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Behavior of Gas Hydrate-Bearing Soils during Dissociation and its Simulation

2014

Hiromasa IWAI
Abstract

Recently, methane hydrates (MHs) have come to represent a new potential energy resource. When the hydrates dissociate, the bonding effect between soil particles may be lost, and the pore water pressure and pore gas pressure may increase. These changes in effective stress and the degradation of the strength of sediments can accelerate the consolidation and shear deformation of the ground and, in turn, produce geological disasters. Thus, characterizing the mechanical properties and responses of hydrate-bearing sediments during dissociation is an important challenge faced by experts in the field of safe natural gas production.

A series of formation and dissociation tests on CO$_2$-hydrates is performed using a temperature-controlled, high-pressure triaxial apparatus in order to characterize the mechanical behavior of gas hydrate sediments subjected to hydrate dissociation. The dissociation tests are conducted using a thermal recovery method under undrained conditions by assuming that the local permeability of the sediments is quite low. It is found that the hydrate dissociation causes a considerable increase in pore pressure and results in significant decreases in effective confining pressure with extensive axial strain. In addition, the experimental results are simulated by a chemo-thermo-mechanically coupled finite element analysis. The simulation results are consistent with the experimental results.

Then, a linear stability analysis is performed in order to investigate the effect of the parameters on the onset of the instability of MH-bearing sediments induced by dissociation. The parameters which have a significant influence on the material instability are the viscoplastic hardening-softening parameter, its gradient with respect to hydrate saturation, the permeability of water and gas, the strain, and the gradient of the dissociation rate with respect to the moles of MHs. With the expansion of strain, material instability may occur even if it is in the region of viscoplastic hardening. In order to investigate the effect of the permeability on the dissociation-deformation behavior, a numerical analysis of the dissociation process in MH-bearing sediments is performed with a one-dimensional finite element mesh. The depressurization method is adopted for the dissociation method. It is found that basically the simulation results become more stable with increases in permeability; however, large permeability enhances hydrate dissociation and causes more gas to be produced, because the depressurized area spreads due to the large permeability.
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# Table of Contents

Chapter 1  INTRODUCTION ......................................................... 1

1.1 Background and Objectives .................................................. 1
  1.1.1 Overview and history of gas hydrates ................................ 1
  1.1.2 Gas hydrates as potential energy resource ............................ 3
  1.1.3 Role of gas hydrates in submarine slope instability ............... 4
  1.1.4 Experimental studies on mechanical properties of gas hydrate-bearing soil .... 6
  1.1.5 Numerical and analytical studies on gas hydrate-bearing sediments .......... 8
  1.1.6 Summary ................................................................. 9

1.2 Scope and Organization ....................................................... 10

Chapter 2  ELASTO-VISCOPLASTIC CONSTITUTIVE MODEL FOR GAS HYDRATE-CONTAINING SOIL AND A CHEMO-THERMO-MECHANICALLY COUPLED ANALYSIS METHOD .... 13

2.1 Introduction ........................................................................... 13

2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil ...... 15
  2.2.1 General setting ............................................................ 15
  2.2.2 Skeleton stress ............................................................ 17
  2.2.3 Elastic stretching tensor .................................................. 19
  2.2.4 Overconsolidation boundary surface .................................. 19
  2.2.5 Static yield function ..................................................... 23
  2.2.6 Viscoplastic potential function ......................................... 23
  2.2.7 Viscoplastic flow rule .................................................... 26
  2.2.8 Temperature-dependency of the viscoplastic parameters .............. 27

2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments .................................. 29
  2.3.1 Equilibrium equation ..................................................... 30
    2.3.1.1 Conservation of momentum for the boundary value problem .... 33
    2.3.1.2 Weak form of the rate type of the equilibrium equations ... 37
    2.3.1.3 Tangent modulus method ......................................... 46
    2.3.2 Conservation of mass .................................................. 50
2.3.2.1 Continuity equation for water and gas ........................................ 50
2.3.2.2 Weak form of the continuity equation for the water phase .......... 53
2.3.2.3 Weak form of the continuity equation for the gas phase ............ 58
2.3.3 Conservation of energy .................................................................. 63
2.3.3.1 Weak form of the conservation of energy ................................. 64
2.3.4 Dissociation rate of methane hydrates .......................................... 69
2.3.5 Effect of the hydrate saturation on permeability ............................. 71
2.3.6 Soil-water characteristic curve ...................................................... 72

2.4 Summary ......................................................................................... 73

Chapter 3  FORMATION AND DISSOCIATION TESTS ON SAND SPECIMENS CONTAINING CARBON DIOXIDE HYDRATES UNDER UNDRAINED CONDITIONS ...................................................... 75

3.1 Introduction ..................................................................................... 75

3.2 Testing Apparatus ............................................................................ 77

3.3 Testing Procedures ........................................................................... 81
3.3.1 Sample preparation ...................................................................... 81
3.3.2 Formation process of carbon dioxide hydrates ............................ 88
3.3.3 Water saturation and consolidation process ................................. 89
3.3.4 Dissociation process .................................................................... 90
3.3.5 Determination of hydrate saturation ............................................ 90
3.3.5.1 Method (A): Water content method ....................................... 91
3.3.5.2 Method (B): Collected gas method ....................................... 92

3.4 Testing Conditions ........................................................................... 94

3.5 Experimental Results ....................................................................... 96
3.5.1 Results of formation process ..................................................... 96
3.5.2 Results of dissociation process ................................................... 101

3.6 Summary ......................................................................................... 112

Chapter 4  NUMERICAL SIMULATION OF DISSOCIATION TESTS BY A CHEMO- THERMO-MECHANICALLY COUPLED METHOD .................................. 115

4.1 Introduction ..................................................................................... 115

4.2 3-Dimensional Finite Element Analysis of Dissociation Tests for Carbon Dioxide Hydrate-Containing Sand Specimen .................................................. 116
Chapter 4

4.2.1 Finite element mesh and boundary conditions ........................................ 116
4.2.2 Simulation cases and material parameters ............................................. 118

4.3 Simulation Results ....................................................................................... 121

4.4 Summary ....................................................................................................... 133

Chapter 5

INSTABILITY ANALYSIS AND NUMERICAL SIMULATION OF
DISSOCIATION PROCESS OF METHANE HYDRATE-BEARING SOIL · 135

5.1 Introduction ................................................................................................... 135

5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials ····· 136
  5.2.1 Governing equations .............................................................................. 136
  5.2.1.1 Stress variables .................................................................................. 136
  5.2.1.2 Conservation of mass ........................................................................ 137
  5.2.1.3 Darcy type of law ............................................................................. 139
  5.2.1.4 Equation of equilibrium ..................................................................... 140
  5.2.1.5 Conservation of energy ...................................................................... 140
  5.2.1.6 Dissociation rate of methane hydrates .............................................. 141
  5.2.1.7 Simplified viscoplastic constitutive equation .................................... 142
  5.2.2 Perturbed governing equations ............................................................. 142
  5.2.3 Conditions of onset of material instability ............................................ 153
    5.2.3.1 Sign of the coefficients \( a_5 \) and \( a_0 \) ........................................... 154
    5.2.3.2 Sign of the coefficients \( a_4 \) and \( a_1 \) .............................................. 157
    5.2.3.3 For the large value of the wave number \( q \) .................................... 158
    5.2.3.4 For the small value of the wave number \( q \) .................................... 162

5.3 Numerical Simulation of Instability Analysis by an Elasto-viscoplastic Model · 162
  5.3.1 One-dimensional model and boundary conditions ............................... 163

5.4 Numerical Results ....................................................................................... 167
  5.4.1 Results of the MH dissociation-deformation problem ........................... 167
  5.4.2 Results of variations in the coefficients ............................................... 176

5.5 Summary ....................................................................................................... 190

Chapter 6 ............................................................................................................ 193

6.1 Concluding Remarks ................................................................................... 193

6.2 Recommendations for Future Work ........................................................... 195

References ....................................................................................................... 197
List of Tables, Figures, and Photos

Table 3.1 Material properties of Toyoura sand (Shiraishi, 2013)
Table 3.2 Test conditions for initial state and formation process
Table 3.3 Test conditions for dissociation process
Table 3.4 B-value and collected gas volume after the dissociation tests
Table 3.5 Maximum values of pore pressure and excess pore pressure, and the pore pressure at final state of dissociation process
Table 4.1 Material parameters for the initial conditions
Table 4.2 Material parameters for the constitutive equations
Table 4.3 Material parameters for the thermal properties
Table 4.4 Material parameters for the soil-water characteristic curve
Table 5.1 Initial conditions of the soil material
Table 5.2 Material parameters for the constitutive equation
Table 5.3 Simulation conditions

Figure 1.1 Methane hydrate (MH21 Research Consortium)
Figure 1.2 Map of discovered gas-hydrate deposits (Makogon et al., 2007)
Figure 1.3 Schematic view of possible hazards in marine sediments induced by gas hydrates dissociation
Figure 1.4 Illustrative view of stable and unstable regions of methane hydrate bearing sediments with and without dissociation
Figure 2.1 Strength-increasing ratio with hydrate saturation
Figure 2.2 OC boundary surface, static yield function, and potential function in the NC region
Figure 2.3 OC boundary surface, static yield function, and potential function in the OC region
Figure 2.4 Isoparametric elements for the soil skeleton, the pore pressures, and temperature
Figure 2.5 Boundary conditions for the whole fluid-solid mixture
Figure 2.6 Boundary conditions for the water phase
Figure 2.7 Boundary conditions for the gas phase
Figure 2.8 Boundary conditions for the temperature
Figure 2.9 Equilibrium curve for methane hydrates (Bejan et al., 2002)
Figure 3.1 Schematic diagram of the triaxial cell and piping systems
Figure 3.2 Grain size distributions of Toyoura sand and samples obtained from Nankai Trough
Figure 3.3 Schematic diagram of the mold and spacer blocks
Figure 3.4 State paths for temperature and pressure on equilibrium curves of carbon dioxide during CO₂ hydrate forming process
Figure 3.5 State paths for temperature and pressure during dissociation process by heating method
Figure 3.6 Schematic diagram of paths used in gas collection process
Figure 3.7-(a)–(d) Time profiles of the pore pressure of each case during formation process (Case-1 ~ Case-4)
Figure 3.7-(e)–(h) Time profiles of the pore pressure of each case during formation process (Case-5 ~ Case-8)
Figure 3.8-(a) Time profiles of the volume injected CO₂ gas, (a) over all view
Figure 3.8-(b) Time profiles of the volume injected CO₂ gas, (b) magnified view
Figure 3.9 Time profiles of the pore pressure and axial strain in Case-7
Figure 3.10 Time profiles of the pore pressure and axial strain in Case-8
Figure 3.11-(a)–(f) Time profiles of pore pressure during dissociation of each case
Figure 3.11-(g)–(h) Time profiles of pore pressure during dissociation of each case
Figure 3.12 Time profiles of the excess pore pressure during dissociation of all cases
Figure 3.13-(a)–(f) Time profiles of effective confining pressure during dissociation of each case
Figure 3.13-(g)–(h) Time profiles of effective confining pressure during dissociation of each case
Figure 3.14 Time profiles of effective confining pressure of all cases
Figure 3.15-(c)–(h) Temperature-pore pressure path during dissociation of each case
Figure 3.16 Temperature-pore pressure paths during dissociation (Case-3 ~ Case-8)
Figure 3.17 Time profiles of the excess pore pressure and the axial strain in Case-7 and Case-8
Figure 4.1 Isoparametric elements used in the three-dimensional finite element method
Figure 4.2 Finite element mesh and boundary conditions for the simulation
Figure 4.3 Conceptual diagram of conditions of the increasing temperature
Figure 4.4-(a), (b) Results of the parametric study on undrained triaxial tests (Iwai, 2010),
Figure 4.5 Soil-water characteristic curve
Figure 4.6 Distributions of the temperature in Case-7, $S_t^H = 15.4 \%$
Figure 4.7 Distributions of the temperature in Case-8, $S_t^H = 32.1 \%$
Figure 4.8 Distributions of CO₂-hydrate remaining ratio in Case-7, $S_r^H = 15.4 \%$
Figure 4.9 Distributions of CO₂-hydrate remaining ratio in Case-8, $S_r^H = 32.1 \%$
Figure 4.10 Distributions of the pore gas pressure in Case-7, $S_p^H = 15.4 \%$
Figure 4.11 Distributions of the pore gas pressure in Case-8, $S_p^H = 32.1 \%$
Figure 4.12 Distributions of the pore water pressure in Case-7, $S_w^H = 15.4 \%$
Figure 4.13 Distributions of the pore water pressure in Case-8, $S_w^H = 32.1 \%$
Figure 4.14 Distributions of the suction in Case-7, $S_r^\mu = 15.4\%$
Figure 4.15 Distributions of the suction in Case-8, $S_r^\mu = 32.1\%$
Figure 4.16 Distributions of the mean effective stress in Case-7, $S_r^\mu = 15.4\%$
Figure 4.17 Distributions of the mean effective stress in Case-8, $S_r^\mu = 32.1\%$
Figure 4.18 Distributions of the viscoplastic volumetric strain in Case-7, $S_r^\mu = 15.4\%$
Figure 4.19 Distributions of the viscoplastic volumetric strain in Case-8, $S_r^\mu = 32.1\%$
Figure 4.20 Distributions of the accumulated viscoplastic deviator strain $\dot{\gamma}^p$ in Case-7, $S_r^\mu = 15.4\%$
Figure 4.21 Distributions of the accumulated viscoplastic deviator strain $\dot{\gamma}^p$ in Case-8, $S_r^\mu = 32.1\%$
Figure 4.22 Skeleton stress path
Figure 4.23 Time profiles of the pore pressure in experiments and the average pore pressure in simulations
Figure 4.24 Time profiles of the effective confining pressure in the experiments and the mean skeleton stress in the simulations
Figure 4.25 Time profiles of the axial strain $\varepsilon_a\%$ in the experiments and the simulations
Figure 5.1-(a), (b) Variation in parameters $C_N$ and $\hat{N}_H$ with respect to $N_H / N_{H0}$
Figure 5.2 Schematic view of the target area of MH-bearing sediments for the numerical simulations
Figure 5.3 Simulation model and boundary conditions
Figure 5.4 Conditions of change in pore pressure change at the depressurization source
Figure 5.5 Stable and unstable regions of permeability and the depressurization level during the MH dissociation process
Figure 5.6 Time profiles of the pore gas pressure $P^G$ [MPa]
Figure 5.7 Time profiles of the remaining MH ration $100 N_H / N_{H0} \%$
Figure 5.8 Time profiles of the average pore pressure $P^e$ [MPa]
Figure 5.9 Time profiles of the mean skeleton stress $\sigma_m^e$ [MPa]
Figure 5.10 Time profiles of the volumetric strain $\varepsilon_v \%$
Figure 5.11-(a)–(f) Time profiles of the coefficients in Case-4-30 with various wave number $q$
Figure 5.12-(a)–(f) Time profiles of the coefficients in Case-4-40 with various wave number $q$
Figure 5.13-(a)–(f) Time profiles of the coefficients in Case-7-30 with various wave number $q$
Figure 5.14-(a)–(f) Time profiles of the coefficients in Case-7-40 with various wave number $q$

Photo 3.1 Exterior appearance of temperature-controlled high-pressure triaxial apparatus
Photo 3.2 Mixture of Toyoura sand and water with a certain water content
Photo 3.3 Instruments for preparing sand specimen
Photo 3.4 Mold for freezing the moist sand and spacer blocks
Photo 3.5 Mold packed in a plastic bag
Photo 3.6 Hydraulic jack
Photo 3.7 Chloroprene-type membrane and silicon-type membrane
Photo 3.8 Inner cell and the connected lines after setting up the specimen
Chapter 1

INTRODUCTION

1.1 Background and Objectives

1.1.1 Overview and history of gas hydrates

Gas hydrates are crystalline compounds which consist of a guest gas molecules trapped inside a host water lattice structures. The gas interacts with the water under the conditions of low temperature and high pressure to form ice-like structure. A unit volume of methane hydrates dissociates into approximately 160 – 170 times the volume of methane gas (at 0°C and 1 atmosphere), although the exact amount varies depending on the measuring environment. Figure 1.1 shows burning artificial methane hydrates. Gas hydrates, commonly methane hydrates (MHs) or carbon dioxide hydrates (CO₂-hydrates), are hydrates with methane gas or carbon dioxide as the guest molecules. They have been regarded as important materials from the viewpoints of a new energy resource and changes in global climate induced by greenhouse gases.

Gas hydrates were recognized in the 1930s when hydrate formations were discovered to be the cause of pipeline blockage during the transmission of natural gas (Hammerschmidt, 1934). Research was then started on the chemical and physical properties and the stability conditions of gas hydrates. In their study, Sloan & Koh (Sloan Jr & Koh, 2007) included references to the first
observation of naturally occurring hydrates found in permafrost regions in 1965 and to the discovery of natural gas hydrate deposits, that is, methane hydrate deposits, within the permafrost regions of the Soviet Union around 1966 (Makogon et al., 1997). In the 1970s, naturally occurring gas hydrates were exploited in permafrost regions, such as the Siberian Messoyakha gas field, the North American Arctic (Kvenvolden & Grantz, 1990), and the Mackenzie Delta in Canada (Judge, 1982). Then, geophysicists and geochemists recognized that natural gas hydrates also occur in shallow sediment under the deep waters of the oceanic outer continental margins (Kvenvolden, 1988). In February 1998, a drilling test for gas hydrate was conducted in the Mackenzie Delta of Canada by the Geological Survey of Canada, the Japan National Oil Company, and the US Geological Survey.

Figure 1.1 Methane hydrate (MH21 Research Consortium)

Figure 1.2 Map of discovered gas-hydrate deposits (Makogon et al., 2007)
On the other hand, the gas hydrates in continental margin sediment have been inferred mainly from the widespread occurrence of a seismic reflector which coincides with the predicted transition boundary at the base of the gas hydrate zone. This reflector is commonly called a BSR (Bottom Simulating Reflector), because it approximately parallels the topography of the seafloor (Kvenvolden, 1988). Figure 1.2 shows a map of the discovered natural gas-hydrate deposits drawn by Makogon et al. (2007). More than 220 gas-hydrate deposits have been found in the world. In Japan, especially in the Nankai Trough area, a huge amount of methane hydrates is believed to exist due to the evidences of the BSR (Satoh et al., 1996).

1.1.2 Gas hydrates as potential energy resource

Research on the mechanical and chemical properties of gas hydrates and the physical properties of methane hydrate-bearing sediments has increased due to the need to develop technologies for extracting natural gas from MH reservoirs and the increase in the discovery of MH deposits all over the world.

In Canada, for example, the first series of international gas production tests from an MH reservoir was conducted at the Mallik site of the Mackenzie Delta in 2002 (Dallimore & Collett, 2005). In the in situ production tests, the thermal recovery method was adopted to extract methane gas, and about 530 m³ of methane gas were produced under standard conditions during a five-day period. The second series of gas production tests was conducted at the Mallik site in 2007 and 2008. The depressurization method was used instead of the thermal recovery method. In the 2008 tests, it was observed that gas and water were produced continuously from the reservoir throughout the test period. The gas production rate ranged from 2,000 to 3,000 m³/day, while the water production rate was 10 to 20 m³/day and the pressure at the bottom hole was rather stable. The total levels of gas and water production throughout the test period were estimated to be about 13,000 m³ and 70 m³, respectively (Kurihara et al., 2011a). These two series of tests in Canada, i.e., 2002 and 2007/2008, indicated that the depressurization method was more efficient than the thermal recovery method from the viewpoint of stable, continuous gas production.

Japan is one of the pioneers in research and development technologies for extracting gas hydrates, because it seems that a huge amount of methane hydrates exists in the deep seabed around Japan, especially in the Nankai Trough. In 2001, the Japanese Ministry of International Trade and Industry (MITI, currently the Ministry of Economy Trade and Industry: METI) presented a strategy for developing naturally occurring gas hydrate deposits in marine sediments around Japan into an energy resource. It was called “Japan’s Methane Hydrate R&D Program”.
Responding to this program, the “Research Consortium for Methane Hydrate Resources in Japan” (known as MH21), an industry-government-academia collaboration research group, was established in 2001 to undertake and to accomplish the challenges announced by "Japan's Methane Hydrate R&D Program". The final goal of the program is to establish a technology platform for commercial gas production from offshore-Japan methane hydrates by 2018 (MH21, 2008).

Japan Oil, Gas, and Metals National Corporation (JOGMEC) conducted the first offshore methane hydrate production trial in March 2013, in the eastern Nankai Trough, Japan. Field work for this trial began in early 2012. Coring and logging operations were conducted to prepare for the gas hydrate production trial. On March 12th, 2013, JOGMEC succeeded in extracting methane gas from the hydrate-bearing sediments by the depressurization method, which was similar to the method used in the production tests at the Mallik site, and approximately 120,000 m$^3$ of methane gas were produced in total during the 6 days. That amount is about 9 times that observed at the Mallik site. However, the production trial was finally terminated due to the increase in sand production. This indicates the importance of evaluating the state of the wellbore in order to produce methane gas safely and efficiently (Uchida et al., 2014; Yamamoto, 2014).

### 1.1.3 Role of gas hydrates in submarine slope instability

As to the other aspects of the new potential energy source, gas hydrates are believed to play a role in the failure of submarine sediments. Kvenvolden (1994, 1993) presented several examples of a possible connection between gas hydrate boundaries and submarine slides, and slump surfaces were recognized. Many of these slides are on gentle slopes which should be stable. Other authors have also reported that gas hydrates, mostly methane hydrates, might affect submarine slides due to dissociation (e.g., Cherkis et al., 1999; Embley, 1980; Nisbet & Piper, 1998; Rothwell et al., 1998; Summerhayes et al., 1979).

Submarine landslides can be caused by an increase in applied shear stress or a reduction in the strength of the soil. When gas hydrates form in sediments, the pore spaces are occupied by the solid phase of gas hydrates, although the gas hydrates themselves can act as a cementation (bonding) agent between soil particles. Then, the permeability of the sediments decreases as more gas hydrates form. The lowering of the sea level, induced by the buildup of continental icecaps, reduces the hydrostatic pressure of the hydrate reservoir and leads to the dissociation of the gas hydrates. The solid phase of the gas hydrates is lost and the hydrates change into the fluid phase, i.e., water and gas. When this released fluid pressure is trapped inside an area of
1.1 Background and Objectives

Sultan et al. (2004a) have analyzed different slope failure events in the Costa target areas, which reflect diverse triggering mechanisms. They indicated some possibilities for the triggering of submarine slides: earthquakes, rapid sediment accumulation, and hydrate dissociation, and they concluded that excess pore pressure is a key parameter to the assessment of slope stability. In order to study the effect of methane hydrate dissociation on slope stability, Sultan et al. (2004b) conducted a theoretical study on the thermodynamic-chemical equilibrium of gas hydrates in sediments, taking into account the influence of changes in temperature, pore pressure, water chemistry, and pore size distribution in the sediments. The model they developed showed that due to increases in temperature, the hydrates began to dissociate first at the top of the hydrate zone, to ensure a chemical equilibrium with the surrounding bulk water, and not at the bottom of the hydrate zone as is often suggested. They applied the numerical model, presented in their paper, to the case of the giant Storegga Slide on the Norwegian margin to investigate the influence of the changes in sea level and the increase in sea water temperature during the last glaciation. They concluded that the failure interface initiated at the top of the hydrate layer, not at the bottom of the hydrate stability zone, due to gas solubility.

The submarine landslides may lead to even worse situation, for example, tsunami disaster. **Figure 1.3** shows a schematic view of possible hazards in marine sediments induced by gas hydrates dissociation. Slope failure in marine sediments can cause enormous turbidity current, and it may generate tsunami. The tsunami produced by slope failure will hit coastal area and offshore structures, and in the worst scenario, many people might be killed by tsunami. In fact,
the landslides occurred off the French coast in 1979. A 0.15-km$^3$ slide off Nice airport that killed 11 people (Nisbet & Piper, 1998). Although this submarine landslide was not caused by hydrate dissociation, this report pointed out that landslides can be a trigger of tsunami disaster.

These examples of gas hydrate dissociation triggering submarine landslides were caused by natural processes, such as changes in sea level. Similar phenomena may occur when producing methane gas from methane hydrate-bearing sediments as a new energy resource. The point is that hydrate dissociation, which can lead to submarine slides, can be artificially induced. Thus, it is an important challenge to characterize the geo-mechanical behavior of gas hydrate-bearing sediments, such as changes in stress and deformation induced by dissociation.

### 1.1.4 Experimental studies on mechanical properties of gas hydrate-bearing soil

Understanding the geo-mechanical properties of these types of sediments, particularly shear strength or deformation, is required for a stability analysis under different environmental conditions of the deep sea. It is difficult, however, to obtain natural hydrate-bearing samples, because many complications are encountered when transferring the preserved samples to the testing apparatus in the laboratory. Even when using pressure cores, the in situ pressure must be released momentarily in order to transfer the core into the triaxial apparatus. Only a few strength tests have been performed on samples containing natural hydrates recovered from drilled wells (Yoneda et al., 2010). Thus, different laboratory methods have been developed to form methane hydrates in reconstituted sediments.

The most time-consuming and laborious method for creating hydrates in a laboratory is the “dissolved gas method”, in which the hydrates are formed from dissolved gas in the absence of free gas (Waite et al., 2009). This method is thought to most closely simulate the natural formation process in both arctic and deep coarse-grained seabed sediments. Buffett & Zatsepina (2000) succeeded in creating CO$_2$-hydrates in porous media using an aqueous solution with dissolved carbon dioxide. They were the first experiments to show that hydrates would form in porous media under realistic conditions even in the absence of free gas. Spangenberg et al. (2005) was the first to report the formation of methane hydrates in a sample of packed glass beads with a dissolved methane gas solution. The methane hydrates saturated up to 95% of the pore spaces with hydrates formed from the dissolved-phase methane over a period of 55 days by pumping methane-saturated water via a gas/water chamber into the sample. After 55 days, they observed a blockage in the flow due to the MH formation.
Another hydrate-formation method, referred to as the “partial water saturation method”, can reduce the formation time substantially by flushing the pressurized methane gas through partially saturated sediment and cooling it in the stability region (e.g., Hyodo et al., 2013a; Miyazaki et al., 2011a; Priest & Best, 2005; Waite et al., 2004; Yoneda et al., 2013a, 2010). The point here is that the formation behavior of the method is quite different from the natural process, although it is easier to form MHs. Each of the methods for forming hydrates produces different methane growth patterns, resulting in different types of macro-scale behavior of seemingly identical sediments. Particularly affected are the mechanical properties of the sediments (Ghiassian & Grozic, 2013; Waite et al., 2009).

Yoneda et al. (2010) performed a series of triaxial compression tests under drained conditions in order to investigate the strength of MH-bearing sand specimens, which were formatted to simulate MH-bearing sediments in the Nankai Trough, with various densities for the host sand, the saturation of the MHs, the confining pressure, the temperature, and the pore pressure. They indicated that the peak strength of the MH-bearing sand increased depending on the density of the sand and the confining pressure. Moreover, the peak strength increased with the increase in saturation of the MHs, the decrease in temperature, and the increase in pore pressure. Miyazaki et al. (2008) conducted drained triaxial tests on artificial MH-bearing sand specimens at various strain rates to investigate the effects of the strain rate on the shear strength. As a result, the peak strength at faster strain rates became higher than that at slower strain rates. It was also found that the strain-rate dependency of the peak strength increased with an increase in methane hydrate saturation. It can be said that the methane hydrate saturation significantly affected the strain-rate dependency of the methane hydrate sediments. Miyazaki et al. (2010) then investigated the effects of the confining pressure on the mechanical properties of artificial methane hydrate sediments. The drained shear strength increased with an increase in the effective confining pressure and hydrate saturation, and the peak strength can be written as a function of the confining pressure and hydrate saturation. They also found that it is possible to obtain the shear strength with different confining pressure levels from only one sand specimen. Ghiassian & Grozic (2013) studied the undrained shear strength of methane hydrate-bearing sand. They used Ottawa sand as the host material and formed the MHs with the partial water saturated method. The strength results indicated that the presence of gas hydrates will enhance the undrained shear strength of the sediments and the stiffness. Through an investigation of the undrained shear strength of laboratory-formed specimens, containing hydrates and other pore fillings, Winters et al. (2007) noted an increase in strength for the hydrate specimens, which was directly related to the hydrate saturation.

These experimental studies mainly focused on the strength of MH-bearing sediments, whereas little information can be found on the mechanical behavior of the seabed ground imposed by
MH dissociation. In the present study, a series of formation and dissociation tests on sand specimens containing carbon dioxide hydrates is performed. The main purpose of these experiments is to investigate the mechanical response, such as changes in stress or the deformation of the gas hydrate-bearing soil induced by dissociation. In the experiments, a temperature-controlled, high pressure triaxial apparatus is used. This specialized apparatus has been modified so that it is possible to create the conditions of low temperature and high pressure based on the conventional triaxial apparatus for soft rocks and hard clays.

### 1.1.5 Numerical and analytical studies on gas hydrate-bearing sediments

There are currently several numerical models that can simulate the flow behavior during dissociation (e.g., Kimoto et al., 2010, 2007a; Kurihara et al., 2011; Moridis et al., 2009; Sakamoto et al., 2009; White and McGrail, 2008; Yoneda, 2013b, etc). Changes in mass, the flows of water, gas, and heat, and the changes in the material properties, such as the permeability of fluids and thermal conductivity, are considered, while solid phases, that is, hydrates and soils, are assumed to be immobile or treated as elastic materials in most simulators. Kimoto et al. (2010, 2007a) have developed a numerical simulator for the deformation of soil containing methane hydrates in order to predict the stability of the ground during hydrate dissociation, in which an elasto-viscoplastic model, considering the effects of suction and hydrate saturation, is used for the constitutive model of the soil sediments. From the numerical results, they reported that ground deformation is induced by the generation and the dissipation of water pressure and gas pressure, and that it localizes around the hydrate dissociation area.

Many experimental and numerical studies have been conducted on the deformation behavior associated with methane hydrate dissociation. Nevertheless, the behavior of methane hydrate-bearing soils during dissociation is still a subject of research, and theoretical analyses to investigate the onset of instability, such as linear stability analyses, have not been performed. Instability analyses for water-saturated soil have been widely performed by many researchers. Rice (1975) investigated the stability of fluid-saturated porous material in quasi-static conditions. Anand et al. (1987), and Zbib and Aifantis (1988) conducted linear perturbation stability analyses for the onset of shear localization. Loret and Harireche (1991) investigated the acceleration waves in inelastic porous media, and Benallal and Comi (2002) showed material instabilities in saturated material under the dynamic state using a perturbation stability analysis. Oka et al. (1995) have been dealing with the strain localization problem of water-saturated clay through the use of viscoplastic constitutive equations because of the rate-dependent nature of cohesive soil. Higo et al. (2005) have studied the effect of permeability and initial heterogeneity
on the strain localization of water-saturated soil. Kimoto et al. (2007b) have performed a linear instability analysis on the thermo-hydro-mechanical coupled material system, and have indicated that strain softening and temperature softening are the main reasons for the material instability. Recently, Garcia et al. (2010) have performed a linear stability analysis in order to investigate which variables have a significant effect on the onset of the instability of an unsaturated viscoplastic material subjected to water infiltration. They have found that the onset of the growing instability of the material system mainly depends on the specific moisture capacity, the suction and the hardening parameter.

In the present study, we conduct a linear stability analysis to investigate the onset of instability during the dissociation process. Figure 1.4 shows an illustration of the stable and unstable regions of methane hydrate-bearing sediments with and without hydrates dissociation.

We discuss which parameters or variables have a significant effect on the instability of methane hydrate-bearing materials when they are subjected to a dissociation process. In the linear stability analysis, we extend the method by Oka et al. (1995), and Garcia et al. (2010) to the chemo-thermo-mechanically coupled material considering hydrate dissociation. Then, we conduct a numerical analysis of gas hydrate dissociation with a viscoplastic model in order to investigate the effect of the parameters related to the hydrate dissociation rate on the system and compare it with the results of an instability analysis.

![Figure 1.4 Illustrative view of stable and unstable regions of methane hydrate bearing sediments with and without dissociation](image)

**1.1.6 Summary**

Gas hydrates play an important role from the viewpoints of a new energy resource and a
1.2 Scope and Organization

Many experimental studies and numerical analyses have been performed to investigate the mechanical properties of MH-bearing sediments. There is little information, however, on the behavior of sediments subjected to hydrate dissociation or on the onset of the instability of the seabed ground. The main objectives of the present study are summarized as follows.

1. to understand the changes in stress and the deformation behavior of CO$_2$-hydrates through dissociation tests under undrained conditions using a temperature-controlled, high-pressure triaxial apparatus;

2. to find the parameters which have a significant effect on the onset of instability during the dissociation process, through a linear stability analysis and chemo-thermo-mechanically coupled finite element simulations.

1.2 Scope and Organization

The present research is roughly divided into three parts. The first part is a formulation of the constitutive equation and the governing equations for gas hydrate-bearing sediments. The second part contains the experimental works using a temperature-controlled, high-pressure triaxial apparatus. The third part is an instability analysis of methane hydrate-bearing sediments. An outline for each chapter is described below.

Chapter 1: The current chapter presents the main purpose of this doctoral research, along with a discussion and background information on gas hydrate-bearing sediments.

Chapter 2: An elasto-viscoplastic constitutive model for gas hydrate-bearing soils is derived based on the model proposed by Adachi and Oka (1982), and governing equations for gas hydrate-bearing sediments are introduced for a chemo-thermo-mechanically coupled analysis as well as discretization methods for a finite element analysis.

Chapter 3: The behavior of sand specimens containing carbon dioxide hydrates is studied during the dissociation process using a temperature-controlled, high-pressure triaxial apparatus. In this chapter, the specialized triaxial system is introduced. It can create the conditions of the deep seabed ground, that is, low temperature and high pressure. After that, the mechanical behavior, such as the changes in stress and deformation during dissociation under undrained conditions, is investigated.
Chapter 4 presents a numerical simulation of dissociation tests on sand specimens containing CO$_2$-hydrates, shown in Chapter 3. The simulation is conducted by the chemo-thermo-mechanically coupled finite element method proposed by Kimoto et al. (2007b). The distributions of pore water pressure, pore gas pressure, volumetric strain and so on, are obtained from the results and compared with the experimental results.

Chapter 5 deals with a linear instability analysis of the dissociation process of methane hydrate-bearing sediments in order to investigate which parameters have a significant effect on the onset of instability, such as strain localization and uncontrollable gas production. In addition, some examples of the numerical simulation of gas hydrate dissociation with a one-dimensional model are shown in order to investigate the effect of the parameters as well as the results discussed in the stability analysis.

Chapter 6 presents the concluding remarks and a summary of this research with suggestions for future works.
1.2 Scope and Organization
Chapter 2

ELASTO-VISCOPLASTIC CONSTITUTIVE MODEL FOR GAS HYDRATE-CONTAINING SOIL AND A CHEMO-THERMO-MECHANICALLY COUPLED ANALYSIS METHOD

2.1 Introduction

It has been well recognized that MH-bearing sediments display higher strength and stiffness than sediments without MHs. Based on a review of the mechanical properties of MH-bearing soil by Waite et al. (2009), the following characteristics are clarified: (1) the peak strength of MH-bearing soils depends on the hydrate saturation; (2) the hydrate contribution to its shear behavior is of a cohesive nature rather than a frictional one; (3) the dilation angle increases with an increase in the hydrate saturation; and (4) the stiffness of gas hydrate-bearing soil is likely to become larger than that of soil without hydrates. Once the hydrates dissociate, either through a depressurization method or a heating method, the MHs in the pores disappear and the soil may
lose its strength or stiffness. The loss of shear resistance can cause some geo-mechanical issues, such as seabed ground settlement, submarine landslides, and the collapse of the wellbore. In order to predict the behavior of MH-bearing sediments and to point out the risks during MH production, it is important to develop a numerical model which can describe the mechanical response of MH-bearing soil subjected to gas production.

The development of numerical models for gas hydrate-bearing sediments requires not only consideration of the mechanical response, but also the thermal and chemical reactions, because the formation and dissociation of gas hydrates are the reactions that accompany phase changes and heat transfer. Many researches and numerical simulations have been conducted in order to simulate the behavior of sediments subjected to hydrate dissociation.

Hong & Pooladi-Darvish (2005) presented a 2-dimensional cylindrical simulator for gas production from MH reservoirs. The model includes equations for the flows of gas and water, conductive and convective heat transfer, and the kinetics of hydrate decomposition. Bejan et al. (2002) and Tsypkin (2000) also considered the multi-fluid flow and heat transfer in porous media. However, they did not consider ground deformation; namely, the solid phase was assumed to be immobile.

From the viewpoint of the mutual relationship between hydrate dissociation and geo-mechanics, Rutqvist & Moridis (2008) and Rutqvist et al. (2009) reported a numerical simulator for analyzing the geo-mechanical performance of hydrate-bearing permafrost. They combined the hydraulic behavior and the mechanical behavior. Uchida et al. (2012) developed a multi-phase coupled numerical model. They considered not only the multi-fluid flow and the heat transfer, but also took into account the bonding effect of MHs on the strength and stiffness in a constitutive model based on the conventional elastoplastic constitutive model. They concluded that volumetric yielding plays an important role in the accurate prediction of ground settlement when MH-bearing soil undergoes a significant change in effective stress and hydrate saturation. Klar et al. (2013) and Zhou et al. (2014) conducted a dissociation-deformation coupled numerical analysis, using the models proposed by Uchida et al. (2012), in order to predict the geo-mechanical behavior of the MH-bearing sediments subjected to the hydrate dissociation.

Kimoto et al. (2010, 2007a) introduced the strength enhancement of MH-bearing soil into the elasto-viscoplastic model, and conducted numerical simulations to investigate the ground deformation during the dissociation process. In the simulations, both the depressurization method and the thermal recovery method were considered. The results indicated that ground deformation was induced by the generation and dissipation of water and gas, and by the
reduction in soil strength due to the loss of hydrates. However, neither the deformation that localized around the dissociated area nor the amount of settlement was so large in the simulation for which the dissociated area was limited around the source, and the hydrate-bearing sediments were distributed uniformly in the horizontal layer. According to Miyazaki et al. (2008), the methane hydrate saturation significantly affected to the strain-rate dependency and then failure mechanism of the methane hydrate sediments. The model proposed by Kimoto et al. (2010, 2007a) is advantageous in that it is able to describe the time-dependent behavior with the viscoplastic constitutive equations. In this chapter, the elasto-viscoplastic model, considering the bonding effect of MHs, and the multiphase coupled analysis method, proposed by Kimoto et al. (2010, 2007a), are introduced.

The following assumptions are adopted in the formulation:

1) Soil particles and water are incompressible.

2) A guest gas, generally methane gas or carbon dioxide, is treated as an ideal gas and the effect of dissolution into water is disregarded.

3) The flows of gas and water are independent and they follow Darcy’s law.

4) The soil and the hydrates behave as a solid mass, so that the velocity of the hydrates is equal to that of the soil.

5) The acceleration term is disregarded in the equation of motion.

### 2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

#### 2.2.1 General setting

Multiphase material $\psi$ is composed of four phases, namely, soil ($S$), water ($W$), gas ($G$), and hydrates ($H$), which are continuously distributed throughout space.
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

\[ \psi = \sum_{\alpha} \psi^\alpha \quad (\alpha = S, W, G, H) \]  \hspace{1cm} (2.1)

in which \( S \) is the soil phase, \( W \) is the water phase, \( G \) is the gas phase, and \( H \) is the hydrate phase. For simplicity, we assume that the hydrates move with the soil particles. In other words, the solid phase, denoted by \( SH \), is composed of soil and hydrates which exist around the soil particles. Total volume \( V \) is obtained from the sum of the partial volumes of the constituents, namely,

\[ \sum_{\alpha} V^\alpha = V \quad (\alpha = S, W, G, H) \]  \hspace{1cm} (2.2)

Total volume of fluids \( V^F \) is given by

\[ V^F = \sum_{\gamma} V^\gamma \quad (\gamma = W, G) \]  \hspace{1cm} (2.3)

Volume fraction \( n^\alpha \) is defined as the local ratio of the volume element with respect to the total volume as:

\[ n^\alpha = \frac{V^\alpha}{V}, \quad \sum_{\alpha} n^\alpha = 1 \quad (\alpha = S, W, G, H) \]  \hspace{1cm} (2.4)

The volume fraction of the void, \( n \), is written as:

\[ n = \sum_{\beta} n^\beta = \frac{V - V^S}{V} = 1 - n^S \quad (\beta = W, G, H) \]  \hspace{1cm} (2.5)

The volume fraction of the fluid, \( n^F \), is given by

\[ n^F = n - n^H \]  \hspace{1cm} (2.6)

In addition, the fluid saturation is given by

\[ s^\gamma = \frac{V^\gamma}{V^F}, \quad \sum_{\gamma} s^\gamma = 1 \quad (\gamma = W, G) \]  \hspace{1cm} (2.7)
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

Water saturation $s^W$ is denoted by water saturation $s$ in the following.

$$s = \frac{V^W}{V^f} = \frac{n^W}{n^f}$$  \hfill (2.8)

The density of each material, $\rho^a$, and the total phase $\rho$ are denoted by

$$\rho = \sum_n n^a \rho^a \hspace{1cm} \alpha = S,W,G,H$$  \hfill (2.9)

### 2.2.2 Skeleton stress

In the theory of porous media, the concept of the effective stress tensor is related to the deformation of the soil skeleton and plays an important role. The effective stress tensor has been defined for water-saturated soil (Terzaghi, 1943). In the case of unsaturated soils, however, the concept needs to be redefined in order to consider compressible materials. In the present study, skeleton stress $\sigma'_{ij}$ is defined and then used for the stress variable in the constitutive relation for the soil skeleton as well as for suction. Total stress tensor $\sigma_{ij}$ is obtained from the sum of the partial stresses, $\sigma^{a}_{ij}$, namely,

$$\sigma_{ij} = \sum_{\alpha} \sigma^{a}_{ij} \hspace{1cm} \alpha = S,W,G,H$$  \hfill (2.10)

in which $\sigma^{a}_{ij}$ represents the stress acting on each phase. The partial stresses for the water and gas phases are expressed as:

$$\sigma^W_{ij} = -n^W \rho^W \delta_{ij}$$  \hfill (2.11)

$$\sigma^G_{ij} = -n^G \rho^G \delta_{ij}$$  \hfill (2.12)
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

where $P^W$ and $P^W$ are the pore water pressure and the pore gas pressure, respectively. Tension is positive for the stresses, whereas compression is positive for the pore water pressure and the pore gas pressure.

For simplicity, we assume that the soil phase and the hydrate phase are in the same phase, namely, the solid phase. Thus, the partial stress of the solid phase is defined as

$$\sigma_{ij}^{SH} = \sigma_{ij}^s + \sigma_{ij}^H$$  \hspace{1cm} (2.13)

The partial stress tensor for the solid phase can be expressed by the analogy with the water saturated one as follows:

$$\sigma_{ij}^{SH} = \sigma_{ij}^t - n^{SH} P^F \delta_{ij}$$  \hspace{1cm} (2.14)

$$n^{SH} = n^s + n^H$$  \hspace{1cm} (2.15)

$$P^F = sP^w + (1-s)P^G$$  \hspace{1cm} (2.16)

where $P^F$ is the average pressure of the fluids surrounding the solid skeleton, and $s$ is the degree of water saturation.

The total stress tensor can be made up of these partial stresses by substituting Equations (2.11) ~ (2.16) into Equation (2.10).

$$\sigma_{ij} = \sum_\alpha \sigma_{ij}^\alpha = \sigma_{ij}^t - n^{SH} P^F \delta_{ij} - n^w P^w \delta_{ij} - n^G P^G \delta_{ij} = \sigma_{ij}^t - P^F \delta_{ij}$$  \hspace{1cm} (2.17)

$\sigma_{ij}^t$ is called the skeleton stress in the present study. It acts on the solid phase and is used as the stress variable in the constitutive equation.

$$\sigma_{ij}^t = \sigma_{ij} + P^F \delta_{ij}$$  \hspace{1cm} (2.18)
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

2.2.3 Elastic stretching tensor

It is assumed that the total strain rate tensor, \( \dot{e}_{ij} \), consists of elastic strain rate tensor \( \dot{e}^e_{ij} \) and viscoplastic strain rate tensor \( \dot{e}^{vp}_{ij} \) as:

\[
\dot{e}_{ij} = \dot{e}^e_{ij} + \dot{e}^{vp}_{ij}
\]  
(2.19)

The elastic strain rate tensor is given by a generalized Hooke type of law, namely,

\[
\dot{e}^e_{ij} = \frac{1}{2G} S_{ij} + \frac{\kappa}{3(1 + e)} \dot{\sigma}^m_{ij} \delta_{ij}
\]  
(2.20)

where \( S_{ij} \) is the deviator stress tensor, \( \sigma^m_{ij} \) is the mean skeleton stress, \( G \) is the elastic shear modulus, \( e \) is the void ratio, \( \kappa \) is the swelling index, and the superimposed dot denotes the time differentiation. Deviator stress tensor \( S_{ij} \) and the mean skeleton stress are defined as,

\[
S_{ij} = \sigma_{ij} - \frac{1}{3} \sigma_{kk} \delta_{ij}
\]  
(2.21)

\[
\sigma^m_{ij} = \frac{1}{3} \sigma^m_{kk}
\]  
(2.22)

2.2.4 Overconsolidation boundary surface

In this model, it is assumed that there is an overconsolidation (OC) boundary surface that delineates the overconsolidation (NC) region, \( f_b < 0 \), from the normal consolidation region, \( f_b \geq 0 \), as follows:

\[
f_b = \bar{n}^*_b + M_m^* \ln \frac{\sigma^r_m}{\sigma^r_{mb}} = 0
\]  
(2.23)
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

\[
\bar{\eta}_{ij}^{(0)} = \sqrt{\left(\eta_{ij}^* - \eta_{ij}^{(0)}) \left(\eta_{ij}^* - \eta_{ij}^{(0)}\right)\right)}
\]  

(2.24)

\[
\eta_{ij}^* = \frac{S_{ij}}{\sigma'_{m}}, \quad \eta_{ij}^{(0)} = \frac{S_{ij(0)}}{\sigma'_{m(0)}},
\]  

(2.25)

where \(\eta_{ij}^*\) is the stress ratio tensor, and subscript \((0)\) denotes the state at the end of consolidation, that is, the initial state before deformation occurs. \(M_m^*\) is the value of \(\eta^* = \sqrt{\eta_{ij} \eta_{ij}}\) when the volumetric strain increment changes from contraction to dilation, which is equal to ration \(M_j^*\) at the critical state, and \(\sigma'_{mb}\) is the hardening parameter, that controls the size of the boundary surface. In the present model, the hardening parameter is assumed to be a function of the viscoplastic volumetric strain \(\varepsilon_{kk}^v\), suction \(P^c = P^G - P^w\), and the saturation of hydrate \(S_H^r\). In order to describe the structural degradation on natural clay, strain-softening with viscoplastic strain is introduced into the hardening parameter in addition to strain-hardening with the viscoplastic volumetric strain (Kimoto et al., 2004).

\[
\sigma'_{mb} = N_s N_m \sigma'_{ma}(z) \exp\left(\frac{1 + \varepsilon^w}{\lambda - \kappa}\varepsilon_{kk}^v\right)
\]  

(2.26)

in which \(N_m\) and \(N_s\) denote the effects of hydrate saturation and suction respectively, \(\sigma_{ma}(z)\) refers to structural degradation with increasing viscoplastic strain, \(\lambda\) is the compression index, and \(\kappa\) is the swelling index. The term \(\sigma'_{ma}(z)\) describes strain-softening, which controls the degradation of the material caused by structural changes, namely,

\[
\sigma'_{ma} = \sigma'_{maf} + \left(\sigma'_{mai} - \sigma'_{maf}\right) \exp\left(-\beta z\right)
\]  

(2.27)
### 2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

\[ z = \int_0^t \dot{z} \, dt, \quad \dot{z} = \sqrt{\dot{\varepsilon}_{ij}^{\text{vp}} \dot{\varepsilon}_{ij}^{\text{vp}}} \]  \hspace{1cm} (2.28)

in which \( \sigma_{\text{mai}}' \) and \( \sigma_{\text{maf}}' \) are the initial and the final values of \( \sigma_{\text{ma}}' \), respectively. \( \beta \) is a material parameter which controls the rate of structural change, and \( z \) is the accumulation of the second invariant of the viscoplastic strain rate \( \dot{\varepsilon}_{ij}^{\text{vp}} \). Since the viscoplastic strain is equal to zero at the initial state, the initial value of the hardening parameter \( \sigma_{\text{mbi}}' \) should be equal to \( \sigma_{\text{mai}}' \).

Parameter \( N_s \) is defined as:

\[
N_s = \begin{cases} 
1 + s_f \exp \left( -s_d \left( \frac{P^C}{P_i^C} - 1 \right) \right), \quad (P^C \neq 0) \\
1, \quad (P^C = 0)
\end{cases}
\]  \hspace{1cm} (2.29)

where \( s_f \) is the strength ratio of unsaturated soils when the value of suction \( P^C \) equals to \( P_i^C \), and \( s_d \) controls the decreasing ratio of strength with decreasing suction. The term \( P_i^C \) is set to be the maximum value for suction. At the initial state, when \( P^C = P_i^C \), the strength ratio of the unsaturated soil to the saturated soil is \( 1 + s_f \) and it decreases with a decline in suction.

Parameter \( N_m \) describes the dependency of the hydrate saturation on the stress-strain behavior, and it is defined as:

\[
N_m = \begin{cases} 
1 + n_m \exp \left( -n_d \left( \frac{S_r^H}{S_r^H} - 1 \right) \right), \quad (S_r^H \neq 0) \\
1, \quad (S_r^H = 0)
\end{cases}
\]  \hspace{1cm} (2.30)
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

in which \( S^H_r \) is the hydrate saturation in the void defined as

\[
S^H_r = \frac{V^H}{V^v}, \tag{2.31}
\]

where \( V^v \) is the volume of voids and \( V^H \) is the volume of hydrates in the soil.

In Equation (2.30), \( n_m \) is a material parameter which describes the ratio of the strength when hydrate saturation \( S^H_r \) equals to \( S^H_{ri} \) to that when hydrate saturation \( S^H_r \) equals zero. The term \( n_d \) is the stress-decreasing ratio with decreasing hydrate saturation. The material parameters which describe the effect of hydrate saturation are determined to be \( S^H_{ri} = 0.51 \), \( n_m = 0.6 \), and \( n_d = 0.75 \), based on the experimental data on carbon dioxide hydrate mixtures (Matsui et al., 2005). Figure 2.1 shows the strength-increasing ratio with various values of the parameter \( n_d \).

![Figure 2.1 Strength-increasing ratio with hydrate saturation](image-url)
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

2.2.5 Static yield function

In order to describe the mechanical behavior of clay at its static equilibrium state, Adachi and Oka(1982) assumed a Cam-clay type of static yield function.

\[ f_y = \eta_i^{*(0)} + \tilde{M}^* \ln \frac{\sigma_m}{\sigma_{my}} = 0 \]  
\[(2.32)\]

In the same way as for the OC boundary surface, the effects of suction and hydrate saturation are introduced in the value of \( \sigma_{my}^{*(i)} \).

\[ \sigma_{my}^{*(i)} = N_m N_i \sigma_{ma}^{*(i)} \sigma_{my}^{*(i)} \exp \left( \frac{1 + \varepsilon_{vp}^{*(i)}}{\lambda - \kappa} \right) = \frac{\sigma_{my}^{*(i)}}{\sigma_{ma}^{*(i)}} \sigma_{mb}^{*(i)} \]  
\[(2.33)\]

where \( \sigma_{my}^{*(i)} \) is the initial value of \( \sigma_{my}^{*(i)} \) when \( N_m = N_i = 1 \).

2.2.6 Viscoplastic potential function

The viscoplastic potential function, \( f_p \), is given as

\[ f_p = \eta_i^{*(0)} + \tilde{M}^* \ln \frac{\sigma_m}{\sigma_{mp}} = 0 \]  
\[(2.34)\]

where \( \tilde{M}^* \) is a dilatancy coefficient; it is assumed to be constant in the normally consolidation region and to vary with the current stress state in the overconsolidation region, as defined in Equation (2.35).

\[ \tilde{M}^* = \begin{cases} \frac{M_m^*}{\eta_i^{*(0)}} & : f_k \geq 0 \text{(NC region)} \\ -\frac{\sqrt{\eta_{ij} \eta_{ij}}}{\ln \left( \frac{\sigma_m}{\sigma_{mc}} \right)} & : f_k < 0 \text{(OC region)} \end{cases} \]  
\[(2.35)\]
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

where $M_m^*$ is the value of $\sqrt{\eta_{ij}^* \eta_{ij}^*} / \sigma_m'$ at the critical state, and $\sigma_m'$ denotes the mean skeleton stress at the intersection of the OC boundary surface and the $\sigma_m'$ axis as

$$\sigma_m' = \sigma_{mb}' \exp \left( \frac{\sqrt{\eta_{ij}(0) \eta_{ij}(0)}}{M_m^*} \right)$$

(2.36)

In the case of the isotropic condition, $\sigma_m'$ equals to $\sigma_{mb}'$. Overconsolidation boundary $f_{b}$, static yield function $f_y$, and viscoplastic potential function $f_p$ at $\eta_{ij}(0) = 0$ for the NC and OC regions are illustrated in the $\sigma_m' - \sqrt{S_{ij} S_{ij}}$ space in Figure 2.2 and Figure 2.3, respectively. $\sigma_{mb}'$ and $\sigma_{my}'$ decrease with an increasing accumulation of viscoplastic strain, with decreasing suction, and with decreasing hydrate saturation due to the dissociation. Since the increments in viscoplastic strain for the overstress type of model depend on the difference between the current stress state and the static state, the shrinkage of the static yield function is due to the degradation of the soil structure, the decreasing suction, and the dissociation of the hydrate yield in the viscoplastic strain increments.
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

Figure 2.2 OC boundary surface, static yield function, and potential function in the NC region

Figure 2.3 OC boundary surface, static yield function, and potential function in the OC region
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

2.2.7 Viscoplastic flow rule

The viscoplastic strain rate tensor, $\dot{\varepsilon}_{ij}^p$, is expressed by the following equation which is based on Perzyna's viscoplasticity theory (Perzyna, 1963):

$$\dot{\varepsilon}_{ij}^p = \gamma \left\{ \Phi_1 \left( f_y \right) \right\} \frac{\partial f_p}{\partial \sigma_i^j}$$

(2.37)

$$\left\{ \Phi_1 \left( f_y \right) \right\} = \begin{cases} \Phi_1 \left( f_y \right) & : f_y > 0 \\ 0 & : f_y \leq 0 \end{cases}$$

(2.38)

where $\Phi_1$ indicates the strain-rate sensitivity. Based on the experimental data of the strain-rate constant triaxial tests, the material function $\Phi_1$ is given as

$$\gamma \Phi_1 \left( f_y \right) = C' \sigma_m' \exp \left( m' f_y \right)$$

$$= C' \sigma_m' \exp \left\{ m' \left( \tilde{\eta}^{(0)} \right) + \tilde{M}^* \ln \frac{\sigma_m'}{\sigma_n^{(s)}} \right\}$$

$$= C' \sigma_m' \exp \left\{ m' \left( \tilde{\eta}^{(0)} \right) + \tilde{M}^* \ln \frac{\sigma_m'}{\sigma_n^{(s)}} \frac{\sigma_m'}{\sigma_n^{(s)}} \right\}$$

$$= C' \sigma_m' \exp \left\{ m' \tilde{\eta}^{(0)} \right\} \exp \left\{ m' \tilde{M}^* \ln \frac{\sigma_m'}{\sigma_n^{(s)}} \right\}$$

$$= C \sigma_m' \exp \left\{ m' \tilde{\eta}^{(0)} \right\}$$

$$C = C' \exp \left( m' \tilde{M}^* \ln \frac{\sigma_m'}{\sigma_n^{(s)}} \right)$$

(2.39)

Finally, using the fourth rank isotropic tensor, $C_{ijkl}$, the viscoplastic strain rate tensor is given by the following equations:
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

\[ \dot{\varepsilon}_{ij}^{vp} = \begin{cases} C_{ijkl} \sigma_{m}^{'} \exp \left\{ m'(\vec{\eta}_{(0)}^{*} + \vec{M}^{*} \ln \frac{\sigma_{m}^{'} - \sigma_{mb}}{\sigma_{mb}}) \right\} \frac{\partial p_{f}}{\partial \sigma_{kl}^{*}} & : f_{y} > 0 \\ 0 & : f_{y} \leq 0 \end{cases} \]  

(2.41)

where the viscoplastic parameter \( C_{ijkl} \) is given by

\[ C_{ijkl} = D \delta_{ij} \delta_{kl} + E \left( \delta_{ik} \delta_{jl} + \delta_{il} \delta_{kj} \right) \]  

(2.42)

in which \( D \) and \( E \) are material constants. Substituting Equation (2.34) into Equation (2.41), the viscoplastic deviator strain rate and the viscoplastic volumetric strain rate are obtained as follows:

\[ \dot{\varepsilon}_{ij}^{vp} = C_{i} \sigma_{m}^{'} \exp \left\{ m'(\vec{\eta}_{(0)}^{*} + \vec{M}^{*} \ln \frac{\sigma_{m}^{'} - \sigma_{mb}}{\sigma_{mb}}) \right\} \frac{n_{ij}^{*} - n_{ij(0)}^{*}}{\vec{\eta}^{*}} \]  

(2.43)

\[ \dot{\varepsilon}_{kk}^{vp} = C_{2} \sigma_{m}^{'} \exp \left\{ m'(\vec{\eta}_{(0)}^{*} + \vec{M}^{*} \ln \frac{\sigma_{m}^{'} - \sigma_{mb}}{\sigma_{mb}}) \right\} \left( \vec{M}^{*} - \frac{n_{\mu \nu}^{*} (n_{\mu \nu}^{*} - n_{\mu \nu(0)}^{*})}{\vec{\eta}^{*}} \right) \]  

(2.44)

\[ C_{1} = 2E, \quad C_{2} = 3D + 2E \]  

(2.45)

where \( C_{1} \) and \( C_{2} \) are the viscoplastic parameters for the deviator and the volumetric strain components, respectively.

2.2.8 Temperature-dependency of the viscoplastic parameters

Based on the experimental results by Boudali et al. (1994), Yashima et al. (1998) showed the following relation between consolidation yield stress \( \sigma_{p}^{'} \) and temperature \( \theta \).
2.2 Elasto-viscoplastic Constitutive Equation for Gas Hydrate-Containing Soil

\[
\frac{\sigma'_p}{\sigma'_{pr}} = \left[ \frac{\theta_r}{\theta} \right]^\alpha
\]  

(2.46)

\[
C_i (\theta) = C'_i \exp \left\{ m' \tilde{M}^* \left( -\ln \left[ \frac{\sigma'_p}{\sigma'_0} \right] \right) \right\}, \quad (i = 1, 2)
\]  

(2.47)

in which \( \sigma'_{pr} \) is the value of \( \sigma'_p \) at the referential temperature \( \theta_r \), \( \alpha \) is the gradient of the line \( \sigma'_p - \log \theta \) curve, and \( \sigma'_0 \) is the pre-consolidation stress. If the stress ratio during the one-dimensional compression is assumed to be constant and initial hardening parameter \( \sigma'_{my}^{(n)} \) corresponds to the consolidation yield stress \( \sigma'_p \), \( C(\theta) \) is a temperature-dependent viscoplastic parameter. Substituting Equation (2.46) into Equation (2.47) yields

\[
C_i (\theta) = C_i (\theta_r) \exp \left\{ m' \tilde{M}^* \left( -\ln \left[ \frac{\theta_r}{\theta} \right]^\alpha \right) \right\}
\]  

(2.48)

\[
C_i (\theta_r) = C'_i \exp \left\{ m' \tilde{M}^* \left( -\ln \left[ \frac{\sigma'_p}{\sigma'_{pr}} \right] \right) \right\}
\]  

(2.49)

From Equation (2.48), the temperature-dependency of the viscoplastic parameter \( C_i \) is obtained as follows:

\[
\frac{C_i (\theta)}{C_i (\theta_r)} = \left[ \frac{\theta}{\theta_r} \right]^{am'\tilde{M}^*}
\]  

(2.50)
Hydrate-bearing soil is composed of four constituents, namely, soil particles, water, gas, and gas hydrates, which are viewed as four independent overlapping continua in the context of the mixture theory. The behavior of multiphase materials can be described within the framework of a macroscopic continuum mechanical approach through the use of the theory of porous media. The theory is considered to be a generalization of Biot's two-phase mixture theory for saturated soil. The four phases represent the constituents as part of the mixture, also referred to as the porous medium. For simplified and practical formulations, the grain particles and the water are assumed to be incompressible.

In the formulations, an updated Lagrangian method with the objective Jaumman rate of Cauchy stress is used for a weak form of the equilibrium equation. An eight-node quadrilateral element with a reduced Gaussian two-point integration is used for the displacement to eliminate shear locking and to reduce the appearance of a spurious hourglass mode. The pore water pressure, the pore gas pressure, and the temperature are defined at the four corner nodes, as shown in Figure 2.4.

![Figure 2.4 Isoparametric elements for the soil skeleton, the pore pressures, and temperature](image-url)
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

2.3.1 Equilibrium equation

In the following, we will derive the rate type of the equilibrium equations by the material derivative of the equilibrium equations in the current configuration. Based on the Truesdell’s “Metaphysical Principles” of the mixture theories (Truesdell, 1984), the momentum balance for each phase is given by,

\[ n^\alpha \rho^\alpha \dot{v}_i^\alpha = \sigma^\alpha_{j,j} + \rho^\alpha n^\alpha \vec{F}_i - \tilde{P}_i^\alpha \quad (\alpha = S, W, G, H) \]  \hspace{1cm} (2.51)

in which the superscript dot (‘) is the time-derivative and subscript \((\ )_j\) indicates the partial derivative with respect to \(x_j\). \(v_i^\alpha\) is the rate vector of phase \(\alpha\), \(\vec{F}_i\) is the gravitational force per unit mass, and \(\tilde{P}_i^\alpha\) is related to the interaction term given in

\[ \tilde{P}_i^\alpha = \sum_\gamma D^{\alpha,\gamma} (v_i^\alpha - \dot{v}_i^\gamma), \quad (\alpha, \gamma = S, W, G) \] \hspace{1cm} (2.52)

where \(D^{\alpha,\gamma}\) \((D^{\alpha,\gamma} = D^{\gamma,\alpha})\) are parameters that describe the interaction with each phase, which is defined as,

\[ D^{WS} = \left(\frac{n^W}{k^W}\right)^2 \rho_w g, \quad D^{GS} = \left(\frac{n^G}{k^G}\right)^2 \rho_g g \] \hspace{1cm} (2.53)

in which \(g\) is the acceleration of gravity, and \(k^W\) and \(k^G\) are the permeability coefficients for the water and gas phases, respectively. The momentum balance for the soil \((S)\) phase and the hydrate \((H)\) phase can be obtained from Equation (2.51) as,

\[ n^S \rho^S \dot{v}_i^S = \sigma^S_{j,j} + \rho^S n^S \vec{F}_i - \tilde{P}_i^S \] \hspace{1cm} (2.54)
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ n^H \rho^H \dot{v}_i^H = \sigma^H_{j,i} + \rho^H n^H \vec{F}_i - \vec{P}_i^H \] (2.55)

We assume that the soil and hydrates behave together as a solid mass; thus, we suppose

\[ v_i^{SH} = \dot{v}_i^s = \dot{v}_i^H \] (2.56)

Considering Equation (2.56) and adding Equations (2.54) and (2.55),

\[ \left( n^s \rho^s + n^H \rho^H \right) \dot{v}_i^{SH} = \left( \sigma^s_{j,i} + \sigma^H_{j,i} \right) + \left( n^s \rho^s + n^H \rho^H \right) \vec{F}_i - \left( \vec{P}_i^s + \vec{P}_i^H \right) \] (2.57)

is obtained. When we consider Equations (2.13) and (2.15), the momentum balance for the solid phase \( (SH) \) is given as

\[ n^{SH} \rho^{SH} \dot{v}_i^{SH} = \sigma^{SH}_{j,i} + n^{SH} \rho^{SH} \vec{F}_i - \vec{P}_i^{SH} \] (2.58)

Substituting Equations (2.11) ~ (2.14) into Equations (2.51) and (2.58), the momentum balance equation for the solid \( (SH) \), water \( (W) \), and gas \( (G) \) phases is obtained, respectively, with the following equations:

Solid:

\[ n^{SH} \rho^{SH} \dot{v}_i^{SH} = \sigma^{SH}_{j,i} - \left( n^{SH} P^e \delta_y \right) + n^{SH} \rho^{SH} \vec{F}_i - D^{SH,W} \left( v_i^{SH} - v_i^W \right) - D^{SH,G} \left( v_i^{SH} - v_i^G \right) \] (2.59)

Water:

\[ n^W \rho^W \dot{v}_i^W = - \left( n^W P^W \delta_y \right) + n^W \rho^W \vec{F}_i - D^{W,SH} \left( v_i^W - v_i^{SH} \right) - D^{W,G} \left( v_i^W - v_i^G \right) \] (2.60)

Gas:

\[ n^G \rho^G \dot{v}_i^G = - \left( n^G P^G \delta_y \right) + n^G \rho^G \vec{F}_i - D^{G,SH} \left( v_i^G - v_i^{SH} \right) - D^{G,W} \left( v_i^G - v_i^W \right) \] (2.61)

When we assume that the deformation is a quasi-static process \( \left( \dot{v}_i = 0 \right) \), the space
derivative of volume fraction $n_i^\beta$ is negligible, and the sum of the momentum balance equations leads to

$$\sigma'_{ji,j} - (n^{SH} p^F \delta_{ji} j) + n^{SH} \rho^{SH} F_i - (n^W p^F \delta_{ji} j) + n^W \rho^W F_i - (n^G p^F \delta_{ji} j) + n^G \rho^G F_i$$

$$= \sigma'_{ji,j} - (n^{SH} + n^W + n^G) (p^F \delta_{ji} j) + (n^{SH} \rho^{SH} + n^W \rho^W + n^G \rho^G) F_i$$

$$= \sigma'_{ji,j} - (p^F \delta_{ji} j) + \rho F_i = 0$$ (2.62)

Substituting Equation (2.18) into Equation (2.62) leads to the equilibrium equation for the whole mixture

$$\sigma_{ji,j} + \rho F_i = 0$$ (2.63)

Following the previous assumptions, disregarding the interaction between the air and water phases, that is, $D^{W,G} = D^{G,W} = 0$, and using Equation (2.53), Equations (2.60) and (2.61) can be written as follows:

$$-(n^W p^W \delta_{ji} j) + n^W \rho^W F_i - \left(\frac{n^W}{k^W}\right)^2 \rho^W g (v_i^W - v^{SH}_i) = 0$$ (2.64)

$$-(n^G p^G \delta_{ji} j) + n^G \rho^G F_i - \left(\frac{n^G}{k^G}\right)^2 \rho^G g (v_i^G - v^{SH}_i) = 0$$ (2.65)

We will define relative velocity vectors $V_{i,\beta}^j$ for the water and air with respect to the solid phase as

$$V_{i,\beta}^j = n^\beta (v_i^\beta - v_i^W), \quad (\beta = W, G)$$ (2.66)
After manipulation of Equations (2.64) and (2.65), and using the relative velocity vector $V^\beta_i$ described in Equation (2.66), a Darcy type of law for the water and gas phases is obtained from the momentum balance equations, respectively, as

$$V^w_i = -\frac{k^w}{\gamma^w} \left( (P^w \delta^w_{ij})_j - \rho^w \overline{F}_i \right), \quad \gamma^w = \rho^w g$$

(2.67)

$$V^G_i = -\frac{k^G}{\gamma^G} \left( (P^G \delta^G_{ij})_j - \rho^G \overline{F}_i \right), \quad \gamma^G = \rho^G g$$

(2.68)

where $g$ is the acceleration of gravity.

### 2.3.1.1 Conservation of momentum for the boundary value problem

In the present study, an updated-Lagrangian method is employed to discretize the governing equations. When we consider an arbitrary domain $V$, with boundary $S$, the increment type of boundary problem is given by

$$\frac{D}{Dt} \int_V \left( \sigma_{ji,j} + \rho \overline{F}_j \right) dV = 0$$

(2.69)

The conservation of linear momentum for the whole mixture in the current configuration is given by the following equation (Higo, 2003; Higo et al., 2006)

$$\frac{D}{Dt} \int_V \rho v_i dV = \int_S t_i dS + \int_V \rho \overline{F}_i dV$$

(2.70)

in which $D/Dt$ is the material time derivative, $\rho$ is the mass density, $v_i$ is the velocity vector, $t_i$ is the surface traction vector, and $\overline{F}_i$ is the body force vector.

In this study, we deal with quasi-static conditions, so that the acceleration and the body force can be assumed to be zero. Consequently, the rate type of equilibrium equation is expressed as follows:
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\frac{D}{Dt} \int_S t_i dS = 0
\]  

(2.71)

The surface traction vector, \( t_i \), is defined by Cauchy’s stress theorem, namely,

\[
t_i = T_{ij} n_j
\]  

(2.72)

where \( T_{ij} \) is the Cauchy’s stress tensor and \( n_j \) is the unit normal vector with respect to the boundary surface \( dS \).

When we take the initial configuration at current time \( t \), the nominal traction \( s_i \) is written as

\[
s_i = \Pi_{ij} N_j
\]  

(2.73)

where \( \Pi_{ij} \) is the nominal stress tensor (the first Piola-Kirchhoff stress tensor) and \( N_j \) is the unit normal vector to \( dS_0 \), which is the surface area in the initial configuration. The relation between the traction force \( t_i dS \) and the nominal traction force \( s_i dS_0 \) can be written as

\[
t_i dS = s_i dS_0
\]  

(2.74)

From Equations (2.72) and (2.73), we obtain the following equation.

\[
T_{ij} n_j dS = \Pi_{ij} N_j dS_0
\]  

(2.75)

Adopting Nanson’s law, that is,
we obtain the relation between the Cauchy’s stress tensor $T_{ij}$ and the nominal stress tensor $\Pi_{ij}$ as

$$\Pi_{ij} = J T_{ij} F^{-1}_{kj}$$

in which $F_{ij}$ is the deformation gradient tensor and $J$ is the Jacobian determinant defined as $J = \text{det}[F]$.

Substituting Equations (2.73) and (2.74) into Equation (2.71) gives the rate type of the equilibrium equations in the initial configuration as

$$\frac{D}{Dt} \int_{S_0} s_j dS_0 = \frac{D}{Dt} \int_{S_0} \Pi_{ji} N_j dS_0 = 0$$

(2.78)

Since $N_j$ and $dS_0$ are independent of time, Equation (2.78) yields

$$\int_{S_0} \dot{\Pi}_{ji} N_j dS_0 = 0$$

(2.79)

where $\dot{\Pi}_{ij}$ is the material time derivative of the nominal stress, which is given by differentiating Equation (2.77) with respect to time

$$\dot{\Pi}_{ji} = J T_{ki} F^{-1}_{kj} + J T_{ki} F^{-1}_{kj} + J T_{ki} F^{-1}_{kj} = J \left( \dot{T}_{ki} + L_{pp} T_{ki} - T_{qi} L_{qk} \right) F^{-1}_{kj}$$

(2.80)

$$L_{iq} = \hat{F}_{ik} F^{-1}_{kj}$$

(2.81)
where \( L_{ij} \) is the velocity gradient tensor.

Combining Equations (2.79) and (2.80) gives

\[
\int_{S_0} J \left( T_{ki} + L_{pp} T_{ki} - T_{qi} L_{qk} \right) F_{kj}^{-1} N_j dS_0 = 0
\]

(2.82)

By adopting Nanson's law and the Gauss theorem, Equation (2.82) provides the rate of the equilibrium equations in the current configuration as

\[
\oint_{V} \hat{S}_{ji,j} dV = 0
\]

(2.83)

\[
\hat{S}_{ji} = \dot{T}_{ji} + L_{pp} T_{ji} - T_{qi} L_{qj}
\]

(2.84)

Finally, the rate type of conservation for the momentum by the material derivative of the equilibrium equation in the current configuration is given by

\[
\hat{S}_{ji,j} = 0
\]

(2.85)

The relation between the Cauchy stress tensor \( T_{ij} \) and the skeleton Cauchy stress tensor \( T_{ij}' \) is given as

\[
T_{ij} = T_{ij}' - P^f \delta_{ij}
\]

(2.86)

\[
\dot{T}_{ij} = \dot{T}_{ij}' - \dot{P}^f \delta_{ij}
\]

(2.87)

where \( \dot{T}_{ij}' \) is the time rate of the skeleton Cauchy stress tensor. The nominal skeleton stress tensor is defined as
### 2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\dot{S}_{\mu j} = \dot{T}_{\mu j} + L_{pp} T_{\mu j} - T_{\mu q} L_{qj} \tag{2.88}
\]

From the above equations, we obtain the relation between the nominal stress tensor and the nominal skeleton stress tensor as follows

\[
\dot{S}_{\mu j} = (\dot{T}_{\mu j} - \dot{P}^F \delta_{\mu j}) + L_{pp} (T_{\mu j} - P^F \delta_{\mu j}) - (T_{\mu q} - P^F \delta_{\mu q}) L_{qj} \\
= (\dot{T}_{\mu j} + L_{pp} T_{\mu j} - T_{\mu q} L_{qj}) - \dot{P}^F \delta_{\mu j} - L_{pp} P^F \delta_{\mu j} + P^F \delta_{\mu q} L_{qj} \tag{2.89}
\]

\[
\dot{S}_{\mu j} = \dot{S}_{\mu j}^t - \dot{P}^F \delta_{\mu j} - L_{pp} P^F \delta_{\mu j} + P^F \delta_{\mu q} L_{qj}
\]

#### 2.3.1.2 Weak form of the rate type of the equilibrium equations

We consider the following boundary conditions for the discretization of the rate type of the equilibrium equations as

\[
\dot{S}_{\mu j} n_j = \dot{S}_{\mu j} \quad \text{on} \quad \partial D_t
\]

\[
v_i = \bar{v}_j \quad \text{on} \quad \partial D_u
\]

Figure 2.5 Boundary conditions for the whole fluid-solid mixture
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ \nu_i = \overline{\nu}_j \text{ on } \partial D_u \quad (2.90) \]

\[ \hat{S}_j^i n_j = \hat{S}_j^i \text{ on } \partial D_i \quad (2.91) \]

in which \( \nu_i \) is the velocity vector, \( n_j \) indicates the unit normal vector to the body, \( \hat{S}_j^i \) is the nominal traction vector, and superposed bar \( (\overline{\cdot}) \) denotes the specified values. The boundary conditions for the whole mixture are illustrated in **Figure 2.5**. The traction vector and displacement should be prescribed at each boundary; they cannot be prescribed at the same point, namely,

\[ \begin{cases} \partial D_i \cap \partial D_i = 0 \\ \partial D_i \cup \partial D_i = \partial D \end{cases} \quad (2.92) \]

As shown in **Figure 2.5**, \( \partial D_i \) and \( \partial D_u \) are the parts of the closed boundary \( \partial D \), on which the stress rate and the displacement rate are prescribed as \( \hat{S}_j^i \) and \( \overline{\nu}_i \), respectively.

Considering the closed domain \( D \) at the current time, \( t = t \), the weak form of the rate type of equilibrium equation, Equation (2.85), is given as follows:

\[ \int_D \hat{S}_{j,i}^i \delta \nu_i dV + \lambda \int_{\partial D} \left( \hat{S}_{j,i}^i - \hat{S}_{j,i}^i n_j \right) \delta \nu_i dS = 0 \quad (2.93) \]

in which \( \delta \nu_i \) is the virtual velocity vector, and \( \lambda \) is an arbitrary scalar.

From the relation

\[ \hat{S}_{j,i}^i \delta \nu_i = \left( \hat{S}_{j,i}^i \delta \nu_i \right)_{j,j} - \hat{S}_{j,i}^i \delta \nu_i \]

Equation (2.93) can be rewritten as

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38
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\int_{\Omega} \left( \hat{S}_{ij} \delta \nu_{ij} \right) dV - \int_{\partial \Omega} \hat{S}_{ij} \delta \nu_{ij} dS + \lambda \int_{\partial \Omega} \left( \hat{S}_{ij} - \hat{S}_{ij} n_{ij} \right) \delta \nu_{ij} dS = 0 \quad (2.95)
\]

Applying the Gauss theorem, and the compatibility equations, i.e., \( \delta \nu_{ij} = \delta L_{ij} \), and considering that the whole boundary can be divided into two parts, that is, \( \partial D_i \) and \( \partial D_u \), Equation (2.95) can be written as

\[
\int_{\partial \Omega} \left( \hat{S}_{ij} \delta \nu_{ij} \right) n_{ij} dS - \int_{\partial \Omega} \hat{S}_{ij} \delta L_{ij} dV + \lambda \int_{\partial \Omega} \left( \hat{S}_{ij} - \hat{S}_{ij} n_{ij} \right) \delta \nu_{ij} dS \\
= \int_{\partial D_u} \left( \hat{S}_{ij} \delta \nu_{ij} \right) n_{ij} dS - \int_{\partial D_i} \hat{S}_{ij} \delta L_{ij} dV + \lambda \int_{\partial D_i} \hat{S}_{ij} \delta \nu_{ij} dS + (1 - \lambda) \int_{\partial D_i} \hat{S}_{ij} n_{ij} \delta \nu_{ij} dS \\
= 0 \quad (2.96)
\]

When we set \( \lambda = 1 \) and \( \delta \nu_{ij} = \delta \overline{\nu}_{ij} \) (constant) on the displacement boundary \( \partial D_u \), Equation (2.96) becomes

\[
\int_{\partial D_i} \hat{S}_{ij} \delta L_{ij} dV = \int_{\partial D_i} \hat{S}_{ij} \delta \overline{\nu}_{ij} dS + \int_{\partial D_u} \hat{S}_{ij} \delta \nu_{ij} n_{ij} dS \quad (2.97)
\]

Now, for the other kinematical admissible strain field, \( \delta L_{ij}^{*} \), and displacement field, \( \delta \nu_{ij}^{*} \),

which is set to be \( \delta \nu_{ij}^{*} = \delta \overline{\nu}_{ij} \) (constant) on the displacement boundary, we obtain a similar weak form. Taking the difference in the weak form and Equation (2.97), we obtain a new weak form, namely,

\[
\int_{\partial D_i} \hat{S}_{ij} \delta L_{ij}^{**} dV = \int_{\partial D_i} \hat{S}_{ij} \delta \nu_{ij}^{**} dS \quad \left( \delta L_{ij}^{**} = \delta L_{ij} - \delta L_{ij}^{*}, \ \delta \nu_{ij}^{**} = \delta \nu_{ij} - \delta \nu_{ij}^{*} \right) \quad (2.98)
\]

By regarding \( \delta L_{ij}^{**} = \delta L_{ij} \) and \( \delta \nu_{ij}^{**} = \delta \nu_{ij} \) again, Equation (2.98) yields

\[
\int_{\partial D_i} \hat{S}_{ij} \delta L_{ij} dV = \int_{\partial D_i} \hat{S}_{ij} \delta \nu_{ij} dS \quad (2.99)
\]
Substituting Equation (2.89) into Equation (2.99) gives

\[
\int_D \hat{S}_{ji}^t \delta L_{ij} dV - \int_D \hat{p}^e \delta \mu \delta L_{ij} dV - \int_D \left( L_{pp} P^e \delta \mu - P^e \delta \nu L_{ij} \right) \delta L_{ij} dV = \int_{\partial D} \hat{S}_{ij}^t \delta v_i dS \quad (2.100)
\]

Since the stretching tensor \( D_{ij} \) is defined as

\[
D_{ij} = \frac{1}{2} \left( L_{ij} + L_{ji} \right) \quad (2.101)
\]

\[
D_{kk} = L_{kk} \quad (2.102)
\]

the following relation is obtained

\[
\delta_{ij} \delta L_{ij} = \delta D_{ij} \quad (2.103)
\]

From the symmetry of the effective Cauchy stress tensor, \( T^e_{ij} = T^e_{ji} \), we obtain

\[
\hat{T}^e_{ij} \delta L_{ij} = \frac{1}{2} \hat{T}^e_{ij} \left( \delta L_{ij} + \delta L_{ji} \right) = \hat{T}^e_{ij} \delta D_{ij} \quad (2.104)
\]

Substituting Equations (2.103), (2.104), and the nominal skeleton stress tensor, Equation (2.88) into Equation (2.100) gives

\[
\int_D \hat{T}^e_{ij} \delta D_{ij} dV + \int_D T^e_{ij} L_{pp} \delta L_{ij} dV - \int_D T^e_{ij} L_{ij} \delta L_{ij} dV - \int_D \hat{p}^e \delta D_{ij} dV

- \int_D \left( L_{pp} P^e \delta \mu - P^e \delta \nu L_{ij} \right) \delta L_{ij} dV = \int_{\partial D} \hat{S}^t_{ij} \delta v_i dS \quad (2.105)
\]

In this formulation, the finite deformation theory and the updated Lagrangian method are adopted. Thus, the Jaumann rate of effective Cauchy stress tensor \( \hat{T}^e_{ij} \) is adopted for the constitutive model. The Jaumann rate of the effective Cauchy stress tensor is the objective
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

tensor; it is defined as

\[
\hat{T}'_{ij} = \hat{T}'_{ij} - W_{ik}T'_{kj} + T'_{ik}W_{kj}
\]  
(2.106)

where \( W_{ij} \) is the spin tensor, namely,

\[
W_{ij} = \frac{1}{2} \left( L_{ij} - L_{ji} \right)
\]  
(2.107)

The stretching tensor is assumed to be a composition of elastic stretching tensor \( D_{ij}^e \) and viscoplastic stretching tensor \( D_{ij}^{vp} \), that is,

\[
D_{ij} = D_{ij}^e + D_{ij}^{vp}
\]  
(2.108)

The relation between elastic stretching tensor \( D_{ij}^e \) and the Jaumann rate of Cauchy stress tensor \( \dot{\hat{T}}_{ij} \) can be obtained as

\[
\dot{\hat{T}}_{ij} = C_{ijkl}^e D_{kl}^e
\]  
(2.109)

where \( C_{ijkl}^e \) is the elastic stiffness matrix.

Herein, the tangent modulus method (Peirce et al., 1984), as will be presented in the next section, is adopted in order to evaluate the viscoplastic stretching tensor \( D_{ij}^{vp} \) (e.g., Higo, 2003; Oka et al., 1992) as

\[
\dot{\hat{T}}_{ij} = C_{ijkl}^{tan} D_{kl} - Q_{ij}
\]  
(2.110)
in which $C_{ijkl}^{\text{tan}}$ is the tangential stiffness matrix, and $Q_j$ is the relaxation stress. Substituting Equation (2.110) into Equation (2.106) yields the following matrix form:

$$\{\ddot{T}\} = [C_{ijkl}^{\text{tan}}] \{D\} - \{Q\} + \{W^*\}$$  \hspace{1cm} (2.111)

where $\{W^*\} = \{WT' - TW\}$ is the vector related to the spin tensor.

According to Equation (2.16), the time derivative of the average pore pressure $P^F$ can be described as

$$\dot{P}^F = \frac{\partial}{\partial t} \left( sP^w + (1-s)P^G \right) = \frac{\partial s}{\partial t} P^w + s\dot{P}^w - \frac{\partial s}{\partial t} P^G + (1-s)\dot{P}^G$$

$$= \frac{\partial s}{\partial P^C} \dot{P}^C P^w + s\ddot{P}^w - \frac{\partial s}{\partial P^C} \dot{P}^C P^G + (1-s)\dot{P}^G$$

$$= \left\{ \frac{\partial s}{\partial P^C} \left( P^w - P^G \right) + (1-s) \right\} \dot{P}^G + \left\{ -\frac{\partial s}{\partial P^C} \left( P^w - P^G \right) + s \right\} \ddot{P}^w$$

$$= \left\{ A_s + (1-s) \right\} \dot{P}^G + \left\{ -A_s + s \right\} \ddot{P}^w$$

$$A_s = \frac{\partial s}{\partial P^C} \left( P^w - P^G \right)$$  \hspace{1cm} (2.112)

For the discretization of the weak form of the equilibrium equation, the following relations are defined:

$$\{v\} = [N]\{v^*\}, \quad \{\delta v\} = [N]\{\delta v^*\}$$  \hspace{1cm} (2.114)

in which $\{v\}$ is the velocity vector in an element, $\{v^*\}$ is the nodal velocity vector, and $[N]$ is a shape function of the eight-node quadrilateral element.

$$\{D\} = [B]\{v^*\}, \quad \{\delta D\} = [B]\{\delta v^*\}$$  \hspace{1cm} (2.115)

in which $[B]$ is the matrix which transforms the nodal velocity vector $\{v^*\}$ into the stretching
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

tensor \( \{D\} \).

\[
\{L\} = [B_m] \{v^r\}, \quad \{\delta L\} = [B_m] \{\delta v^r\}
\]  
(2.116)

where \([B_m]\) is the matrix which transforms the nodal velocity vector into the vector form of velocity gradient vector \( \{L\} \)

\[
\text{tr} \{D\} = [B_v]^T \{v^r\}, \quad \text{tr} \{\delta D\} = [B_v]^T \{\delta v^r\}
\]  
(2.117)

in which \([B_v]\) is the vector which transforms the nodal velocity into the trace of \( \{D\} \).

\[
\{p^w\} = \{N_h\} \{p^{wr}\}, \quad \{\dot{p}^w\} = \{N_h\} \{\dot{p}^{wr}\}
\]  
(2.118)

\[
\{p^s\} = \{N_{h,i}\} \{p^{wr}\} = [B_h] \{p^{wr}\}
\]  
(2.119)

\[
\{p^g\} = \{N_h\} \{p^{gr}\}, \quad \{\dot{p}^g\} = \{N_h\} \{\dot{p}^{gr}\}
\]  
(2.120)

\[
\{p_s^g\} = \{N_{h,i}\} \{p^{gr}\} = [B_h] \{p^{gr}\}
\]  
(2.121)

in which \( \{N_h\} \) represents the element shape function which transforms the nodal pore pressures \( \{p^{wr}\} \) and \( \{p^{gr}\} \) and the nodal pore pressure rate vectors \( \{\dot{p}^{wr}\} \) and \( \{\dot{p}^{gr}\} \) into the pore pressure vectors \( \{p^w\} \) and \( \{p^g\} \) and the rate vectors \( \{\dot{p}^w\} \) and \( \{\dot{p}^g\} \). \([B_h]\) is the matrix that transforms the nodal pore pressure vector into the vector form of the pore pressure gradient.
In addition to the above equations, the other two terms in Equation (2.105) are expressed as

$$\{ -T^r_{pq} L_{pq} \} = [D^r] [B_M] \{ \nu^r \}$$  \hspace{1cm} (2.122)

$$L_{pq} P^P \delta_{ij} - P^P \delta_{ij} L_{pq} = [U] [B_M] \{ \nu^r \}$$  \hspace{1cm} (2.123)

By all the matrix and vector relations obtained previously, Equation (2.105) can be written as

$$\int_D \{ \delta \nu^r \}^T \{ B \}^T \left[ C^{tan} \right] [B] \{ \nu^r \} dV - \int_D \{ \delta \nu^r \}^T \{ B \}^T \{ Q \} dV$$

$$+ \int_D \{ \delta \nu^r \}^T \{ B \}^T \{ W^* \} dV$$

$$+ \int_D \{ \delta \nu^r \}^T \{ B_M \}^T \{ T' \} \{ B \} \{ \nu^r \} dV + \int_D \{ \delta \nu^r \}^T \{ B_M \}^T \{ D^r \} [B_M] \{ \nu^r \} dV$$

$$- \int_D \{ \delta \nu^r \}^T (-A_i + s) \{ B_i \} \{ N_i \} \{ \dot{P}^{tan} \} dV$$

$$- \int_D \{ \delta \nu^r \}^T (A_i + (1-s)) \{ B_i \} \{ N_i \} \{ \dot{P}^{ext} \} dV$$

$$- \int_D \{ \delta \nu^r \}^T \{ B_M \}^T \{ U \} [B_M] \{ \nu^r \} dV = \int_{\partial D} \{ \delta \nu^r \}^T \left[ N \right]^T \{ \tilde{S} \} dS$$

Since the nodal virtual velocity $\{ \delta \nu^r \}$ is arbitrary, from Equation (2.124) we have

$$[K] \{ \nu^r \} + [K_L] \{ \nu^r \} - (-A_i + s) [K_i] \{ \dot{P}^{tan} \} - (A_i + (1-s)) [K_i] \{ \dot{P}^{ext} \} = \{ V \} + \{ F \}$$  \hspace{1cm} (2.125)

in which

$$[K] = \int_D \{ B \}^T \left[ C^{tan} \right] [B] dV$$  \hspace{1cm} (2.126)

$$[K_L] = \int_D \{ B_M \}^T \{ D^r \} [B_M] dV + \int_D \{ B_M \}^T \{ T' \} \{ B_M \}^T dV + \int_D \{ B_M \}^T \{ U \} [B_M] dV$$  \hspace{1cm} (2.127)
2.3 Multiphase Finite Element Formulation for a Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
[K_v] = \int_D \{B_v\}^T \{N_h\} dV
\]

\[\{V\} = \int_{\partial D} [N]^T \{\mathbf{S}\} dS\]

\[
\{F\} = \int_D [B]^T \{Q\} dV - \int_D [B]^T \{W^*\} dV
\]

The relation between the nodal velocity vector \(\{v^*\}\) and the nodal displacement increment vector \(\{\Delta u^*\}\) can be obtained by using Euler's approximation as

\[
\{v^*\} = \frac{\{\Delta u^*\}}{\Delta t}
\]

Similarly, the time derivative of the pore water pressure and the pore air pressure can be obtained as

\[
\{\dot{p}^{W^*}\} = \frac{\{p^{W^*}\}_{t+\Delta t} - \{p^{W^*}\}_t}{\Delta t}, \quad \{\dot{p}^{G^*}\} = \frac{\{p^{G^*}\}_{t+\Delta t} - \{p^{G^*}\}_t}{\Delta t}
\]

Incorporating Equations (2.131) and (2.132) into Equation (2.125) gives

\[
\left(\left[ [K_v] \right] + [K_L] \right) \frac{\{\Delta u^*\}}{\Delta t} - (-A_s + s) [K_v] \frac{\{p^{W^*}\}_{t+\Delta t} - \{p^{W^*}\}_t}{\Delta t}
\]

\[
- (A_s + (1-s)) [K_v] \frac{\{p^{G^*}\}_{t+\Delta t} - \{p^{G^*}\}_t}{\Delta t} = \{V\} + \{F\}
\]

Finally, we obtain the weak form of the equilibrium equations as

\[
\left(\left[ [K_v] \right] + [K_L] \right) \{\Delta u^*\} - (-A_s + s) [K_v] \{p^{W^*}\}_t - (A_s + (1-s)) [K_v] \{p^{G^*}\}_t
\]

\[
= \Delta t \left( \{V\}_t + \{F\}_t \right) - (-A_s + s) [K_v] \{p^{W^*}\}_t - (A_s + (1-s)) [K_v] \{p^{G^*}\}_t
\]
2.3.1.3 Tangent modulus method

In this section, we will derive the relation between the Jaumann rate of Cauchy's stress and the stretching tensor using the tangent stiffness method (Oka et al., 1992; Peirce et al., 1984). As shown in Equation (2.108), the total stretching tensor is being divided between the elastic stretching tensor $D_{ij}^e$ and the viscoplastic stretching tensor $D_{ij}^{vp}$. The elastic stretching tensor is being defined in Equation (2.20). The viscoplastic stretching tensor is defined as

$$D_{ij}^{vp} = C_{ijkl} \left\{ \Phi(f_y) \right\} \frac{\partial f_p}{\partial T_{kl}}$$

(2.135)

Material function $\Phi(f_y)$ depends only on the Cauchy stress tensor $T_{ij}'$ and the viscoplastic volumetric strain $v^{vp} \left( = e_{kk}^{vp} \right)$. Then, the time derivate of $\Phi(f_y)$ can be written as

$$\dot{\Phi}(f_y) = \frac{\partial \Phi}{\partial T_{ij}'} \dot{T}_{ij}' + \frac{\partial \Phi}{\partial v^{vp}} \dot{v}^{vp}$$

(2.136)

where the viscoplastic volumetric strain $v^{vp}$ is expressed as follows

$$v^{vp} = \int_0^t D_{kk}^{vp} dt$$

(2.137)

The rate of Cauchy stress can be derived from the Jaumann rate of Cauchy stress as

$$\dot{T}_{ij}' = \dot{T}_{ij}' - W_{ik} T'_{kj} + T'_{ik} W_{kj}$$

(2.138)

where $W_{ij}$ is the spin tensor.

Using two scalars, $A$ and $B$, and a symmetric tensor $U_{ij}$, we can write
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij} = \frac{\partial \Phi}{\partial T'_{ij}} \left( \dot{T}'_{ij} - W_{ik} T'_{kj} + T'_{ik} W_{kj} \right)
\]

\[
= \frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij} + \frac{\partial \Phi}{\partial T'_{ij}} \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right)
\]

\[
= \frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij} + (A U_{ij} + B \delta_{ij}) \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right)
\]

\[
= \frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij} + A U_{ij} \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right) + B \delta_{ij} \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right)
\]

We then have

\[
A U_{ij} \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right) = A \left( U_{ik} T'_{kj} - U_{ik} T'_{kj} \right) W_{kj} = 0
\]

\[
B \delta_{ij} \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right) = B \left( -W_{ik} T'_{kj} + T'_{ik} W_{kj} \right) = 0
\]

Substituting Equations (2.140) and (2.141) into Equation (2.139), we obtain

\[
\frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij} = \frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij}
\]

Then, substituting Equation (2.142) into Equation (2.136), we have

\[
\Phi = \frac{\partial \Phi}{\partial T'_{ij}} \dot{T}'_{ij} + \frac{\partial \Phi}{\partial v''_{ij}} v''^{ij}
\]

We define the increment of the material function by

\[
\Phi_{t+\Delta t} = \Phi_{t} + \Delta \Phi = \Phi_{t} + \Delta \Phi_{t}
\]

and employ a linear interpolation within the time increment using the tangent modulus parameter \( \theta \) as
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ \Phi = (1 - \theta) \Phi_t + \theta (\Phi_t + \Delta t \Phi_{t+\Delta t}) \]  \hspace{1cm} (2.145)

the parameter \( \theta \) can range from 0 to 1, with \( \theta = 0 \) corresponding to a simple Euler time integration scheme.

Applying Equations (2.143), (2.144), and (2.145), we obtain

\[
\Phi = (1 - \theta) \Phi_t + \theta \left( \Phi_t + \frac{\partial \Phi}{\partial \Delta t} \frac{\Delta t}{\Delta t} \frac{\Delta t}{\Delta t} \right) \\
= (1 - \theta) \Phi_t + \theta \left[ \Phi_t + \frac{\partial \Phi}{\partial \Delta t} \frac{1}{\Delta t} \frac{\Delta t}{\Delta t} + \frac{\partial \Phi}{\partial \Delta t} \frac{1}{\Delta t} \frac{\Delta t}{\Delta t} \right] \\
(2.146)
\]

Using the Equations (2.109) and (2.135), the Jaumann rate for the Cauchy stress can be written as

\[ \dot{T}_{ij} = C_{ijkl}^{e} \left( D_{kl} - D_{kl}^{vp} \right) = C_{ijkl}^{e} \left( D_{kl} - C_{klln} \Phi \frac{\partial f_{p}}{\partial T_{nn}} \right) \]  \hspace{1cm} (2.147)

From Equation (2.135), and the time derivative of viscoplastic volumetric strain \( \dot{\psi}^{vp} \) is expressed as

\[ \dot{\psi}^{vp} \Delta t = D_{kk}^{vp} \Delta t = C_{klln} \Phi \frac{\partial f_{p}}{\partial T_{nn}} \Delta t \]  \hspace{1cm} (2.148)

Substituting Equations (2.147) and (2.148) into Equation (2.146) yields

\[ \Phi = (1 - \theta) \Phi_t + \theta \left\{ \Phi_t + \frac{\partial \Phi}{\partial \Delta t} C_{ijkl}^{e} \left( D_{kl} - C_{klln} \Phi \frac{\partial f_{p}}{\partial T_{nn}} \right) \Delta t + \frac{\partial \Phi}{\partial \Delta t} C_{klln} \Phi \frac{\partial f_{p}}{\partial T_{nn}} \Delta t \right\} \]  \hspace{1cm} (2.149)

Manipulating Equation (2.149) yields
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\Phi \left\{ 1 + \left( \frac{\partial \Delta t}{\partial T_{ij}} \right) \left( \Phi_{ijkl} C_{klmm} - \Phi_{ijkl} C_{klmm} \right) - \left( \frac{\partial \Delta t}{\partial v_{ij}} \right) \Phi_{ijkl} C_{klmm} \right\} \right\} = \Phi_{ijkl} D_{ijkl} (2.150)
\]

\[
\Phi = \frac{1}{1 + \xi'} \left\{ \Phi_{ijkl} + \left( \frac{\partial \Delta t}{\partial T_{ij}} \right) C_{ijkl} D_{ijkl} \right\} (2.151)
\]

\[
\xi' = \left( \frac{\partial \Delta t}{\partial T_{ij}} \right) C_{ijkl} C_{klmm} - \left( \frac{\partial \Delta t}{\partial v_{ij}} \right) C_{ijkl} C_{klmm} \left( \frac{\partial f_{ijkl}}{\partial T_{mn}} \right) (2.152)
\]

Substituting Equation (2.151) into Equation (2.135) leads to

\[
D_{ij} = C_{ijkl} \frac{1}{1 + \xi'} \left\{ \Phi_{ijkl} + \left( \frac{\partial \Delta t}{\partial T_{pq}} \right) C_{ijkl} D_{ijkl} \right\} \left( \frac{\partial f_{ijkl}}{\partial T_{kl}} \right) (2.153)
\]

From Equations (2.153) and (2.147), we obtain

\[
\hat{T}_{ij} + C_{ijkl} D_{kl} = C_{ijkl} C_{klmm} \frac{1}{1 + \xi'} \left\{ \Phi_{ijkl} + \left( \frac{\partial \Delta t}{\partial T_{pq}} \right) C_{ijkl} D_{ijkl} \right\} \left( \frac{\partial f_{ijkl}}{\partial T_{mn}} \right) (2.154)
\]

When we define the tangential stiffness matrix \( C^\text{tan}_{ijkl} \) and the relaxation stress \( Q_{ij} \) as,

\[
C^\text{tan}_{ijkl} = C_{ijkl} - C_{ijkl} C_{ijkl} \frac{1}{1 + \xi'} \left( \frac{\partial \Delta t}{\partial T_{pq}} \right) C_{ijkl} C_{ijkl} \left( \frac{\partial f_{ijkl}}{\partial T_{mn}} \right) (2.155)
\]

\[
Q_{ij} = C_{ijkl} C_{klmm} \frac{1}{1 + \xi'} \left( \frac{\partial f_{ijkl}}{\partial T_{mn}} \right) \Phi_{ijkl} (2.156)
\]
we have the relation between the Jaumann rate of Cauchy’s skeleton stress \( \dot{T}_{ij}^{s} \) and the total stretching tensor \( \dot{D}_{ij} \) as

\[
\dot{T}_{ij}^{s} = C_{ijkl}^{\tan} \dot{D}_{kl} - Q_{ij} = (2.157)
\]

### 2.3.2 Conservation of mass

#### 2.3.2.1 Continuity equation for water and gas

The conservation law of mass for each phase, i.e., soil (\( S \)), water (\( W \)), gas (\( G \)), and hydrate (\( H \)) is given in the following equation as

\[
\frac{\partial}{\partial t} \left( n^\alpha \rho^\alpha \right) + \left( n^\alpha \rho^\alpha v_i^\alpha \right) = \dot{m}^\alpha, \quad (\alpha = S, W, G, H) \tag{2.158}
\]

in which \( \rho^\alpha \) is the material density for each phase, \( \dot{m}^\alpha \) is the mass-increasing ratio per unit volume, generated by dissociation, and \( v_i^\alpha \) is the velocity of phase \( \alpha \).

Considering that the mass-increasing rate for the soil phase should be zero in both dissociation and the formation process of gas hydrates, namely, \( \dot{m}^S = 0 \), Equation (2.158) yields

**Soil:**

\[
\dot{n}^S \rho^S + n^S \dot{\rho}^S + n^S \rho^S v_i^S + n^S \rho^G v_i^G + n^S \rho^H v_i^H = 0 \tag{2.159}
\]

**Water:**

\[
\dot{n}^W \rho^W + n^W \dot{\rho}^W + n^W \rho^W v_i^W + n^W \rho^G v_i^G + n^W \rho^H v_i^H + n^W \dot{\rho}^S v_{ij}^S = 0 \tag{2.160}
\]

**Gas:**

\[
\dot{n}^G \rho^G + n^G \dot{\rho}^G + n^G \rho^G v_i^G + n^G \rho^S v_i^S + n^G \rho^H v_{ij}^H = 0 \tag{2.161}
\]

**Hydrate:**

\[
\dot{n}^H \rho^H + n^H \dot{\rho}^H + n^H \rho^H v_i^H + n^H \rho^G v_i^G + n^H \rho^S v_{ij}^S - \dot{\dot{m}}^H = 0 \tag{2.162}
\]
when the densities of soil, water, and hydrates are assumed to be constant, that is, 
\[ \dot{\rho}^S = \dot{\rho}^W = \dot{\rho}^H = 0, \]
and the spacial gradients of densities and volume fractions are neglected, that is, \( \rho_j^\alpha = n_j^\alpha = 0 \), and considering Equations (2.5)–(2.8), Equations (2.159)–(2.162) yield

**Soil:**

\[
-\dot{n}_i^S \rho^S + (1-n) \rho^S v_{i,j}^S = 0 \tag{2.163}
\]

**Water:**

\[
\begin{align*}
\dot{n}_i^F s \rho^W + n_i^F \dot{s} \rho^W + n_i^F s \rho^W v_{i,j}^W - \dot{m}_i^W \\
= (\dot{n}_i^H - \dot{n}_j^H) s \rho^W + (n_i^H - n_j^H) \dot{s} \rho^W + (n_i^H - n_j^H) s \rho^W v_{i,j}^W - \dot{m}_i^W = 0
\end{align*}
\tag{2.164}
\]

**Gas:**

\[
\begin{align*}
\dot{n}_i^F (1-s) \rho^G - n_i^F \dot{s} \rho^G + n_i^F (1-s) \dot{\rho}^G + n_i^F (1-s) \rho^G v_{i,j}^G - \dot{m}_i^G \\
= (\dot{n}_i^H - \dot{n}_j^H) (1-s) \rho^G - (n_i^H - n_j^H) \dot{s} \rho^G + (n_i^H - n_j^H) (1-s) \dot{\rho}^G \\
+ (n_i^H - n_j^H) (1-s) \rho^G v_{i,j}^G - \dot{m}_i^G = 0
\end{align*}
\tag{2.165}
\]

**Hydrate:**

\[
\dot{n}_i^H = \frac{\dot{m}_i^H}{\rho^H} - n_i^H v_{i,j}^H \tag{2.166}
\]

Multiplying both sides of Equation (2.163) by \( s \rho^W / \rho^S \), and adding Equation (2.164), we obtain the continuity equation for the water phase as

\[
\begin{align*}
(1-n^H) s v_{i,j}^{SH} + \dot{s} n^F - \dot{n}^H s + n^F s (v_{i,j}^W - v_{i,j}^{SH}) - \frac{\dot{m}_i^W}{\rho^W} = 0
\end{align*}
\tag{2.167}
\]

in which the soil and the hydrates are assumed to behave as a solid mass, so that, we suppose \( v_i^{SH} = v_i^S = v_i^H \). Similarly, the continuity equation for the gas phase is obtained by adding Equation (2.163) multiplied by \( (1-s) \rho^G / \rho^S \) and adding Equation (2.165) as
2.3  Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
(1-n^H)(1-s)v_{i,j}^{SH} - n^F \dot{s} - n^H (1-s) + n^F (1-s)(v_{i,j}^G - v_{i,j}^{SH}) + n^F (1-s) \frac{D^G}{\rho^G} - \frac{\dot{m}^G}{\rho^G} = 0
\] (2.168)

The relation between the stretching tensor and the velocity vector for each phase can be written as

\[
D^G_{ij} = \frac{1}{2}(v^G_{i,j} + v^G_{j,i})
\] (2.169)

Tension is considered to be positive in this study, and from Equation (2.169),

\[
v^G_{i,j} = D^G_{ii} = D_{ii}
\] (2.170)

Applying Equation (2.170) and the relative velocity vector described above in Equation (2.66), the conservation law for each phase can finally be defined as

**Soil:**

\[
\dot{n} = (1-n)D_{ii}
\] (2.171)

**Water:**

\[
(1-n^H)sD_{ij} + \dot{s}n^F - \dot{n}^H \dot{s} + V_{i,j}^W - \frac{\dot{m}^W}{\rho^W} = 0
\] (2.172)

**Gas:**

\[
(1-n^H)(1-s)D_{ij} - n^F \dot{s} - \dot{n}^H (1-s) + V_{i,j}^G + n^F (1-s) \frac{D^G}{\rho^G} - \frac{\dot{m}^G}{\rho^G} = 0
\] (2.173)

**Hydrate:**

\[
\dot{n}^H = \frac{\dot{m}^H}{\rho^H} - n^H D_{ii}
\] (2.174)

As for describing changes in the gas density, the equation of ideal gas is used, i.e.,

\[
\rho^G = \frac{M^G}{R \theta} \bar{P}^G
\] (2.175)

in which \( M^G \) is the molecular weight of gas, \( \bar{P}^G \) is the atmospheric pressure, \( R \) is the gas

constant, \( \theta \) is the temperature, and tension is positive in the equation. Considering the time derivative of Equation (2.175), we have

\[
\frac{\dot{\rho}^G}{\rho^G} = \frac{\dot{p}^G}{\bar{p}^G} - \frac{\dot{\theta}}{\bar{\theta}}
\]

(2.176)

As for the mass conservation of the hydrate phase, given by Equation (2.174), \( \dot{n}^H \) is negative when dissociation occurs, and the mass-increasing ratio of hydrates, \( \dot{n}^H \), is calculated from the dissociation rate.

### 2.3.2.2 Weak form of the continuity equation for the water phase

As shown in Figure 2.6, \( D \) demonstrates a closed domain. In a similar manner to that of the boundary for the fluid-solid mixture, the whole boundary surface \( \partial D \) can be divided into two parts, namely, \( \partial D_p \) and \( \partial D_q \).

\[
\begin{aligned}
\partial D_p & \cap \partial D_q = 0 \\
\partial D_p & \cup \partial D_q = \partial D
\end{aligned}
\]

(2.177)

in which \( \partial D_p \) is the boundary where the pore water pressure is specified and \( \partial D_q \) is the boundary where the flow of water is specified.

The boundary conditions for the discretization of the continuity equation of the liquid phase are given by
2.3  Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ P^w = \bar{P}^w \text{ on } \partial D_p \]

\[ V^w = \bar{V}^w \text{ on } \partial D_q \]

**Figure 2.6 Boundary conditions for the water phase**

The specified values are designated by a superposed bar, and \( V^w \) is the velocity of the pore water through the boundary surface \( \partial D_q \).

Considering the test function \( \hat{P}^w \), which satisfies \( \hat{P}^w = \bar{P}^w \text{ on } \partial D_p \), and using the arbitrary scalar \( \lambda \), we obtain the weak form of the continuity equation for the water phase as...
\[
\int_D \left( (1 - n^H) s_D + \dot{s}_n^F - n^H s + V_i - \frac{\dot{m}_w}{\rho_w} \right) \hat{P}_w dV + \lambda \int_{\partial D_s} \left( \overline{V}_i - V_i \right) \hat{P}_w dS = 0 \quad (2.180)
\]

where \( n_i \) is the unit normal vector to the body. From the relation

\[
V_i \hat{P}_w = \left( V_i \hat{P}_w \right)_d - V_i \hat{P}_w
\]

(2.181)

and applying the Gauss theorem, Equation (2.180) can be rewritten as

\[
\int_{\partial D} \hat{P}_w V_i n_i dS - \int_D V_i \hat{P}_w dV + \int_D \hat{P}_w \left( 1 - n^H \right) s_D dV - \int_D \hat{P}_w \sin^H dV
\]

\[
+ \int_D \hat{P}_w \dot{s}_n^F dV - \int D \frac{\dot{m}_w}{\rho_w} dV + \lambda \int_{\partial D_s} \left( \overline{V}_i - V_i \right) \hat{P}_w dS = 0
\]

(2.182)

When we set \( \lambda = 1 \) and \( \hat{P}_w = \overline{P}_w \) (constant) on the pressure boundary, \( \partial D_p \), Equation (2.182) becomes

\[
\int_{\partial D} \overline{P}_w V_i n_i dS - \int_D \hat{P}_w V_i dV + \int_D \hat{P}_w \left( 1 - n^H \right) s_D dV - \int_D \hat{P}_w \sin^H dV
\]

\[
+ \int_D \hat{P}_w \dot{s}_n^F dV - \int_D \frac{\dot{m}_w}{\rho_w} dV + \int_{\partial D_s} \overline{P}_w \overline{V}_i n_i dS = 0
\]

(2.183)

In the same manner as the weak form of the equilibrium equation, considering the other pore water pressure field \( \hat{P}^{w'} \), which is set to be \( \hat{P}^{w'} = \overline{P}_w \) on the pressure boundary \( \partial D_p \), we obtain a similar weak form to Equation (2.183). Then taking the difference in these equations, we get a new weak form of the continuity equation for the water phase, namely,

\[
- \int_D \hat{P}_w^{w*} V_i^w dV + \int D \hat{P}_w^{w*} \left( 1 - n^H \right) s_D dV - \int_D \hat{P}_w^{w*} \sin^H dV
\]

\[
+ \int_D \hat{P}_w^{w*} \dot{s}_n^F dV - \int D \frac{\dot{m}_w^{w*}}{\rho_w} dV + \int_{\partial D_s} \hat{P}_w^{w*} \overline{V}_i n_i dS = 0
\]

(2.184)
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

where \( \hat{p}^{w*} = \overline{p}^w - \hat{p}^w \), and we regard \( \hat{p}^{w*} \) as \( \hat{p}^w \) afterward.

Considering the fact that the degree of saturation \( s \) only depends on the suction \( P^C \), then \( \hat{s} \) is can be given as

\[
\hat{s} = \frac{\partial s}{\partial P^C} \hat{p}^C = \frac{\partial s}{\partial P^C} (\hat{p}^G - \hat{p}^w)
\]

(2.185)

Applying a Darcy type of law, shown in Equations (2.67) and (2.68), to the relative velocity vector \( V^w_i \), and substituting Equation (2.185) into Equation (2.184), we obtain

\[
- \int_D \hat{p}^w \frac{k^w}{\gamma^w} (-P^w + \rho^w F_i) V^w_i dV + \int_D \hat{p}^w (1 - n^H) sD^w dV - \int_D \hat{p}^w s\dot{n}^H dV
\]

\[
+ \int_D \hat{p}^w n^F \frac{\partial s}{\partial P^C} (\hat{p}^G - \hat{p}^w) dV - \int_D \hat{p}^w \frac{\dot{m}^w}{\rho^w} dV + \int_{\partial\Omega} \hat{p}^w \nabla_i n^w dS = 0
\]

(2.186)

Incorporating Equations (2.115), and Equations (2.117)–(2.120) into Equation (2.186), we have

\[
\int_D \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} B_h \end{bmatrix}^T \frac{k^w}{\gamma^w} \begin{bmatrix} P^{w*} \end{bmatrix} dV - \int_D \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} B_h \end{bmatrix}^T \rho^w \begin{bmatrix} F \end{bmatrix} dV
\]

\[
+ \int_D \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} N_h \end{bmatrix}^T (1 - n^H) s \begin{bmatrix} B_h \end{bmatrix} \begin{bmatrix} v^* \end{bmatrix} dV - \int_D \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} N_h \end{bmatrix}^T \dot{s} \dot{n}^H dV
\]

\[
+ \int_D \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} N_h \end{bmatrix}^T n^F \frac{\partial s}{\partial P^C} \begin{bmatrix} \hat{p}^G \end{bmatrix} - \begin{bmatrix} \hat{p}^{w*} \end{bmatrix} dV - \int_D \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} N_h \end{bmatrix}^T \frac{\dot{m}^w}{\rho^w} dV
\]

\[
+ \int_{\partial\Omega} \begin{bmatrix} \hat{p}^{w*} \end{bmatrix}^T \begin{bmatrix} N_h \end{bmatrix}^T \begin{bmatrix} n \end{bmatrix}^T \nabla \hat{w} dS = 0
\]

(2.187)

Considering that the test function \( \{ \hat{p}^{w*} \} \) is arbitrary, the above equation yields
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ -\int_D \{N_h\}^T \left(1-n^H\right) s \{B_v\}^T \{v^s\} dV - \int_D [B_h]^T \frac{k^W}{\gamma^W} [B_v] \{P^{w*}\} dV \]

\[ + \int_D \{N_h\}^T n^F \frac{\partial s}{\partial \rho^c} \{N_h\} \{\dot{P}^{w*}\} dV - \int_D \{N_h\}^T n^F \frac{\partial s}{\partial \rho^c} \{N_h\} \{P^{G}\} dV \]

\[ = \int_{\partial D_v} \{N_h\}^T \{n\}^T \{\bar{V}^w\} dS - \int_D \{N_h\}^T \left(\frac{\dot{m}^W}{\rho^w} + sn^H\right) dV - \int_D [B_h]^T \frac{k^W}{\gamma^W} \rho^w \{F\} dV \]  

(2.188)

Finally, the discretization of the continuity equation for the liquid phase is obtained as

\[ -\left(1-n^H\right) s [K_v]^T \{v^s\} = k^W [K_h] \{P^{w*}\} + n^F \frac{\partial s}{\partial \rho^c} [K_n] \{\dot{P}^{w*}\} \]

\[ -n^F \frac{\partial s}{\partial \rho^c} [K_n] \{\dot{P}^{G}\} = \{V^w\} + \{M^w\} + \{F^w\} \]  

(2.189)

where

\[ [K_v] = \int_D \{N_h\}^T \{B_v\} dV \]  

(2.190)

\[ [K_h] = \int_D [B_h] [B_v] dV \]  

(2.191)

\[ [K_n] = \int_D \{N_h\}^T \{N_h\} dV \]  

(2.192)

\[ \{V^w\} = \int_{\partial D_v} \{N_h\}^T \{n\}^T \{\bar{V}^w\} dS \]  

(2.193)

\[ \{M^w\} = -\int_D \{N_h\}^T \left(\frac{\dot{m}^W}{\rho^w} + sn^H\right) dV \]  

(2.194)

\[ \{F^w\} = -\frac{k^W}{\gamma^W} \int_D [B_h] \rho^w \{F^w\} dV \]  

(2.195)
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

Substituting the Euler’s approximation Equations (2.131) and (2.132) into Equation (2.189), the final discretization equation for the continuity equation for the water phase is obtained as

\[
-\left(1-n^n\right) s \left[K_n\right]^T \left\{\Delta u^s\right\} - \Delta t \frac{k_w}{\gamma_w} \left[K_n\right] \left\{P^W\right\}_{i+\Delta t} + n^n \frac{\partial s}{\partial P^G} \left[K_n\right] \left\{P^G\right\}_{i+\Delta t} = n^n \frac{\partial s}{\partial P^G} \left[K_n\right] \left\{P^G\right\}_i - n^F \frac{\partial s}{\partial P^G} \left[K_n\right] \left\{P^G\right\}_i + \Delta t \left[\left\{V^W\right\} + \left\{M^W\right\} + \left\{F^W\right\}\right] \tag{2.196}
\]

2.3.2.3 Weak form of the continuity equation for the gas phase

As shown in Figure 2.7, the boundary conditions of the discretization of the continuity equation of gas phase are given by

\[
P^G = \bar{P}^G \quad \text{on} \quad \partial D_p \tag{2.197}
\]

\[
V^G = \bar{V}^G \quad \text{on} \quad \partial D_q \tag{2.198}
\]

in which \(\partial D_p\) is the boundary where the pore water pressure is specified and \(\partial D_q\) is the boundary where the flow of water is specified, and they should be satisfy Equation (2.177).

The specified values are designated by a superposed bar, and \(V^G\) is the velocity of the pore water through the boundary surface \(\partial D_q\).

Considering the test function \(\hat{G}\), which satisfies \(\hat{G}^G = \bar{P}^G\) on \(\partial D_p\), and using the arbitrary scalar \(\lambda\), we obtain the weak form of the continuity equation for gas phase as
2.3 Multiphase Finite Element Formulation for Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

Figure 2.7 Boundary conditions for the gas phase

\[
\begin{align*}
\int_{D} \left\{ (1-n^H)(1-s) D_n - \dot{\text{s}} n^F - \dot{n}^H (1-s) + V_{i,n}^G + n^F (1-s) \frac{\dot{\rho}^G}{\rho^G} - \frac{\dot{m}^G}{\rho^G} \right\} \dot{p}^G dV \\
+ \lambda \int_{\partial D} (\vec{V}_{i,n} - \vec{V}_{i,n}) \dot{p}^G dS = 0
\end{align*}
\]  

(2.199)

where \( \vec{n} \) is the unit normal vector to the body. From the relation,

\[
V_{i,j}^G \tilde{p}^G = \left( V_{i,j}^G \tilde{p}^G \right)_j - V_{i,j}^G \tilde{p}^G
\]  

(2.200)

and applying the Gauss theorem, Equation (2.199) can be written as
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ \int_{\partial D} \hat{P}^G V^G_i n_i dS - \int_D V^G_i \hat{P}_d^G dV + \int_D \hat{P}^G (1-n^H)(1-s) D_s dV - \int_D \hat{P}^G n^H (1-s) dV \]

\[ -\int_D \hat{P}^G sn^F dV + \int_D \hat{P}^G n^F (1-s) \frac{\hat{P}^G}{\rho^G} dV - \int_D \hat{P}^G \hat{n}^G dV \]

\[ + \lambda \int_{\partial D_s} (\vec{V}^G_i n_i - V^G_i n_i) \hat{P}^G dS = 0 \]  \hspace{1cm} (2.201)

When we set \( \lambda = 1 \) and \( \hat{P}^G = \bar{P}^G \) (constant) on the pressure boundary \( \partial D_p \), Equation (2.201) becomes

\[ \int_{\partial D_p} \bar{P}^G V^G_i n_i dS - \int_D V^G_i \hat{P}_d^G dV + \int_D \bar{P}^G (1-n^H)(1-s) D_s dV - \int_D \bar{P}^G n^H (1-s) dV \]

\[ -\int_D \hat{P}^G sn^F dV + \int_D \bar{P}^G n^F (1-s) \frac{\hat{P}^G}{\rho^G} dV - \int_D \hat{P}^G \hat{n}^G dV + \int_D V^G_i n_i \bar{P}^G dS = 0 \]  \hspace{1cm} (2.202)

In the same manner as the weak form of the equilibrium equation, considering the other pore water pressure field, \( \hat{P}^{G'} \), which is set to be \( \hat{P}^{G'} = \bar{P}^G \) on the pressure boundary, \( \partial D_p \), we obtain a similar weak form to Equation (2.202). Then taking the difference in these equations, we get a new weak form of the continuity equation for the water phase, namely,

\[ -\int_D V^G_i \hat{P}_d^G dV + \int_D \hat{P}^{G*} (1-n^H)(1-s) D_s dV - \int_D \hat{P}^{G*} n^H (1-s) dV \]

\[ -\int_D \hat{P}^{G*} sn^F dV + \int_D \hat{P}^{G*} n^F (1-s) \frac{\hat{P}^{G*}}{\rho^{G*}} dV - \int_D \hat{P}^{G*} \hat{n}^{G*} dV + \int_D \vec{V}^G_i n_i \hat{P}^{G*} dS = 0 \]  \hspace{1cm} (2.203)

where \( \hat{P}^{G*} = \hat{P}^G - \hat{P}^{G'} \), and we regard \( \hat{P}^{G*} \) as \( \hat{P}^G \) afterward.

Using Equation (2.185) to describe \( \hat{\dot{s}} \) and Equation (2.176) for the time derivative of gas density \( \hat{\dot{\rho}}^G \), and applying a Darcy type of law, shown in Equations (2.67) and (2.68), to the relative velocity vector \( V^G_i \), Equation (2.203) yields
2.3 Multiphase Finite Element Formulation for a Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
- \int_D \hat{P}_i^G k_i^G \left( \frac{\partial \Phi_i^G}{\partial \hat{n}_i^G} \right) (1-n_i^H) (1-s) dV + \int_D \hat{P}_i^G (1-n_i^H) (1-s) D_i dV - \int_D \hat{P}_i^G n_i^F (1-s) \left( \frac{\dot{P}_i^G}{P_i^G} - \theta \right) dV - \int_D \hat{P}_i^G \dot{m}_i^G \frac{\dot{\rho}}{\rho_i^G} dV
\]

+ \int_{\partial D_v} \nabla_i^G \cdot \hat{\nu}_i^G dS = 0

Incorporating Equations (2.115), and Equations (2.117)–(2.121) into Equation (2.204), we have

\[
- \int_D \{\hat{P}_i^G\}^T \left[ B_h \right]^T \frac{k_i^G}{\gamma_i^G} \left[ B_h \right] \{\rho G\} dV - \int_D \{\hat{P}_i^G\}^T \left[ B_h \right]^T \frac{k_i^G}{\gamma_i^G} \{\rho G\} \{F\} dV
\]

+ \int_D \{\hat{P}_i^G\}^T \left[ N_h \right]^T \left( \frac{(1-n_h^H)(1-s)}{\gamma_h^G} \right) \left( \frac{\dot{P}_i^G}{P_i^G} - \frac{\dot{\rho}}{\rho_i^G} \right) dV

+ \int_D \{\hat{P}_i^G\}^T \left[ N_h \right]^T n_i^F \left( \frac{(1-n_h^H)(1-s)}{\gamma_h^G} \right) \left( \frac{\dot{P}_i^G}{P_i^G} - \frac{\dot{\rho}}{\rho_i^G} \right) dV

- \int_D \{\hat{P}_i^G\}^T \left[ N_h \right]^T n_i^F \left( \frac{(1-n_h^H)(1-s)}{\gamma_h^G} \right) \left( \frac{\dot{P}_i^G}{P_i^G} - \frac{\dot{\rho}}{\rho_i^G} \right) dV

- \int_{\partial D_v} \{\hat{P}_i^G\}^T \left[ N_h \right]^T \left( \frac{\dot{m}_i^G}{\rho_i^G} \right) dS - \int_{\partial D_v} \left[ \{\hat{P}_i^G\}^T \left[ N_h \right]^T \left( \frac{\dot{m}_i^G}{\rho_i^G} \right) \{\dot{\nu}_i^G\} \right] dV

- \int_D \left[ B_h \right]^T \frac{k_i^G}{\gamma_i^G} \{\rho G\} \{F\} dV

\]

\[
(2.205)
\]

Since the test function \( \{\hat{P}_i^G\} \) is arbitrary, the previous equation becomes

\[
- \int_D \{N_h\}^T \left( \frac{(1-n_h^H)(1-s)}{\gamma_h^G} \right) \left( \frac{\dot{P}_i^G}{P_i^G} - \frac{\dot{\rho}}{\rho_i^G} \right) dV

+ \int_D \left[ \{N_h\}^T n_i^F \left( \frac{(1-n_h^H)(1-s)}{\gamma_h^G} \right) \left( \frac{\dot{P}_i^G}{P_i^G} - \frac{\dot{\rho}}{\rho_i^G} \right) dV

- \int_D \left[ \{N_h\}^T n_i^F \left( \frac{(1-n_h^H)(1-s)}{\gamma_h^G} \right) \left( \frac{\dot{P}_i^G}{P_i^G} - \frac{\dot{\rho}}{\rho_i^G} \right) dV

= \int_{\partial D_v} \left[ \{N_h\}^T n_i^F \left( \frac{\dot{m}_i^G}{\rho_i^G} \right) \{\dot{\nu}_i^G\} \right] dS

- \int_D \left[ B_h \right]^T \frac{k_i^G}{\gamma_i^G} \{\rho G\} \{F\} dV

\]

\[
(2.206)
\]
Finally, the discretization of the continuity equation for the gas phase is obtained as

\[
-(1-n^H)(1-s)[K_v]^T \{v^*\} - \frac{k_G}{\gamma_G} [K_h]\{P^{G^*}\} - n^F \frac{\partial s}{\partial P^c} [K_n]\{\dot{P}^{w^*}\} \\
+ n^F \frac{\partial s}{\partial P^c} [K_n]\{\dot{P}^{G^*}\} - (1-s)n^F [K'_c] \{\dot{P}^{G^*}\} + (1-s)n^F [K'_c] \{\dot{\theta}^*\}
\]

\[
= \{V^G\} + \{M^G\} + \{F^G\} \tag{2.207}
\]

where

\[
[K_v] = \int_{D} \{N_h\}^T \{B_v\}^T dV \tag{2.208}
\]

\[
[K_h] = \int_{D} \{B_h\}^T [B_h] dV \tag{2.209}
\]

\[
[K_n] = \int_{D} \{N_h\}^T \{N_h\} dV \tag{2.210}
\]

\[
[K'_c] = \int_{D} \frac{\{N_h\}^T \{N_h\}}{P^G} dV \tag{2.211}
\]

\[
[K'_c] = \int_{D} \frac{\{N_h\}^T \{N_h\}}{\theta} dV \tag{2.212}
\]

\[
\{V^G\} = \int_{\partial D_v} \{n\}^T \{\vec{V}^G\} dS \tag{2.213}
\]

\[
\{M^G\} = -\int_{D} \{N_h\} \left( \frac{n^G}{P^G} + n^H (1-s) \right) dV \tag{2.214}
\]
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\{ F^{G} \} = - \frac{k^{G}}{ \gamma^{G}} \int_{V} [B_{h}]^{T} \rho^{G} \{ \ddot{F} \} dV = 0 \quad (2.215)
\]

Substituting the Euler’s approximation Equations (2.131) and (2.132) into Equation (2.207), the final discretization equation for the continuity equation for the water phase is obtained as

\[
\begin{align*}
-(1-n^{H})(1-s)[K^{i}]^{T} \{ \Delta u^{*} \} - \Delta t \, k^{G} \left[ K_{h} \right]^{i} \{ P^{G} \}_{r+\Delta t} - n^{E} \frac{\partial s}{\partial P^{C}} \left[ K_{s} \right]^{i} \{ P^{W} \}_{r+\Delta t} \\
+ n^{E} \frac{\partial s}{\partial P^{C}} \left[ K_{s} \right]^{i} \{ P^{W} \}_{r+\Delta t} -(1-s)n^{E} \left[ K_{h}^{P} \right] \{ P^{G} \}_{r+\Delta t} + (1-s)n^{E} \left[ K_{h}^{G} \right] \{ \theta^{*} \}_{r+\Delta t}
\end{align*}
\]

\[
= \Delta t \left( \{ V^{G} \} + \{ M^{G} \} + \{ F^{G} \} \right) - n^{E} \frac{\partial s}{\partial P^{C}} \left[ K_{s} \right]^{i} \{ P^{W} \}_{r} + n^{E} \frac{\partial s}{\partial P^{C}} \left[ K_{s} \right]^{i} \{ P^{G} \}_{r} \\
- (1-s)n^{E} \left[ K_{h}^{P} \right] \{ P^{G} \}_{r} + (1-s)n^{E} \left[ K_{h}^{G} \right] \{ \theta^{*} \}_{r} \quad (2.216)
\]

2.3.3 Conservation of energy

The following energy conservation equation is applied in order to consider the heat conductivity and the heat sink rate associated with hydrate dissociation (Vardoulakis, 2001),

\[
(\rho c)^{E} \dot{\theta} = D_{ij}^{vp} \sigma_{ij} - \dot{h}_{i} + \dot{Q}^{H} \quad (2.217)
\]

\[
(\rho c)^{E} = \sum_{\alpha} n^{\alpha} \rho^{\alpha} c^{\alpha}, \quad (\alpha = S, W, G, H) \quad (2.218)
\]

where \( c^{\alpha} \) is the specific heat, \( \theta(K) \) is the temperature for all phases, \( D_{ij}^{vp} \) is the viscoplastic stretching tensor, \( \dot{h}_{i} \) is heat flux, and \( \dot{Q}^{H} \) is the dissociation heat rate per unit volume due to hydrate dissociation, which is defined as

\[
\dot{Q}^{H} = \frac{\hat{N}^{H} Q}{V} \quad (2.219)
\]
in which \( Q(\text{kJ/kmol}) \) is the dissociation heat per unit kilomole and it changes with temperature (Kuustraa & Hammershainb, 1983), namely,

\[
Q = 56599 - 16.744\theta
\]  
(2.220)

The term \( D_{ij}^p\sigma'_{ij} \) corresponds only to the work dissipated by the deformation of the solid phase, since all the dissipation in the fluid phase is neglected (Vardoulakis, 2002).

As for the heat flux \( \bar{h}_i \), we assume the validity of Fourier’s law as

\[
\bar{h}_i = -\lambda^E \frac{\partial \theta}{\partial x_i}
\]  
(2.221)

\[
\lambda^E = \sum_{\alpha} n^\alpha \lambda^\alpha, \quad (\alpha = S,W,G,H)
\]  
(2.222)

where \( \lambda^\alpha \) is the thermal conductivity of the \( \alpha \) phase.

Substituting Equation (2.221) into Equation (2.217) yields

\[
(\rho c)^E \dot{\theta} = D_{ij}^p\sigma'_{ij} + \lambda^E \theta_{ii} + \dot{Q}^H
\]  
(2.223)

### 2.3.3.1 Weak form of the conservation of energy

As shown in Figure 2.8, the boundary surface \( \partial D \) can be divided into two parts, namely, \( \partial D_0 \) and \( \partial D_h \).
In the same manner as for the previous boundary conditions, \( \partial D_\theta \) and \( \partial D_h \) must satisfy the following relation:

\[
\begin{align*}
\partial D_\theta \bigcap \partial D_h &= 0 \\
\partial D_\theta \bigcup \partial D_h &= \partial D
\end{align*}
\]  

(2.224)

in which \( \partial D_\theta \) is the boundary where the temperature is specified and \( \partial D_h \) is the boundary where the heat flux is specified.

The boundary conditions for the discretization of the conservation of law of energy are given by

\[
\theta = \bar{\theta} \quad \text{on} \quad \partial D_\theta
\]  

(2.225)
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ h = \bar{h} \quad \text{on} \quad \partial D_h \]  
(2.226)

The superposed bar \( \bar{\cdot} \) indicates the specified values on each boundary.

Considering the test function \( \hat{\phi} \), which satisfies \( \hat{\phi} = \bar{\phi} \) on \( \partial D_\phi \), and using the arbitrary scalar \( \lambda \), we derive the weak form of the conservation law of energy.

\[ \int_D \left\{ \left( \rho c \right)^E \hat{\phi} \sigma_d - D_{ij}^{\sigma} \sigma_{ij} + h_{i,j} - Q^H \right\} \hat{\phi} dV + \lambda \int_{\partial D_h} \left( \bar{h}_i n_i - h_i n_i \right) \hat{\phi} dS = 0 \]  
(2.227)

where \( n_i \) is the unit normal vector to the body. From the relation,

\[ h_{i,j} \hat{\phi} = \left( h_i \hat{\phi} \right)_j - h_i \hat{\phi}_j \]  
(2.228)

and applying the Gauss theorem, Equation (2.227) can be rewritten as

\[ \int_D \hat{\phi} \left( \rho c \right)^E \sigma_d dV - \int_D \hat{\phi} D_{ij}^{\sigma} \sigma_{ij} dV + \int_{\partial D} \hat{\phi} h_i n_i dS - \int_{\partial D_h} \hat{\phi}_j h_j dV - \int_{\partial D_h} \hat{\phi} Q^H dV 
+ \lambda \int_{\partial D_h} \left( \bar{h}_i n_i - h_i n_i \right) \hat{\phi} dS = 0 \]  
(2.229)

When we set \( \lambda = 1 \) and \( \hat{\phi} = \bar{\phi} \) (constant) on the temperature boundary \( \partial D_\phi \), Equation (2.229) becomes

\[ \int_D \hat{\phi} \left( \rho c \right)^E \sigma_d dV - \int_D \hat{\phi} D_{ij}^{\sigma} \sigma_{ij} dV - \int_{\partial D} \hat{\phi}_j h_j dV - \int_{\partial D_h} \hat{\phi} Q^H dV 
+ \int_{\partial D_h} \bar{\phi} h_i n_i dS + \int_{\partial D_h} \bar{h}_i \hat{\phi} dS = 0 \]  
(2.230)

By considering the other temperature field, \( \hat{\phi}' \), which is set to be \( \hat{\phi}' = \bar{\phi} \) on the temperature...
boundary $\partial D_{\theta}$, we obtain a similar weak form to Equation (2.230), then taking the difference in these equations, we have a new weak form of the conservation law of energy as

$$\int_D \left( (\rho c)^E \delta \dot{\theta}^* dV - D_{\gamma}^\theta \sigma^* \dot{\theta}^* dV - \int_{\partial D} \dot{h}_{\gamma} \delta \dot{\theta}^* dS - \int_D \dot{Q}^H \dot{\theta}^* dV + \int_{\partial D} \bar{h}_{\gamma} \delta \dot{\theta}^* dS \right) = 0 \quad (2.231)$$

where $\delta \dot{\theta}^* = \dot{\theta} - \dot{\theta}'$, and we regard $\delta \dot{\theta}^*$ as $\dot{\theta}$ afterward.

For the discretization of the weak form of the equilibrium equation, the following relations are defined

$$\hat{\theta} = \{N_h\}\{\theta^*\}, \quad \dot{\theta} = \{N_h\}\{\dot{\theta}^*\} \quad (2.232)$$

$$\theta_j = \{N_{h,j}\}\{\theta^*\} = [B_h]\{\theta^*\} \quad (2.233)$$

$$h_i = -\lambda^E \theta_j = -\lambda^E [B_h]\{\theta^*\} \quad (2.234)$$

Substituting Equations (2.232)–(2.234) into Equation (2.231) yields

$$\int_D \left\{ \{\dot{\theta}^*\}^T \{N_h\}^T (\rho c)^E \{N_h\}\{\dot{\theta}^*\} dV - \int_D \left\{ \{\dot{\theta}^*\}^T \{N_h\}^T \{\sigma^*\}^T \{D_{\gamma}^\theta\} \right\} dV + \int_{\partial D} \left\{ \{\dot{\theta}^*\} \left[ B_h \right] \{\theta^*\} dV - \int_D \left\{ \{\dot{\theta}^*\}^T \{N_h\}^T \dot{Q}^H \right\} dV + \int_{\partial D} \left\{ \{\dot{\theta}^*\}^T \{N_h\}^T \{\bar{h}\} \{n\} \right\} dS = 0 \quad (2.235)$$

Since the test function $\{\theta^*\}$ is arbitrary, the previous equation becomes
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[
\int_D \{N_h\}^T (\rho c)^E \{\dot{\theta}^r\} dV - \int_D \{N_h\}^T \{\sigma'_{ij}\}^T \{D_{ij}^p\} dV \\
+ \int_D [B_h]^T \lambda^E [B_h] \{\dot{\theta}^r\} dV - \int_D \{N_h\}^T \dot{Q}^H dV + \int_{\partial D_h} \{N_h\}^T \{\vec{h}\} \{n\} dS = 0 
\] (2.236)

Finally, the discretization of the conservation law of energy is obtained as

\[
(\rho c)^E [K_h] \{\dot{\theta}^r\} + \lambda^E [K_h] \{\dot{\theta}^r\} = \{V^o\} + \{M^o\} + \{F^o\} 
\] (2.237)

where \([K_h]\) and \([K_n]\) are the same as Equations (2.209) and (2.210), respectively, and other vectors are defined as follows:

\[
\{V^o\} = \int_{\partial D_h} \{N_h\}^T \{\vec{h}\} \{n\} dS 
\] (2.238)

\[
\{M^o\} = \int_D \{N_h\}^T \dot{Q}^H dV 
\] (2.239)

\[
\{F^o\} = \int_D \{N_h\}^T \{\sigma'_{ij}\}^T \{D_{ij}^p\} dV 
\] (2.240)

In the same manner as for Equations (2.131) and (2.132), the time derivative of the temperature is expressed as

\[
\{\dot{\theta}^r\} = \frac{\{\theta^r\}_{t + \Delta t} - \{\theta^r\}_t}{\Delta t} 
\] (2.241)

Substituting Equation (2.241) into Equation (2.237), the final discretization equation for the conservation law of energy is obtained as

\[
(\rho c)^E [K_n] \{\theta^r\}_{t + \Delta t} + \Delta t \lambda^E [K_h] \{\theta^r\}_{t + \Delta t} = \Delta t \left(\{V^o\} + \{M^o\} + \{F^o\}\right) \\
+ (\rho c)^E [K_n] \{\theta^r\}_t 
\] (2.242)
2.3.4 Dissociation rate of methane hydrates

Figure 2.9 schematically illustrates the equilibrium curve for methane hydrates. The stability of the hydrates depends on temperature and pressure, and the hydrates are stable at low temperature and high pressure conditions, which is shown above the equilibrium curve in Figure 2.9. Bejan et al. (2002) proposed the equilibrium pressure \( P^e [\text{MPa}] \) as a function of the current temperature \( \theta_c [\text{K}] \) as

\[
P^e = c \exp \left( a - \frac{b}{\theta_c} \right), \quad a = 49.3185, \quad b = 9459, \quad c = 1.0 \times 10^{-6} \quad [\text{MPa}] \tag{2.243}
\]

When the current pore pressure is larger than the equilibrium pressure, namely, \( P > P^e \), the methane hydrates can exist as hydrates, whereas, in the case of \( P < P^e \), the methane hydrates are outside the stable region, namely, the methane hydrates dissociate into gas and water. The equation for the decomposition reaction of methane hydrates is expressed as
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ \text{CH}_4 \cdot n\text{H}_2\text{O(hydrate)} \rightarrow \text{CH}_4(\text{gas}) + n\text{H}_2\text{O(water)} \quad (2.244) \]

where \( n \) is a hydrate number and is assumed to be 5.75.

Kim et al. (1987) proposed the following equation for the decomposition rate of methane hydrates:

\[ \dot{N}^H = - \left( \frac{7.44 \times 10^{11}}{\Psi P^H D_0} \right) \exp \left( - \frac{9400}{\theta} \right) \left( P^e - P \right) N_{H_0}^3 N_H^2 \quad (2.245) \]

where \( N_H \) is the moles of hydrates in a volume \( V \ [\text{m}^3] \) and \( N_{H_0} \) is the value at the initial state, \( \Psi \) is the sphericity factor, \( P^H \ [\text{mol}/\text{m}^3] \) is the density of moles per unit volume, and \( D_0 \ [\text{m}] \) is the diameter of the hydrate particle. Kim et al. (1987) assumed that the hydrate particles have the same diameter \( D_0 \) before the decomposition, and may be not variable either before or after the dissociation. The term \( \Psi P^H D_0 \) is related to the geometry of the hydrate particles, and it is defined as

\[ \frac{\Psi P^H D_0}{6} = 2.12 \times 10^{-2} \ [\text{mol}/\text{m}^2] \quad (2.246) \]

Substituting Equation (2.246) into Equation (2.245), we obtain the decomposition rate of methane hydrates as

\[ \dot{N}^H = -D^H \exp \left( - \frac{9400}{\theta} \right) \left( P^e - P \right) N_{H_0}^3 N_H^2 \ [\text{kmol/sec}] \quad (2.247) \]

\[ D^H = 5.85 \times 10^{12} \ [\text{l}/\text{MPa} \cdot \text{sec}] \quad (2.248) \]

From Equations (2.244) and (2.247), the generating rate [\text{kmol/sec}] of water and gas due to
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

the dissociation are given as

\[
\dot{N}^W = -5.75\dot{N}^H, \quad \dot{N}^G = -\dot{N}^H
\]  
(2.249)

The mass-increasing ratios for the methane hydrate, water, and gas phases, which are required in the conservation law of mass described Equations (2.160)–(2.162) (see Section 2.3.2) can be calculated from the above equations as follows:

\[
\dot{m}^H = \frac{\dot{N}^HM_H^H}{V} \times 10^{-3} \text{ [t/m}^3\text{·sec]}  
\]  
(2.250)

\[
\dot{m}^W = \frac{\dot{N}^WM_W^W}{V} \times 10^{-3} = -\frac{5.75\dot{N}^HM_W^W}{V} \times 10^{-3} \text{ [t/m}^3\text{·sec]}  
\]  
(2.251)

\[
\dot{m}^G = \frac{\dot{N}^GM_G^G}{V} \times 10^{-3} = -\frac{\dot{N}^HM_G^G}{V} \times 10^{-3} \text{ [t/m}^3\text{·sec]}  
\]  
(2.252)

where \( M_H^H, M_W^W, \) and \( M_G^G \) are the molar mass \([\text{kg/kmol}]\) of each phase.

As for the ratio of dissociation heat rate per unit due to hydrate dissociation \( \dot{Q}^H \) described in Equation (2.219), it can also be calculated from Equation (2.247).

### 2.3.5 Effect of the hydrate saturation on permeability

The development of a mathematical model that is able to estimate the gas production rate from hydrate reservoirs is an important aspect of production scenarios. Some researchers have pointed out that permeability is an important parameter for predicting the flows of water and gas in the reservoir (Hong & Pooladi-Darvish, 2005; Masuda et al., 1997; Ordonez et al., 2009). Hong and Pooladi-Darvish (2005) have illustrated the variation in gas production predictions depending on the hydrate content of the soil, and its behavior depends significantly on the growth habits of the hydrates. Masuda et al. (2002) have proposed the following equation of permeability coefficients for water and gas as a function of the hydrate saturation \( S_r^H \).
2.3 Multiphase Finite Element Formulation for A Dissociation-Deformation Coupled Analysis of Methane Hydrate-Bearing Sediments

\[ k_D = k_{D0} \left(1 - S_r^H \right)^N \] (2.253)

\[ k^W = k_0^W \exp\left(\frac{e-e_0}{2}\right) \left(1 - S_r^H \right)^N, \quad k^G = k_0^G \exp\left(\frac{e-e_0}{2}\right) \left(1 - S_r^H \right)^N \] (2.254)

where \( N \) is the reduction parameter of the permeability. In the present models, we employ Equation (2.254) as the permeability coefficients for water and gas.

2.3.6 Soil-water characteristic curve

The soil-water characteristic curve (SWCC) for soil is defined as the relationship between the volumetric water content or degree of saturation and the suction of the soil. The SWCC for unsaturated soil is required to describe the behavior of the gas hydrates-bearing sediments, because the sediments will become unsaturated state due to the hydrate dissociation. It can be described as the water content changes when it is subjected to various levels of the suction. In the model, we adopt the following type of formulation for the SWCC proposed by van Genuchten (van Genuchten, 1980).

\[
\begin{align*}
    s_{re} &= \left\{1 + \left(\alpha P^c \right)^{n'}\right\}^{-m} \\
    m &= 1 - \frac{1}{n'}
\end{align*}
\] (2.255)

where \( \alpha \), \( n' \) are material parameters and \( s_{re} \) is effective degree of the water saturation, namely,

\[ s_{re} = \frac{s - s_{\text{min}}}{s_{\text{max}} - s_{\text{min}}} \] (2.256)

in which \( s_{\text{max}} \) and \( s_{\text{min}} \) are the maximum and minimum water saturation, respectively.
2.4 Summary

In this chapter, an elasto-viscoplastic constitutive equation for the gas hydrate-bearing sediment proposed by Kimoto et al. (2007a) was introduced. The constitutive model is based on the overstress-type of viscoplastic theory proposed by Adachi and Oka (1982) with considerations of soil structure degradation for saturated soil (Kimoto & Oka, 2005; Kimoto et al., 2004) and the effect of the suction using the skeleton stress and the suction for independent stress variables (Oka et al., 2008a). In addition, Kimoto et al. (2007a) developed a model taking into account the phase changes from solids to fluids, that is, water and gas, the flows of water and gas, and the heat transfer, in order to simulate the ground deformations subjected to gas hydrate dissociation. In addition, a finite element formulation for the introduced governing equations, based on the finite deformation theory, has been shown in this chapter. The examples of a numerical analysis for MH-bearing sediments will be discussed in a later chapter.
2.4 Summary
Chapter 3

FORMATION AND DISSOCIATION TESTS ON SAND SPECIMENS CONTAINING CARBON DIOXIDE HYDRATES UNDER UNDRAINED CONDITIONS

3.1 Introduction

Methane hydrates have been viewed as a new potential energy resource. They compose an unconventional resource; thus, in order to produce methane gas from a methane hydrate reservoir, the methane hydrates should be dissociated in the sediments. At least four ways are proposed to dissociate the methane hydrates: (1) the thermal recovery method, (2) the depressurization method, (3) inhibitor injection, and (4) the CH₄-CO₂ swapping method. With all the approaches, the methane hydrates may dissociate into water and gas, and these fluid flows may alter the pore fluid pressures and the effective stress in the methane hydrate reservoir. When hydrates dissociate, the bonding effect between soil particles may be lost, and the pore water pressure and pore gas pressure may increase. These changes in effective stress and the degradation of the strength of the sediments can accelerate the consolidation and shear
deformation of the ground and, in turn, produce geological disasters. Thus, characterizing the geo-mechanical properties and responses of hydrate-bearing sediments during dissociation is an important challenge faced by experts in the field of natural gas production.

Some experimental studies on the mechanical properties of gas hydrate-bearing sediments have been conducted (e.g., Ghiassian and Grozic, 2013; Grozic and Ghiassian, 2008; Hyodo et al., 2014a, 2014b; Miyazaki et al., 2011a, 2011b, 2010). They mentioned that the shear strength of hydrate-bearing soils becomes larger than soils without hydrates. However, there are still many geo-mechanical issues in the production process to be addressed, especially in relation to the ground deformation caused by dissociation. A few studies have been developed to investigate the deformation behavior of methane hydrate-bearing sediments induced by dissociation.

Sakamoto et al. (2008) conducted experimental studies on consolidation and gas production behavior during MH dissociation by the depressurization method, using a special apparatus. In order to reproduce the real flow conditions of gas and water in the deep seabed ground, they used disc-shaped samples as the simulated MH sediments and the horizontal radial flow during dissociation was produced; the vertical load system was used to simulate hydrate-bearing sediments. From the experimental observation, it was found that the increase in effective stress produced by depressurization was the dominant factor in the compaction behavior and that the deformation after the constant effective stress condition was primarily dependent on the creep effect rather than the MH dissociation.

Wu et al. (2008) reported experimental results on carbon dioxide hydrate-bearing sand for the purpose of understanding the physical effects imposed by hydrate dissociation, such as changes in stress and deformation. They conducted nine tests under three different pressure levels and three different temperatures where gas hydrates were formed, and subsequently dissociated, using a special high-pressure, low-temperature triaxial apparatus created to study the geo-mechanical behavior of gas hydrates and their interaction with soil. The triaxial test data indicated that (1) higher pressure levels produce higher hydrate contents, (2) lower temperatures can create higher hydrate contents, (3) greater amounts of gas hydrates initially existing in the specimen cause greater reductions in effective stress due to dissociation and lower strength, and (4) the dissociation of even a small percentage of gas hydrates causes a dramatic reduction in the soil strength and can lead to failure. These tests were conducted under undrained conditions; however, the hydrate saturation was around 10%, which is relatively low compared to that of real sediments.

Hyodo et al. (2014b) investigated the shear strength and deformation behavior of methane
hydrate-bearing sediments during dissociation using both the thermal recovery method and the depressurization method. Shear tests were conducted under drained conditions. From the experiments, it was found that (1) the failure strength of isotropically consolidated methane hydrate-bearing sediments, dissociated by the heating method, is less than that of pure Toyoura sand, and (2) the depressurization method will not cause the collapse of methane hydrate-bearing sediments during depressurization, although the recovery of water pressure after depressurization will lead to failure when the axial load is larger than the strength of the methane hydrate-bearing sediments. They pointed out the possibility that the shear strength may change before and after the dissociation.

For the safe and economical production of methane gas from MH reservoirs, further studies on the deformation behavior during dissociation are required. This chapter notes the results of an experimental program conducted on carbon dioxide hydrate-bearing sand to understand the physical effects, such as changes in stress and deformation, imposed by hydrate dissociation. Of course, it is preferable to use methane gas and to form MHs in order to simulate the real environment of sediments containing MHs. However, some researchers have pointed out that the differences in mechanical properties between CO$_2$-hydrates and MHs are small (e.g., Espinoza & Santamarina, 2011; Hyodo et al., 2014a; Miyazaki et al., 2013).

In the experiments, CO$_2$ gas is chosen as the guest gas, because it is nonflammable and CO$_2$-hydrates can be formed at relatively lower pressure levels compared to MHs. As the host soil material, Toyoura sand is used. After forming CO$_2$-hydrates in the sand specimen, dissociation tests are conducted using the thermal recovery method under undrained conditions. The tests are conducted under undrained conditions because it is important to consider the most dangerous situations where the pore pressure will not be allowed to dissipate after hydrate dissociation and the seabed ground will be exposed locally under undrained conditions.

### 3.2 Testing Apparatus

In order to create an environment where hydrates can be formed, namely, the conditions of the seabed ground, we have developed a special triaxial apparatus for the experimental program. It is a high-pressure, low-temperature triaxial apparatus which has been made specially to investigate the geo-mechanical behavior of gas hydrate-bearing sand specimens.
3.2 Testing Apparatus

Photo 3.1 Exterior appearance of temperature-controlled high-pressure triaxial apparatus

Figure 3.1 Schematic diagram of the triaxial cell and piping systems

(1) Specimen
(2) Cell pressure pump
(3) Pore pressure pump (upper)
(4) Pore pressure pump (lower)
(5) Outer cell
(6) Inner cell
(7) CO₂ gas saturated water chamber
(8) CO₂ gas tank
(9) Cell fluid cooling tank
(10) Cooling fluid circulating room
(11) Thermometer
(12) Load cell
(13) Volume change meter
(14) Gas flow meter
(15) Pore pressure transducers
(16) Cell pressure transducer
(17) Vertical displacement transducer
3.2 Testing Apparatus

The system is almost the same as a conventional triaxial testing system for soft rocks or hard clays, and we newly installed a thermal control tank, a high-pressure gas application system, and a double-cell structure in order to measure the volumetric strain during the dissociation process of hydrates. Photo 3.1 shows the exterior appearance of the equipment, and Figure 3.1 illustrates the schematic diagram of the triaxial system. Details for each part of the device are provided below.

1) **Test specimen**: Toyoura sand is used for the specimen. The specimen is cylindrical with a diameter of 35 mm and a height of 70 mm.

2) **Cell pressure pump**: The cell pressure of both outer and inner cells is controlled using this device. There is a piston inside the cylinder with a diameter of 50 mm; the piston is controlled by a stepping motor. The maximum pressure is 20 MPa and the maximum volume is 416 cm$^3$.

3) **Pore pressure pump (upper)**: This pump can control pore pressure up to 20 MPa from both the top and bottom of the specimen. The system is almost the same as the one for the cell pressure. The differences are that the diameter of the piston is 25 mm and the total volume of the cylinder is 105 cm$^3$.

4) **Pore pressure pump (lower)**: This pump can control the pore pressure in the same way as the upper pore pressure pump and the cell pressure pump. The diameter of the piston is 80 mm and the total volume of the pump is 1000 cm$^3$, while the maximum pressure is 8 MPa, which is less than that of the other pressure cylinder.

5) **Outer cell**: This cell is designed to resist pressure up to 20 MPa applied by the cell pressure pump. For the cell fluid, we employ ethylene glycol brine, which is an antifreeze solution that can be employed as liquid at $-40^\circ$C. It is useful for reproducing low temperature conditions.

6) **Inner cell**: There is a possibility that the specimen will become unsaturated during testing, especially in the dissociation process. Thus, we adopt a double-cell system to measure the volume changes of the specimen. The inner cell is filled with brine, and the volume changes of the cell fluid in the inner cell are measured with a volume change transducer, numbered (13) in Figure 3.1. The volume changes of the specimen can be obtained indirectly. The maximum capacity of this cell pressure is also 20 MPa.

7) **CO$_2$ gas-saturated water chamber**: In this chamber, it is possible to saturate water with
carbon dioxide gas that is required in the process of the water saturation. In order to prevent the carbon dioxide hydrates in the specimen from dissolving into pure water, it is necessary that water saturated with carbon dioxide gas be prepared. Then, the water is injected into the specimen using the lower pore pressure pump.

(8) **CO₂ gas tank:** In this test program, we use carbon dioxide gas instead of methane gas, although it is preferable to use methane hydrates to test real situations, because methane hydrates are nonflammable and can be formed at a relatively lower pressure than methane gas. The carbon dioxide gas is supplied from this tank to the pore space (1), the CO₂ gas-saturated water tank (7), and both upper and lower pore pressure pumps (3) and (4).

(9) **Cell fluid thermal control tank:** The temperature of the brine in the tank can be controlled in the range of -30°C ~ +50°C. This tank has a pump that can circulate the liquid brine through the outer cell in order to cool the system. The brine is not only an antifreeze liquid, but also a non-corrosive liquid against several metallic materials.

(10) **Cooling fluid circulating room:** This room is connected to the cell fluid thermal control tank (9). By circulating cooling fluid inside the room, the temperature of the cell can be maintained at the desired value.

(11) **Thermometer:** A thermometer is installed near the side of the specimen in the triaxial room; it can measure the temperature in the inner cell.

(12) **Load cell:** The load cell, which can apply vertical pressure is equipped in the triaxial cell. The maximum load is 200kN.

(13) **Volume-change transducer:** A volume-change transducer is equipped in order to measure the volume changes of the specimen. The device has a piston in the cell, and it is connected to the cell pressure intensifier and the inner cell. When the volume of the brine in the inner cell changes, the piston of the transducer may also change. Thus, we can obtain the volume changes of the specimen. The maximum pressure is 20 MPa, and it can measure volume changes up to 50.9 cm³.

(14) **Gas flow meter:** In order to estimate the hydrate saturation in the specimen, a gas flow meter is equipped in the system. The gas produced by the dissociation will be collected through the device and calculated under the standard status. It can measure the amount of gas volume in the range of 30 cm³/min ~ 777 cm³/min.
(15) **Pore pressure transducer:** There are two pore pressure transducers; one measures the pore pressure at the top of the specimen, and the other measures the pressure at the bottom. They can measure a maximum of 20 MPa of pore pressure. We do not have a device that can divide the pore pressure into pore water pressure and pore gas pressure, although the specimen may become unsaturated due to hydrate dissociation. Hence, it is impossible to measure the pore water pressure and the pore gas pressure independently in this experiment.

(16) **Cell pressure transducer:** The cell pressure transducer measures the pressure of the inner cell. It is assumed that the outer cell pressure and the inner cell pressure are the same, because they are supplied from one piston, as shown in Figure 3.1-(2).

(17) **Vertical displacement transducer:** A vertical displacement transducer is installed on the outside of the outer cell. The measurable range is 25 mm, that is, about 35% of the axial strain of the specimen with a height of 70 mm.

All the measuring devices and stepping motors are connected to an external computer. Each pressure intensifier is equipped with a stepping motor and can be controlled by pulses produced by the computer. The control program is written in Visual Basic and was developed by Microsoft, Inc.

### 3.3 Testing Procedures

#### 3.3.1 Sample preparation

As for the host soil material, we chose Toyoura sand, because according to the report by Suzuki et al. (2009), the methane hydrates in the Nankai Trough exists in the sandy layer. They have investigated the grain size distribution curve, shown in Figure 3.2, of an undisturbed core sample taken from the Nankai Trough. In preparing the sand specimen, we refer to the method proposed by Hyodo et al. (2013a). The material properties of Toyoura sand are described in Table 3.1 (Shiraishi, 2013). Details of the sample preparation are given as follows.

**Step-1.** First, the amount of water and Toyoura sand are mixed so that the water content becomes the target value of the hydrate saturation as shown in Photo 3.2. The weight of Toyoura sand is determined from the void ratio. The hydrate saturation is expressed by
the following equation:

\[ S_r^H = \frac{V^H}{V^v} \]  

(3.1)

where \( V^H \) and \( V^v \) are the volume of the hydrates and the void, respectively.

**Step-2.** Next, the moist sand is then placed in a mold using the moist tamping method. The mold measures 35 mm in diameter and 150 mm in height. The picture and the pattern diagram of the mold and spacer blocks are shown in Photo 3.3, Photo 3.4, and Figure 3.3, respectively.

<table>
<thead>
<tr>
<th>Table 3.1 Material properties of Toyoura sand (Shiraishi, 2013)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average grain size ( D_{50} ) [mm] 0.24</td>
</tr>
<tr>
<td>Uniformity coefficient ( U_c = D_{60}/D_{10} ) 1.27</td>
</tr>
<tr>
<td>Density ( \rho^s ) [g/cm3] 2.64</td>
</tr>
<tr>
<td>Maximum void ratio ( e_{\text{MAX}} ) 0.975</td>
</tr>
<tr>
<td>Minimum void ratio ( e_{\text{MIN}} ) 0.614</td>
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</tbody>
</table>

Figure 3.2 Grain size distributions of Toyoura sand and samples obtained from Nankai Trough
3.3 Testing Procedures

Photo 3.2 Mixture of Toyoura sand and water with a certain water content

Photo 3.3 Instruments for preparing sand specimen
3.3 Testing Procedures

Photo 3.4 Mold for freezing the moist sand and spacer blocks

Figure 3.3 Schematic diagram of the mold and spacer blocks
3.3 Testing Procedures

**Step-3.** Then, two spacer blocks are set in the bottom of the mold. OHP film is also placed in the mold in order to prevent the moist specimen from becoming dry.

**Step-4.** Moist sand with a certain water content is placed in the mold in about 10 layers, and each layer is compacted by a tamper, as shown in Photo 3.3.

**Step-5.** Similar to Step-1, OHP film is again placed on the compacted sand, and two other spacers are placed on that.

**Step-6.** In order to maintain the water content in the specimen, the mold is packed in a plastic bag, as shown in Photo 3.5.

**Step-7.** The mold is then put in a freezer and left for one day so that the specimen can stand by itself.

**Step-8.** In advance, the pedestal is set on the triaxial cell. Then, a porous metal and a paper filter are placed on the pedestal sequentially from the pedestal.

**Step-9.** Next, the frozen specimen is removed from the mold with a hydraulic jack. Photo 3.6 illustrates the jack used to remove the specimen. It is placed on the pedestal and covered with a membrane. A low temperature and high pressure will be imposed on the specimen. Furthermore, it is possible for carbon dioxide gas to osmose through the latex membrane, which is usually used in conventional triaxial tests. Thus, chloroprene-type membranes with a thickness of 0.5 mm were used in the tests, as illustrated in Photo 3.7.
3.3 Testing Procedures

Photo 3.5 Mold packed in a plastic bag

Photo 3.6 Hydraulic jack
Step-10. A top cap is installed on the specimen and the double cell is assembled. The pipes are connected to both the top and bottom of the specimen, and the inner cell is filled with brine at a temperature of 10°C. Photo 3.8 is the photo of the inner cell and the connected lines after setting up the specimen.
Step-11. Finally, the triaxial room is enclosed by the outer cell and filled with brine. Then, the line of brine is switched to the circulating room in order to control the temperature.

3.3.2 Formation process of carbon dioxide hydrates

After the preparation of the specimen and the setting process, the specimen is subjected to the formation process of the carbon dioxide hydrates. We adopt the partial water saturation method to form the hydrates in the specimen, and this method involves two approaches. In the first approach, referred to as “excess-gas” (Priest et al., 2009; Waite et al., 2004), the soil specimen is prepared with a predetermined moisture content and compacted to form a partially saturated specimen. The specimen is then pressurized with methane gas and cooled in the stability zone to promote hydrate formation. In the second approach, referred to as “excess-water”, the sample is initially prepared, fully saturated with water, and then methane is percolated through the sample, pushing a predetermined amount of water out of the sample prior to cooling it (Winters et al., 2000). They showed that for this excess-water formation methodology, hydrates will form around the gas bubbles and lead to a frame-supporting response at small strain levels. In these tests, the distribution of hydrates may not be adequately controlled.

In our study, we use the method of “excess-gas” to form carbon dioxide hydrates. The phase equilibrium curve for carbon dioxide is shown in Figure 3.4. The red solid line indicates the boundary between the CO₂-hydrate (H), CO₂-gas (G), and liquid water (L_w), while the black dashed line is the boundary of liquid (L_CO2) and gas (G). Hence, the hydrates can be formed in the shadowed area. The pressure and temperature paths to form the hydrates are also described in Figure 3.4.

Step-12. First, the carbon dioxide gas is injected with a manual regulator from the tank into the specimen from both the top and the bottom of the sample.

Step-13. Then, the back pressure of the CO₂ gas is gradually increased up to 2.3 MPa, while keeping the temperature constant at 10°C, as shown in Figure 3.4-(a). The cell pressure is maintained so that the effective confining pressure, which is defined as the difference between the applied cell pressure and the pore pressure measured by the pore pressure transducers, remains constant at 0.2 MPa.

Step-14. In order to thaw the frozen specimen and make the CO₂ gas dissolve enough into the water, it is left for half a day.
Step-15. After that, the temperature in the triaxial cell is cooled to 1°C, as shown in Figure 3.4-(b), where the carbon dioxide hydrates are stable, and the specimen environment is kept under constant pore pressure conditions. In this process, it is important to maintain a constant effective confining pressure to prevent premature sample failure or the overconsolidation of the specimen. If hydrates begin to form, pore gas is consumed for forming the hydrates and the pore gas pressure will drop suddenly. Thus, we use the accurate computer-controlled pore pressure intensifier illustrated in Figure 3.1-(3). When the pore gas pressure drops, due to the hydrate formation, a temperature change, and/or a leak in the system, the pore intensifier, via computerized pressure control, causes an upheaval of the piston into the cylinder to maintain the set pressure.

Step-16. In order to ensure the hydrate formation, the system is then left for at least 1 day.

![Figure 3.4 State paths for temperature and pressure on equilibrium curves of carbon dioxide during CO₂ hydrate forming process](image)

3.3.3 Water saturation and consolidation process

After generating carbon dioxide hydrates, the specimen is then saturated with distilled water. We use pure water for injection in the tests, although some CO₂-hydrates can dissolve in pure water. This is because water saturated with CO₂ gas may cause the formation of additional CO₂-hydrates more easily, and this makes the permeability in the pores rather low.
During the saturation, the pure water is passed from the bottom to the top of the specimen under a certain level of pressure gradient in order to replace the CO₂ gas with the pure water. After that, the back pressure (water pressure) and the cell pressure are increased to the target values of effective stress required for the consolidation process. At this stage, a Skempton’s B-test is performed to ensure the saturation (Skempton, 1954). Then, consolidation is carried out under a specified effective confining pressure.

### 3.3.4 Dissociation process

The dissociation of gas hydrates is brought on by increasing the temperature, namely, the heating method, while monitoring the changes in pore pressures and deformations of the specimen. Figure 3.5 demonstrates the pressure and temperature paths during the heating method. The initial temperature is around 1°C, and then the triaxial cell is heated up to 18°C. The dissociation tests are conducted under undrained conditions. After consolidation, all the valves connected to the specimen are closed.

Figure 3.5 State paths for temperature and pressure during dissociation process by heating method

### 3.3.5 Determination of hydrate saturation

Hydrate saturation is defined as Equation (3.1), and is calculated in the following two ways:
3.3 Testing Procedures

(A) the method using the water contents after dissociation

(B) the method using the volume of carbon dioxide gas collected after the dissociation test

3.3.5.1 Method (A): Water content method

In method (A), we assume that all the water in the specimen reacts with the carbon dioxide gas and forms CO$_2$-hydrates. The reaction equation of the CO$_2$-hydrates is described as follows:

$$\text{CO}_2 \cdot 5.75\text{H}_2\text{O}(\text{hydrate}) \Leftrightarrow \text{CO}_2(\text{gas}) + 5.75\text{H}_2\text{O}(\text{liquid})$$  \quad (3.2)

When the water content, after the dissociation test, is expressed by $w_{after} \ [%]$, the moles of water in the pores can be calculated by the following equation.

$$N_{water} = \frac{w_{after} m_s}{100 M_{water}} \ [\text{mol}]$$  \quad (3.3)

where $N_{water} \ [\text{mol}]$ indicates the moles of water determined from the water content, $m_s \ [\text{g}]$ is the mass of soil phase, and $M_{water} \ [\text{g/mol}]$ is the molar mass of water; we use 18.0 in this study. A unit mole of CO$_2$-hydrates produces 5.75 times the moles of water, namely,

$$N_{hydrate} = \frac{N_{water}}{5.75} \ [\text{mol}]$$  \quad (3.4)

Considering the molar mass of CO$_2$-hydrates $M_H$, the mass of CO$_2$-hydrates in the specimen becomes

$$\begin{cases} m^H = N_{hydrate} M_H \ [\text{g}] \\ M_H = 44 + 5.75 \times 18 = 147.5 \ [\text{g/mol}] \end{cases}$$  \quad (3.5)

Using Equation (3.5) and the density of carbon dioxide hydrates $\rho_{CO2}^H \ [\text{g/cm}^3]$, we obtain the
volume of the hydrates in the pores as follows

\[ V^H = \frac{m^H}{\rho_{CO2}^H} = \frac{N_{water} M_H}{5.75 \rho_{CO2}^H} \text{ [cm}^3\text{]} \]  \hspace{1cm} (3.6)  

\[ \rho_{CO2}^H = 1.12 \text{ [g/cm}^3\text{]} \]  \hspace{1cm} (3.7)  

in which the density of CO\textsubscript{2}-hydrates \( \rho_{CO2}^H \) is given by Udachin et al. (2001). The volume of the void can be described as follows:

\[ V^v = \frac{e}{1+e} \frac{\pi HD^2}{4} \text{ [cm}^3\text{]} \]  \hspace{1cm} (3.8)  

where \( e \) is the void ratio, and \( H \) [cm] and \( D \) [cm] are the height and the diameter of the specimen, respectively.

Considering Equations (3.1), (3.6) and (3.8), we have the hydrate saturation of the specimens.

Method (A) is useful only for the specimens without the water saturation process, because the water content may change if water is injected into the specimens after the formation of the CO\textsubscript{2}-hydrates.

\subsection*{3.3.5.2 Method (B): Collected gas method}

After the dissociation test, the CO\textsubscript{2} gas produced by dissociation is collected in the lower pore pressure pump, as shown in Figure 3.1-(4). Before collecting the gas, the cylinder is empty, and then we open the valves close to the specimen to allow the CO\textsubscript{2} gas to flow into the cylinder. Figure 3.6 illustrates the schematic diagram of the paths used in the gas collection process. Red lines indicate the path filled with CO\textsubscript{2} gas. The piston of the pump drops down in order to maintain the pressure in the cylinder at atmospheric pressure, monitoring the displacements of the piston. The displacements of the piston can be converted to volume changes. In collecting the gas, a small water flow is applied to try to push the remaining gas out of the specimen. After that, we obtain the volume of CO\textsubscript{2} gas at atmospheric pressure, at which point the temperature in the cylinder is 20°C, which is the same as that of the experimental room. The collected gas volume is then recalculated at standard conditions (0°C, 101.325 kPa) using Charles’s law,
3.3 Testing Procedures

namely,

\[ V'_{st} = \frac{T_{st}}{T_m} V_m \]  

\[ \begin{align*}
V_m &: \text{Measured volume of CO}_2 \text{ gas [cm}^3]\text{]} \\
T_m &: \text{Temperature of experimental room, } 273 + 20 = 293[\text{K}] \\
V_{st} &: \text{CO}_2 \text{ gas volume at standard conditions [cm}^3]\text{]} \\
T_{st} &: \text{Temperature at standard conditions, } 273[\text{K}] 
\end{align*} \]

Since one cubic meter of gas hydrates decomposes at the standard temperature and pressure into 164.6 m\(^3\) of gas (Kvenvolden, 2000), the volume of the hydrates in the pores, \( V^H \) [cm\(^3\)], can be described as

\[ V^H = \frac{V_{st}}{164.6} \text{[cm}^3]\text{]} \]  

In the same way as for method (A), the hydrate saturation is calculated from Equations (3.8) and (3.10). The method is usable for the specimens saturated with water.

One negative aspect of both method (A) and method (B) is that the resolution of CO\(_2\) gas into the pore water is not considered, for the sake of simplicity. It will be a future task to consider the solubility of CO\(_2\) gas into water for measuring the precise values of the hydrate saturation.
3.4 Testing Conditions

The experimental conditions are summarized in Table 3.2 and Table 3.3. We have conducted eight tests under different pore pressures and hydrate saturations. Case-1 is the specimen without CO₂-hydrates, that is, the water-saturated specimen. In this case, the process of the specimen preparation is the same as the others; however, CO₂ gas is not passed through the specimen to prevent CO₂-hydrates from forming during the cooling process. Case-2 and Case-3 are the tests for the CO₂-hydrate-containing samples, while water is not passed through the specimens after forming hydrates. The hydrate saturations of Case-2 and Case-3 are determined by the water contents after the dissociation process, that is, Method (A), and the water contents become 5.0% in Case-2 and 5.1% in Case-3, respectively. Case-4 ~ Case-8 are the cases where the specimens are saturated with water. Thus, the collected gas method, Method (B), is used to determine the hydrate saturation. Table 3.4 shows B-values measured after the water saturation process and the CO₂ gas volume collected after the dissociation tests at standard conditions. The hydrate saturations shown in Table 3.3 are determined from the collected gas volume as described in Table 3.4. The initial water content is determined from the target hydrate saturation.
that is, 55%. The hydrate saturation in Case-4 ~ Case-8 varies among the cases, although the initial water contents and the experimental procedures are the same among the cases. It is likely that the water saturation process affects the results of the hydrate saturation. In other words, the remaining CO₂ gas in the specimen reacts with the injected water and forms the additional CO₂-hydrates, or the existing CO₂-hydrates dissolve into the water.

The pore pressure during the formation process is 2.0 MPa in Case-1, whereas the pore pressure in the other cases is the same at 2.3 MPa. Then, back pressure is applied up to 4.0 MPa in Case-3 ~ Case-7, and up to 6.0 MPa in Case-8. By increasing the pore water pressure, the CO₂ gas remaining in pores may dissolve into the water. In Case-2, the back pressure is not applied after forming hydrates, and the dissociation process is conducted at the same pore pressure as the formation process.

The effective confining pressure, which is defined as the difference between the cell pressure and the pore pressure, is set at 1.0 MPa in Case-1 and Case-2, and in the other cases, it is set at 3.0 MPa. It is assumed that the pore pressure measured by the pore pressure transducers is the pore water pressure after water injection. The triaxial system is not able to measure pore water pressure and pore gas pressure independently. Consequently, suction cannot be detected if the specimens are under unsaturated conditions.

During the dissociation test, the cell pressure is kept at constant, and the effective confining pressure may change with increases or decreases in the pore pressure due to the hydrate dissociation.

### Table 3.2 Test conditions for initial state and formation process

<table>
<thead>
<tr>
<th>Case-No.</th>
<th>Initial water content</th>
<th>Initial void ratio</th>
<th>Pore pressure during formation</th>
<th>Temperature for formation</th>
<th>State of specimen</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( w_{\text{in}} ) [%]</td>
<td>( e_0 )</td>
<td>[MPa]</td>
<td>[°C]</td>
<td></td>
</tr>
<tr>
<td>Case-1</td>
<td>12.0</td>
<td>0.69</td>
<td>2.0</td>
<td>1.0</td>
<td>Without hydrates</td>
</tr>
<tr>
<td>Case-2</td>
<td>15.3 (5.0)</td>
<td>0.69</td>
<td>2.3</td>
<td>1.0</td>
<td>Unsaturated</td>
</tr>
<tr>
<td>Case-3</td>
<td>15.0 (5.1)</td>
<td>0.67</td>
<td>2.3</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Case-4</td>
<td>15.0</td>
<td>0.75</td>
<td>2.3</td>
<td>1.0</td>
<td>Water saturated</td>
</tr>
<tr>
<td>Case-5</td>
<td>11.0</td>
<td>0.75</td>
<td>2.3</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Case-6</td>
<td>15.0</td>
<td>0.75</td>
<td>2.3</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Case-7</td>
<td>15.0</td>
<td>0.79</td>
<td>2.3</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Case-8</td>
<td>15.2</td>
<td>0.76</td>
<td>2.3</td>
<td>1.0</td>
<td></td>
</tr>
</tbody>
</table>
3.5 Experimental Results

3.5.1 Results of formation process

Figure 3.7-(a)–(h) indicates the time profiles of the pore pressure in each case during the formation process. The pore pressure should be maintained at a constant pressure, that is, at the initial one. If hydrates begin to form, the CO$_2$ gas in the pores will be consumed for forming hydrates and the pore gas pressure will drop suddenly. Thus, we use the computer-controlled pore pressure intensifier which is illustrated in Figure 3.1-(3). By calculating the volume change in the cylinder, the volume of CO$_2$ gas consumed for hydrate formation can be obtained.
3.5 Experimental Results

Its time profiles are illustrated in Figure 3.8-(a) and (b).

The initial pore pressure is 2.0 MPa in Case-1, and it is 2.3 MPa in the other cases. The black red lines indicate the value of the pore pressure transducer in the lower and upper parts of the specimen, respectively. The pore pressure in Case-1 was kept constant at the initial value. Only a small injection of the piston into the specimen was observed, as shown in Figure 3.8-(a) and (b). This was because the cooling of the triaxial cell caused the contraction of the remaining gas in the pores or helped the gas dissolve into the pore water. Hence, the pore pressure was likely to decrease, and the piston injected more CO₂ gas into the specimen. The pore pressure in Case-2 was also kept constant, whereas the volume of injected CO₂ gas suddenly increased after 3.0 hours, as shown in Figure 3.8-(a) and (b). It was evidence of the consumption of CO₂ gas for forming the hydrates, and the piston moved up in order to maintain a constant pore pressure. The volume change of the piston settled after 5 hours; however, it began to increase again at a constant rate.

It can be seen that the pore pressure dropped drastically after 2 hours in Case-3, Case-5, and Case-6, as shown in Figure 3.7-(c), (e), and (f), respectively, despite of the rapid injection of the piston. It is likely that the rate of the pore pressure reduction due to the hydrate formation was larger than the rate of the CO₂ gas injection. The pore pressure recovered to 2.3 MPa after 5 hours by the piston maintaining the injection of CO₂ gas in Case-3 and Case-5. The total volume of the injected CO₂ gas was calculated at 60 cm³ in Case-3 and 140 cm³ in Case-5, and the hydrate saturation was 25.4% and 39.1%, respectively. The higher the injected gas volume becomes, the more hydrates can be formed in the pores. In Case-6, on the other hand, the pore pressure continued to decrease despite the consecutive injection of CO₂ gas. It was difficult to keep the constant pressure because of the high rate of consumption. After 17 hours, the piston of the pore pressure intensifier reached the limit, and the piston stopped moving up. Consequently, it became impossible to supply CO₂ gas into the specimen any more. This resulted in a significant decrease in pore pressure, as shown in Figure 3.7-(f). The hydrate saturation in Case-6 is 42.4 %, which is the highest among all cases.

The volume of injected CO₂ gas in Case-8 became higher than that of the other cases until 6 hours had passed. A larger amount of CO₂ gas was consumed for the hydrate formation. After that, the piston became calm and settled at about 70 cm³, while the continuous injection can be found in Case-6 and Case-7, where the hydrate saturation is larger than in Case-8.

Figure 3.9 and Figure 3.10 illustrate the time profiles for the pore pressure and axial strain in Case-7 and Case-8, respectively. The axial strain is positive in compression and negative in tension. The load cell was controlled to maintain isotropic conditions.
In Case-7, the compressive axial strain can be seen in the first hour, because of the decrease in pore pressure due to the shrinkage of CO$_2$ gas or the dissolution of gas into the pore water. The axial strain increased in the direction of tension after 2 hours, although the pore pressure continued decreasing. It may have been caused by a difference in density between water and the CO$_2$-hydrates. A unit volume of CO$_2$-hydrates can produce about 0.8 times the volume of water; namely, when the water changes into hydrates, the volume will increase.

The relationship between the pore pressure and the axial strain in Case-8 is similar to that in Case-7. The negative axial strain, that is, tensile strain, was found, while the pore pressure was constant at 2.3 MPa, as shown in Figure 3.10. It reached to -0.25% after 2 hours, and then it settled after 4 hours. The injection of CO$_2$ gas also became calm at the same time when the change in the axial strain became small, as shown in Figure 3.8-(a) or (b).

![Figure 3.7-(a)–(d) Time profiles of the pore pressure of each case during formation process (Case-1 – Case-4)](image-url)
3.5 Experimental Results

Figure 3.7-(e)–(h) Time profiles of the pore pressure of each case during formation process (Case-5 ~ Case-8)

Figure 3.8-(a) Time profiles of the volume injected CO$_2$ gas

(a) overall view
3.5 Experimental Results

Figure 3.8-(b) Time profiles of the volume injected CO$_2$ gas

(b) magnified view

Figure 3.9 Time profiles of the pore pressure and axial strain in Case-7
3.5 Experimental Results

3.5.2 Results of dissociation process

Figure 3.11-(a)–(h) shows the time profiles of the pore pressure during the dissociation process for each case, while the excess pore pressure for each case is illustrated in Figure 3.12. In Case-1, that contains no hydrates, little increase in the pore pressure was observed. The pore pressure increased from 2.0 MPa to 2.1 MPa. The pore air that dissolved in pore water might change into air bubbles due to the increase in temperature and cause the pore pressure to increase.

In the other cases, a drastic increase in the pore pressure was observed just before 1 hour had passed. The pore pressure was kept constant at the initial pore pressure until 1 hour from the beginning of heating. A small increase in pore pressure in the first hour may have been caused by the expansion of the remaining CO₂ gas or the gasification of the dissolved gas from the pore water induced by the rise in temperature. The maximum values of the pore pressure of each case and the final value are listed in Table 3.5. The pore pressure reached up to 3.9 MPa in Case-2, 6.0 MPa in Case-3, 5.6 MPa in Case-4, 6.9 MPa in Case-5, 10.0 MPa in Case-6, 6.9 MPa in Case-7, and 8.3 MPa in Case-8.

In Case-3–Case-8, the pore pressures dropped after reaching the peak value. In all cases, except for Case-4, the pore pressure begins to decline slowly after 2 or 3 hours, and then the pore pressure at the final state settled around 6.0 MPa in Case-3, Case-5, and Case-7, and eventually around 7.7 MPa in Case-8, as shown in Figure 3.11-(c), (e), (g), and (h). In Case-4,
3.5 Experimental Results

the pore pressure of the upper part of the specimen rises drastically, as well as in the other cases, and then, when it reaches 5.0 MPa, it suddenly drops and then increases again after 2 hours, as shown in Figure 3.11-(d). Finally, it settles at 5.6 MPa. In comparison to the hydrate saturation in Case-3, Case-7, and Case-8, the hydrate saturation in Case-4 was higher than in the other cases; however, the maximum value of the excess pore pressure was lower.

The change in the pore pressure in Case-6 is more drastic, as shown in Figure 3.11-(f) and Figure 3.12. The gradient of the pore pressure with respect to time in Case-6 is larger than in the other cases because of the high hydrate saturation, namely, 42.4%. When the pore pressure reaches 7 MPa, after about 1.5 hours, the effective confining pressure is about to become zero. Thus, the cell pressure increases to 10.0 MPa in order to prevent the specimen or the membrane from being punctured due to the high pore pressure. The pore pressure becomes larger with an increase in the cell pressure and finally reaches 10.0 MPa. Then, the pore pressure drops suddenly and settles around 6.0 MPa, as well as in the other cases. The reason why the pore pressure becomes about 6.0 MPa at the final state in many cases will be discussed later.

Table 3.5 Maximum values of pore pressure and excess pore pressure, and the pore pressure at final state of dissociation process

<table>
<thead>
<tr>
<th>Case No. (( S^H_r ))</th>
<th>Pore pressure before heating [MPa]</th>
<th>Pore pressure (Max.) [MPa]</th>
<th>Excess pore pressure (Max.) [MPa]</th>
<th>Pore pressure at final state [MPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case-1 (0.0%)</td>
<td>2.0</td>
<td>2.1</td>
<td>0.1</td>
<td>2.1</td>
</tr>
<tr>
<td>Case-2 (22.8%)</td>
<td>2.3</td>
<td>3.9</td>
<td>1.6</td>
<td>3.7</td>
</tr>
<tr>
<td>Case-3 (25.4%)</td>
<td>3.0</td>
<td>6.0</td>
<td>3.0</td>
<td>5.7</td>
</tr>
<tr>
<td>Case-4 (28.5%)</td>
<td>4.0</td>
<td>5.6</td>
<td>1.6</td>
<td>5.6</td>
</tr>
<tr>
<td>Case-5 (39.1%)</td>
<td>4.0</td>
<td>6.9</td>
<td>2.9</td>
<td>5.9</td>
</tr>
<tr>
<td>Case-6 (42.4%)</td>
<td>4.0</td>
<td>10.0</td>
<td>6.0</td>
<td>5.7</td>
</tr>
<tr>
<td>Case-7 (15.4%)</td>
<td>4.0</td>
<td>6.9</td>
<td>2.9</td>
<td>6.0</td>
</tr>
<tr>
<td>Case-8 (32.1%)</td>
<td>6.0</td>
<td>8.3</td>
<td>2.3</td>
<td>7.7</td>
</tr>
</tbody>
</table>
3.5 Experimental Results

Figure 3.11-(a)–(f) Time profiles of pore pressure during dissociation of each case

(a) Case-1, $S_H^0=0.0\%$ (without hydrate)
(b) Case2, $S_H=22.8\%$
(c) Case-3, $S_H=25.4\%$
(d) Case-4, $S_H=28.5\%$
(e) Case-5, $S_H=39.1\%$
(f) Case-6, $S_H=42.4\%$
Figure 3.11-(g)~(h) Time profiles of pore pressure during dissociation of each case

Figure 3.12 Time profiles of the excess pore pressure during dissociation of all cases

Time profiles of the effective confining pressure for each case are plotted in Figure 3.13-(a) ~ (h) and Figure 3.14, where the effective confining pressure is defined as the difference between the cell pressure and the pore pressure. In Case-1, which contains no hydrates, the cell pressure is kept at 2.1 MPa during the heating process; therefore, the effective confining pressure is 0.1 MPa. The effective confining pressure does not change so largely, although a small change due to the rising temperature is observed as well as pore pressure. In Case-2, the initial effective confining pressure is 1.0 MPa. When hydrate dissociation starts, the effective confining pressure decreases due to the drastic increase in pore pressure. After about 1 hour, the effective confining pressure becomes approximately zero, and then the cell pressure is increased in order to prevent
the specimen and the membrane from being punctured. At the same time, the effective confining pressure is recovered to 0.7 MPa. The results of Case-2 indicate that it is easy for the pore pressure produced by hydrate dissociation to exceed 1.0 MPa of the effective confining pressure under undrained conditions. Consequently, the initial effective confining pressure in the other cases is set at 3.0 MPa, so that the pore pressure will not exceed the initial effective confining pressure.

The effective confining pressure in Case-3 drops drastically just before 1 hour has passed due to the hydrate dissociation, as shown in Figure 3.13-(c). After 3 hours, it decreases to approximately 0.0 MPa, and this state continues until 4 hours have passed. After that, the effective confining pressure begins to increase. This is because the pore pressure gradually decreases, as previously mentioned in Figure 3.11-(c) and Figure 3.12. The behavior of the effective confining pressure in the other cases is basically the same as that in Case-3. The effective confining pressure begins to decrease rapidly when the hydrates dissociate and excess pore pressure is produced. The effective confining pressure is reduced to 1.4 MPa in Case-4, 0.1 MPa in Case-5 and Case-7, and 0.7 MPa in Case-8. After that, the effective confining pressure gradually moves towards recovery, as shown in Figure 3.13-(c)~(h). In Case-6, however, the change in effective confining pressure is more abrupt than in the other cases, as shown in Figure 3.13-(f). The reduction rate of the effective confining pressure in Case-6 is quite a bit larger than in the other cases, and it reaches 0.0 MPa before 2 hours have passed, which is earlier than in the other cases. This is because the hydrate saturation in Case-6 is higher than that in the other cases and more and more CO₂ gas is produced due to the dissociation. Due to the risk of puncturing the membrane or an explosion of the specimen, the cell pressure is raised to 10.0 MPa. Then, the pore pressure also increases to 10.0 MPa with an increase in the cell pressure, as described in Figure 3.11-(f). When it reaches to 10.0 MPa, it increases suddenly, because the pore pressure decreases drastically, which is can be seen in Figure 3.11-(f).

The results of the effective confining pressure indicate that the dissociation of hydrates causes a significant reduction in the effective stress. In a real situation, when methane hydrates dissociate around an area where the permeability is quite low locally, it is possible that the strength of the sediments may decrease and that submarine slope instabilities may be triggered.
Figure 3.13-(a)–(f) Time profiles of effective confining pressure during dissociation of each case
In order to explain why the pore pressure gradually decreases after the peak and finally settles around 5.6 ~ 6.0 MPa, and why there is a sequential recovery of the effective confining pressure, the relationships between temperature and pore pressure are illustrated in Figure 3.15-(c)–(h) and Figure 3.16. In Case-1 and Case-2, instruments which gather temperature data have not been equipped; hence, it is impossible to draw the temperature-pore pressure relationships for Case-1 and Case-2.

The dashed line indicates the liquefied boundary of CO₂ gas. CO₂ gas exists as gas under the dashed line, while the gas changes to the liquid phase above the boundary. In Case-3, the initial
3.5 Experimental Results

state of the heating process is around the point of (1.0°C, 3.0 MPa), that is, below the liquid boundary. The pore pressure is kept constant for a while, from the beginning of the heating, and then the pore pressure begins to increase when the temperature becomes 7.0°C. The increase in pore pressure is produced by the hydrate dissociation. Then, the pore pressure exceeds the liquefied boundary at the time when the temperature reaches 10.0°C. The pore pressure continues increasing with an increase in temperature, although the increasing rate of the pore pressure decreases gradually after 10.0°C. In Case-3, the pore pressure drops from 6.0 MPa to 5.7 MPa, which is consistent with that of the boundary between liquid CO₂ and the gas phase at the temperature of 18.0°C, as shown in Figure 3.15-(c). It is possible that the reduction in pore pressure after the peak, as previously described, may be caused by the liquefaction of CO₂ gas produced by the hydrate dissociation.

In the other cases, that is, Case-4, Case-5, Case-6, and Case-7, on the other hand, the initial state is above the liquefied boundary. The first increase in pore pressure occurs at the point where the temperature-pore pressure path and the liquefied boundary intersect. The behavior is the same among Case-4 ~ Case-7. This is because the remaining CO₂ gas exists as the liquid phase at the initial state, and it changes into the gas phase again due to the rise in temperature, although enough volume of water was passed through the specimen during the water saturation process after the hydrate formation. The second increase in pore pressure occurs after the point of 10.0°C, and it may be caused by the hydrate dissociation. The second buildup of pore pressure was obvious, especially in Case-4, Case-5, and Case-6, whereas it was subtle in Case-7.

In Case-4, the temperature-pore pressure path moves along with the line of liquefied boundary after the first increases in pore pressure, and then it gradually rises until reaching the final state. The reason why the peak value for the excess pore pressure in Case-4 is lower than that in Case-3 and Case-7, even though the hydrate saturation in Case-4 is higher than in the others cases, can be explained as follows. The liquefaction of CO₂ gas and the gas production induced by the dissociation may occur simultaneously, especially in Case-4. And the effect of the liquefaction is stronger than that of the dissociation. On the other hand, the effect of the dissociation is stronger than that of liquefaction in the other cases. Thus, the behavior of the excess pore pressure during the heating process is different among those cases, although the conditions of the temperature and pore pressure at the final state are the same.

Figure 3.15-(f) illustrates the temperature and pore pressure path in Case-8. The initial point is at (1.5°C, 6.0 MPa). The reason why the initial pore pressure is set at 6.0 MPa, which is larger than in the other cases, is to remove the effect of the gasification of CO₂ gas from the liquid phase. In Case-4 ~ Case-7, the pore pressure begins to increase before it reaches the hydrate
boundary, because the temperature-pore pressure path first intersects with the liquefied boundary, and the CO$_2$ gas remaining in the pores as liquid may change into the gas phase. This will make the initiation of the dissociation unclear, and then it becomes difficult to distinguish which is the main reason for the significant increase in pore pressure, gasification or hydrate dissociation. The pore pressure in Case-8 is kept constant until the temperature becomes 10.0°C, and then it starts to increase drastically, as shown in Figure 3.15-(g) and (h). This large buildup of pore pressure is mainly induced by the hydrate dissociation. The maximum value of excess pore pressure becomes approximately 8.3 MPa, and the effective confining pressure is then reduced to 0.7 MPa by the dissociation. Of course, there is a possibility that the CO$_2$ may be produced as the liquid phase, when the temperature-pore pressure conditions are above the liquefied boundary. However, the behavior of the increasing pore pressure in Case-8 is very similar to that in Case-3, and the dissociation process in Case-3 should start under the liquefied boundary. Therefore, the buildup of pore pressure in Case-8 may be caused by the gas production. The reduction in pore pressure after the peak is also evidence that the increase in pore pressure should be caused by CO$_2$ gas and not by CO$_2$ liquid. If it was caused by CO$_2$ liquid, the pore pressure would increase gradually up to the final state without a drop in pressure like in Case-4, as shown in Figure 3.15-(d) and Figure 3.16.

These results indicate that hydrate dissociation will initially produce CO$_2$ gas even if the conditions of temperature and pore pressure exist in the liquefied region, and the gas production will lead to the significant decrease in effective stress.

In the experiments, the effective confining pressure recovered due to the liquefaction of CO$_2$ gas. On the other hand, the liquefaction of methane gas may occur at rather low temperatures, and this means that the excess pore pressure produced by the methane hydrate dissociation will be maintained at high pressure levels. The conditions may become more dangerous than those of the CO$_2$ gas or the CO$_2$-hydrates.
Figure 3.15-(c)–(h) Temperature-pore pressure path during dissociation of each case
The relationships between the excess pore pressure and the axial strain in Case-7 and Case-8 are shown in Figure 3.17. The axial strain is positive in compression and negative in expansion. During dissociation tests, the load cell was controlled to maintain isotropic conditions. In both cases, the expansive strain progressed with the increase in excess pore pressure. The change in axial strain was small until the beginning of the pore pressure buildup. The tensile strain was observed with an increase in pore pressure, and when the pore pressure became the maximum value, the axial strain also reached the peak. The tensile strain reached -0.45% in both Case-7 and Case-8. The results indicate that hydrate dissociation may lead to the uplift of the sediment due to the large increase in pore pressure with the significant decrease in effective stress if the permeability of the sediments is low.
In this chapter, a series of formation and dissociation tests has been performed using a temperature-controlled, high-pressure triaxial apparatus in order to characterize the mechanical behavior of gas hydrate sediments subjected to hydrate dissociation. The apparatus that has been developed enables the control of low temperature and high pressure environments for the formation of gas hydrate-containing sand specimens and the performance of sequential dissociation tests. In the experiments, CO$_2$ gas was chosen as the guest gas for its safety and easy handling. In both the formation and the dissociation processes, the load cell was controlled to maintain the isotropic conditions, and the axial strain of the specimen was measured. The dissociation tests were conducted using the thermal recovery method under undrained conditions by assuming that the local permeability of the sediments was quite low. The obtained conclusions are summarized as follows:

1. In forming CO$_2$-hydrates in specimens, a drastic drop in pore pressure and the simultaneous consumption of CO$_2$ gas were observed. This is the evidence of the formation of CO$_2$-hydrates.

2. The installed pore pressure intensifier was able to control the pore pressure constant in some cases; however, it became more difficult to maintain the pore pressure constant in the
3.6 Summary

case of high hydrate saturation, because the consumption of CO$_2$ gas became larger and the reduction of the pore pressure became more drastic.

3. Tensile axial strain was found in the formation process, while the pore pressure was kept constant. The difference in density between water and hydrates produced such a tensile strain; namely, the water change in the pores changed into hydrates with the expansion of the volume.

4. The results of the dissociation tests indicate that hydrate dissociation can cause significant increases in pore pressure and can result in significant decreases in effective confining pressure under undrained conditions. In real situations, when MHs dissociate inside the area where the local permeability is quite low, the possibility of the sediments being unstable becomes higher.

5. CO$_2$ gas was produced by both the hydrate dissociation and the gasification from the liquid CO$_2$ remaining in the pores, and then the pore pressure increased to a very high pressure level. Since it increased over the liquid boundary, the pore pressure decreased to the liquefaction boundary. The gas production due to the dissociation and the liquefaction of CO$_2$ gas may occur simultaneously, and the maximum value of the excess pore pressure depends on the balance of these two reactions.

6. The relationship between dissociation and deformation was found, and a tensile axial strain was observed with a decrease in the effective confining pressure due to dissociation. However, it was not possible to measure the volumetric strain because of a leak in the inner cell. It will be a future task to characterize the dissociation-deformation relationships.
3.6 Summary
Chapter 4

NUMERICAL SIMULATION OF DISSOCIATION TESTS BY A CHEMO-THERMO-MECHANICALLY COUPLED METHOD

4.1 Introduction

A series of dissociation tests using the temperature-controlled, high-pressure triaxial apparatus was reported in the previous chapter. In this chapter, the results are numerically simulated by the three-dimensional finite element method using an elasto-viscoplastic constitutive equation and the finite element formulation introduced in Chapter 2. The experimental results for Case-7 and Case-8 are chosen in order to compare the dissociation deformation behavior with that of the simulation subjected to hydrate dissociation, because the data for the axial strain was measured in those cases. In addition to the comparative study, the numerical simulation can give the dissociation-deformation behavior in more detail, such as the distributions of volumetric strain, the degree of the CO₂-hydrate dissociated area, and the effective stress inside the specimen.
4.2 3-Dimensional Finite Element Analysis of Dissociation Tests for Carbon Dioxide Hydrate-Containing Sand Specimen

The finite element formulations used in this chapter are the same as those introduced in Chapter 2. The governing equations are equilibrium equations for the solid-fluid mixture, continuity equations for pore water and pore gas, the conservation law of heat transfer, and the dissociation rate of the hydrates. The model expressed in Chapter 2 is developed for MH-bearing soils; however, we assume that the mechanical properties of CO₂-hydrate-bearing soils are similar to those of MH-bearing soils, as some researchers have mentioned (e.g., Espinoza & Santamarina, 2011; Hyodo et al., 2014a; Miyazaki et al., 2013). Since there is not enough data on CO₂-hydrates, the equations for the dissociation rate of CO₂-hydrates and dissociation heat are assumed to be the same as those for MHs, that is, Equation (2.247) and Equation (2.220), although it is preferable to use the one for carbon dioxide. Based on the finite deformation theory, an updated Lagrangian method with the objective Jaumann rate of Cauchy stress is used for the weak form of the rate type of the equilibrium equations. The finite element code is formulated in a three-dimensional setting, which was developed by Fukuda (2011) based on the three-dimensional finite element analysis method for unsaturated soils (Oka et al., 2008a). The element types used in the three-dimensional analysis are shown in Figure 4.1. A twenty-node isoparametric element, with a reduced Gaussian (2×2×2) integration for the soil skeleton, and an eight-node isoparametric element, with a full (2×2×2) integration for the pore fluids and the temperature, are used.

4.2.1 Finite element mesh and boundary conditions

Figure 4.2 shows the finite element meshes and the boundary conditions which are set up according to the same boundary conditions as those in the experiments. The mesh has a total of 160 elements, that is, \( x \times y \times z = 4 \times 4 \times 10 \), and 965 nodes. In the experiments, the dissociation tests were performed under undrained conditions for both water and gas. Thus, all the boundaries are assumed to be impermeable in the simulation, while the transport of pore water and pore gas between the elements is allowed. As for the top surface, the deviator stress was kept constant at isotropic pressure and the axial strain was measured in the experiments. Thus, the top boundary deforms in the z-direction, and deformation is not allowed in the x and y-directions due to contact with the top cap. All the deformations at the bottom surface are fixed.

116
in all directions. The dissociation tests were carried out by the thermal recovery method by increasing the temperature of the cell fluid from 1°C to 18°C. In the simulation, the temperature of the top, bottom, and side surfaces should be increased from 1°C to 18°C in 2.5 hours, which are the same conditions as in the experiments. A conceptual diagram of the increasing temperature is illustrated in Figure 4.3.

Figure 4.1 Isoparametric elements used in the three-dimensional finite element method

Figure 4.2 Finite element mesh and boundary conditions for the simulation
4.2 3-Dimensional Finite Element Analysis of Dissociation Tests for Carbon Dioxide Hydrate-Containing Sand Specimen

Figure 4.3 Conceptual diagram of conditions of the increasing temperature

4.2.2 Simulation cases and material parameters

The target results of the experiments are Case-7 and Case-8, as shown in Chapter 3, and the material parameters for the initial state of the simulation are listed in Table 4.1. The initial parameters are basically determined from the experimental results. As for the initial water saturation, it is set to be 0.9. The specimen is unsaturated assuming that there is a possibility of CO₂ gas remaining in the pores, and the initial pore gas pressure is assumed to be the same as the initial pore water pressure, 4.0 MPa in Case-7 and 6.0 MPa in Case-8. Table 4.2 shows the material parameters required for the constitutive equations of gas-hydrate bearing soils introduced in Chapter 2. The parameters are obtained from the results of parametric studies performed by Iwai (2010) on triaxial compression tests of Toyoura sand under undrained conditions presented by Takyu (2003) as shown in Figure 4.4-(a) and (b). The thermal properties of water, CO₂ hydrates, and CO₂ gas are listed in Table 4.3. The heat capacity of CO₂-hydrate \( c_{CO₂}^H \) is estimated from the results presented by Handa (1986), and the heat conductivity of CO₂-hydrate \( \lambda_{CO₂}^H \) is set based on both the experimental and analytical results reported by Rosenbaum et al. (2007). The parameters required for the soil-water characteristic curve (SWCC) are listed in Table 4.4, and Figure 4.5 illustrates the shape of SWCC. The total time of the simulation is 6 hours.
### Table 4.1 Material parameters for the initial conditions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Case-7</th>
<th>Case-8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial void ratio $e_0$</td>
<td>0.79</td>
<td>0.76</td>
</tr>
<tr>
<td>Initial hydrate saturation [%]</td>
<td>$S_r^H$</td>
<td>15.4</td>
</tr>
<tr>
<td>Initial water saturation $s_r$</td>
<td>0.9</td>
<td></td>
</tr>
<tr>
<td>Initial average pore pressure [MPa] $P_{ini}$</td>
<td>4.0</td>
<td>6.0</td>
</tr>
<tr>
<td>Initial mean skeleton stress [MPa] $\sigma_{ini}'$</td>
<td>3.0</td>
<td></td>
</tr>
<tr>
<td>Initial temperature [$^\circ$C] $\theta_{ini}$</td>
<td>1.0</td>
<td></td>
</tr>
</tbody>
</table>

### Table 4.2 Material parameters for the constitutive equations

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Case-7</th>
<th>Case-8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Normalized elastic shear modulus $G_0/\sigma_{m0}'$</td>
<td>2023.6</td>
<td></td>
</tr>
<tr>
<td>Compression index $\lambda$</td>
<td>0.0112</td>
<td></td>
</tr>
<tr>
<td>Swelling index $\kappa$</td>
<td>0.0026</td>
<td></td>
</tr>
<tr>
<td>Compression yield stress [MPa] $\sigma_{mbi}'$</td>
<td>3.0</td>
<td></td>
</tr>
<tr>
<td>Stress ration at critical state $M_m$</td>
<td>1.13</td>
<td></td>
</tr>
<tr>
<td>Viscoplastic parameter $m'$</td>
<td>29.0</td>
<td></td>
</tr>
<tr>
<td>Viscoplastic parameter [1/s] $C_o$</td>
<td>1.0×10⁻⁹</td>
<td></td>
</tr>
<tr>
<td>Structural parameter $\sigma_{muf}'/\sigma_{mau}'$</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Structural parameter $\beta$</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Hardening parameter $B^*$</td>
<td>50.0</td>
<td></td>
</tr>
<tr>
<td>Thermo viscoplastic parameter $\alpha$</td>
<td>0.15</td>
<td></td>
</tr>
<tr>
<td>Suction parameter (1) $S_I$</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>Suction parameter (2) $s_d$</td>
<td>0.25</td>
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<tr>
<td>Suction parameter (3) [kPa] $P_i^C$</td>
<td>100</td>
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<tr>
<td>CO₂-hydrate parameter (1) $n_m$</td>
<td>0.6</td>
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<tr>
<td>CO₂-hydrate parameter (2) $n_d$</td>
<td>0.75</td>
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<tr>
<td>CO₂-hydrate parameter (3) $S_{ri}^H$</td>
<td>0.154</td>
<td>0.321</td>
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<tr>
<td>Coefficient of water permeability [m/s] $k_w$</td>
<td>1.0×10⁻⁴</td>
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<tr>
<td>Coefficient of gas permeability [m/s] $k_g$</td>
<td>1.0×10⁻³</td>
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</tr>
<tr>
<td>Permeability reduction parameter $N$</td>
<td>7</td>
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Table 4.3 Material parameters for the thermal properties

<table>
<thead>
<tr>
<th>Case-7, and 8</th>
<th>Density of water [t/m³] ( \rho^W )</th>
<th>1.0</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Specific heat capacity of water [kJ/t⋅K] ( c^W )</td>
<td>4200</td>
</tr>
<tr>
<td></td>
<td>Heat conductivity of water [kW/m⋅K] ( \lambda^W )</td>
<td>( 0.58 \times 10^{-3} )</td>
</tr>
<tr>
<td></td>
<td>Molar mass of water [kg/kmol] ( M^W )</td>
<td>18.0</td>
</tr>
<tr>
<td></td>
<td>Density of CO₂-hydrate [t/m³] ( \rho^H_{CO2} )</td>
<td>1.12</td>
</tr>
<tr>
<td></td>
<td>Specific heat capacity of CO₂-hydrate [kJ/t⋅K] ( c^H_{CO2} )</td>
<td>2200</td>
</tr>
<tr>
<td></td>
<td>Heat conductivity of CO₂-hydrate [kW/m⋅K] ( \lambda^H_{CO2} )</td>
<td>( 0.68 \times 10^{-3} )</td>
</tr>
<tr>
<td></td>
<td>Molar mass of CO₂-hydrate [kg/kmol] ( M^H_{CO2} )</td>
<td>147.5</td>
</tr>
<tr>
<td></td>
<td>Specific heat capacity of CO₂-hydrate [kJ/t⋅K] ( c^G_{CO2} )</td>
<td>826</td>
</tr>
<tr>
<td></td>
<td>Heat conductivity of CO₂-hydrate [kW/m⋅K] ( \lambda^G_{CO2} )</td>
<td>( 1.45 \times 10^{-5} )</td>
</tr>
<tr>
<td></td>
<td>Molar mass of CO₂-hydrate [kg/kmol] ( M^G_{CO2} )</td>
<td>44.0</td>
</tr>
<tr>
<td></td>
<td>Gas constant [kJ/kmol⋅K] ( R )</td>
<td>8.314</td>
</tr>
</tbody>
</table>

Table 4.4 Material parameters for the soil-water characteristic curve

<table>
<thead>
<tr>
<th></th>
<th>( s_{\text{max}} )</th>
<th>0.9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Minimum saturation ( s_{\text{min}} )</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Shape parameter for SWCC ( \alpha^C )</td>
<td>( 2.5 \times 10^{-3} )</td>
<td></td>
</tr>
<tr>
<td>Shape parameter for SWCC ( n' )</td>
<td>10.0</td>
<td></td>
</tr>
</tbody>
</table>

Figure 4.4-(a), (b) Results of the parametric study on undrained triaxial tests (Iwai, 2010), (a) effective stress path, (b) stress-strain relationship
4.3 Simulation Results

The dissociation tests using the thermal recovery method under undrained conditions were simulated. The boundary conditions for the temperature of the specimen are that it was increased from the top, the bottom, and side surfaces from 1°C to 18°C in 2.5 hours. Figure 4.6 and Figure 4.7 display the distributions of the increasing temperature during the simulations in Case-7 and Case-8, respectively. It can be seen that the temperature increased with progress in the heating of the specimen, and that the temperature in the center part of the specimen was lower than that of surrounding elements. That is because the heat transfer came from the boundary surfaces.

Figure 4.8 and Figure 4.9 show distributions of CO₂-hydrate remaining ratio in Case-7 and Case-8, respectively. The hydrate remaining ration is defined as a fraction of the current moles of the CO₂-hydrates \( N_H \) with respect to the initial moles \( N_{H0} \), namely,

\[
\text{CO}_2\text{-hydrate remaining ratio} = \frac{N_H}{N_{H0}}
\]  

(4.1)

The hydrates began to dissociate after about 1.5 hours in Case-7 and after 2 hours in Case-8, and the dissociation progressed from the top, the bottom, and the side surfaces according to the temperature increase as shown in Figure 4.6 and Figure 4.7. In Case-7, the dissociation
finished within 3 hours, whereas some hydrates were still remained in Case-8. The hydrate dissociation in Case-8 started about 20 or 30 minutes after that in Case-7, because the initial pore pressure of Case-8 was higher than that of Case-7, and it took more time to reach the hydrate equilibrium temperature. The difference might be produced also by the differences of the hydrate saturation between the two cases. The more hydrates that are inside the specimen at the initial state, the more time it will take for the dissociation reaction to come to an end. In Case-8, the hydrates dissociation was still going on even after 6 hours. It was probably because that the pore pressure reached at the equilibrium pressure at the temperature of 18°C, and the dissociation process almost stopped.

Figure 4.10 and Figure 4.11 illustrate the distributions of the pore gas pressure in Case-7 and Case-8, respectively. The pore gas pressure increased when the hydrate dissociation initiated. It seems that the differences were few among each element, namely, the distributions of the pore gas pressure were homogeneous. The pore gas pressure increased from 4.0 MPa to about 10.3 MPa in Case-7, and from 6.0 MPa to about 11.4 MPa in Case-8, respectively. The excess pore gas pressure became about 6.3 MPa in Case-7, and 5.4 MPa in Case-8. The excess pore gas pressure in Case-7 became higher than that in Case-8, even though the initial hydrate saturation was smaller. In Case-7, the hydrates dissociated completely within 3 hours, on the other hand, in Case-8, the hydrates which existed around the center part of the specimen remained even after 6 hours. Thus, the excess pore gas pressure in Case-7 became higher than that in Case-8 despite of the lower initial hydrate saturation. As for the pore water pressure, the behavior was similar to that of the pore gas pressure. The pore water pressure increased when the hydrate dissociation started, and the distributions were uniform in each element as shown in Figure 4.12 and Figure 4.13. The maximum values of the pore water pressure became less than the pore gas pressure in both Case-7 and Case-8, and the differences between the pore gas pressure and the pore water pressure were expressed in the distributions of the suction \( P^C = P^G - P^W \). The distributions of suction are illustrated in Figure 4.14 and Figure 4.15. Suction plays an important role in the mechanical properties of the soil. From Figure 4.14 and Figure 4.15, the initial suction was 0.0 kPa, which means the pore gas pressure was equal to the pore water pressure at the initial state. Then, the suction increased with time, even before the hydrate dissociation. This was because the gas density changed due to the temperature increase, and it led to a small buildup of the pore gas pressure. After that, a drastic increase in suction was observed; it was caused by the hydrate dissociation and the sequential rise in pore gas pressure. Finally, the suction reached 348 kPa in Case-7 and 346 kPa in Case-8, respectively.

The mean skeleton stress decreased as the pore gas pressure increased, as shown in Figure 4.16 and Figure 4.17. The mean skeleton stress in both cases dropped to approximately 0.0 MPa.
4.3 Simulation Results

in almost all elements. However, in contrast to the uniform distributions of the pore gas pressure, the mean skeleton stress became higher in the elements around the top and bottom surfaces than in the other elements. The maximum mean skeleton stress became more than 3MPa, that is, the initial values of the mean skeleton stress. The mean skeleton stress was controlled not to become negative, thus the mean skeleton stress was set to be \( 0.01 \times \sigma_{m0} \) at the time when it reached 0.0MPa in the simulations.

The distributions of viscoplastic volumetric strain \( \varepsilon_{vp} \) in both cases are illustrated in Figure 4.18 and Figure 4.19. The signs for the volumetric strain are the same as in the experimental results, that is, positive for compression and negative for expansion. Large expansive volumetric strain can be found, whereas little compressive strain was observed in either Case-7 or Case-8. The maximum value became -13.0% in Case-7 and -9.0% in Case-8. The larger expansive volumetric strain occurred in Case-7 than that in Case-8. This might be because that the excess pore pressure in Case-7 was bigger than Case-8. It can be said that the larger the excess pore pressure was produced due to the hydrate dissociation, the larger deformation will occur. The distributions of the volumetric strain were rather more non-uniform than the other results. The expansive strain localized especially around the center part of the specimen, where the mean skeleton stress became quite lower than that of the bottom and top surfaces, as shown in Figure 4.16 and Figure 4.17. This indicates that hydrate dissociation may produce the high pore fluids pressure and a sequential reduction in the mean skeleton stress under undrained conditions, as well as the larger expansive volumetric strain occurs in the area where the mean skeleton stress becomes lower.

Figure 4.20 and Figure 4.21 display accumulated viscoplastic deviator strain \( \gamma^{vp} \), which is defined by the following equation:

\[
\gamma^{vp} = \int_{0}^{t} \dot{\gamma}^{vp} \, dt, \quad \dot{\gamma}^{vp} = \sqrt{\dot{\varepsilon}_{ij}^{vp} \dot{\varepsilon}_{ij}^{vp}} \quad (4.2)
\]

where \( \dot{\varepsilon}_{ij}^{vp} \) is the viscoplastic deviator strain rate, and \( \dot{\gamma}^{vp} \) is its second invariant. Accumulated viscoplastic strain \( \gamma^{vp} \) indicates the cumulative quantities of shear strain. Hereafter, it will just be called deviator strain. The deviator strain behaved similarly to the viscoplastic
volumetric strain. It localized around the center part of the specimen; however, the strain level was smaller than the volumetric strain. The maximum values of the deviator strain became 1.1% in Case-7 and 0.8% in Case-8. Similar to the viscoplastic volumetric strain, the degree of the deviator strain became larger in Case-7 than that in Case-8. It may also depend on the degree of the excess pore pressure and the sequential reduction in the mean skeleton stress.

The skeleton stress paths in the target elements are plotted in Figure 4.22. The number of target elements is four, that is, element-1 (Layer-1, the bottom layer), element-83 (Layer-6), element-117 (Layer-8), and element-149 (Layer-10, the top layer), as shown in Figure 4.2. The initial state of the paths was under isotropic conditions with 3.0MPa of mean skeleton stress in each element. The mean skeleton stress began to decrease after the initiation of the hydrate dissociation in both Case-7 and Case-8. In elements-1, 117, and 149, the deviator stress increased simultaneously and gradually approached the failure line. On the other hand, in element-83, the deviator stress remained almost 0.0 MPa all the time, and only the mean skeleton stress moved toward the origin. All the skeleton stress paths reached to the failure line. In element-1, both the mean skeleton stress and the deviator stress increased along with the failure line after intersecting the failure line. In elements-117 and 149, the mean skeleton stress continued decreasing and the deviator stress increasing after the path reaching the failure line, then the mean skeleton stress became approximately 0.0 MPa. Finally, only the deviator stress increased, leaving the mean skeleton stress around 0.0 MPa. This could be the reason why the deviator strain localized around the center part of the specimen. The results indicate that the hydrate dissociation should lead to a significant reduction in the mean skeleton stress and will cause material failure.

Figure 4.23 – Figure 4.25 show time histories for the average pore pressure $P^p$, the mean skeleton stress $\sigma^i_m$, and the axial strain $\varepsilon_a$ of the selected elements, respectively, and the experimental data are also plotted on the graph in order to compare the stress and deformation behavior between the experiments and the simulations. Figure 4.23 displays the time profiles of the average pore pressure $P^p$ in the simulation, and the pore pressure measured in the experiments is plotted in the same figure. In the experiments, it was impossible to measure the pore water pressure and the pore gas pressure separately, and it was also impossible to calculate average pore pressure $P^p$; thus, only the pore pressure is used. The drastic increase in the average pore pressure was visible after 1.5 hours in Case-7 and after 1.8 hours in Case-8. The increasing behavior was very similar to that of the experimental results; the gradient of the pore pressure became steep after the dissociation, although the time when the average pore pressure began to increase was low-reproducible between the experimental results and the simulation one. The maximum values were also different between the experiments and the simulations. The
4.3 Simulation Results

maximum average pore pressure of each element was more than 10MPa in both Case-7 and Case-8, while these values were less than 10MPa in the experiments, namely, 6.9MPa in Case-7 and 8.3MPa in Case-8, respectively. In the experiments, the possibility of a CO$_2$ gas-changing liquid phase was pointed out in Chapter 3; however, the liquefaction of CO$_2$ was not considered and the gas was treated as ideal gas in the simulation. That is one of the reasons for the differences in the peak values between the experiments and the simulations.

The time profiles for the mean skeleton stress $\sigma_m'$ in the simulation and the effective confining pressure (E.C.P.) obtained from the experimental results moved similarly with time, as displayed in Figure 4.24, although it is difficult to compare those parameters simply because of a difference in their definitions. The mean skeleton stress is defined by Equation (2.22) and it requires the average pore pressure $P^F$ for the definition. However, the E.C.P is defined as just the differences between the cell pressure and the pore pressure, as described in Chapter 3. In both the experiments and the simulations, the E.C.P and the mean skeleton stress came to be approximately 0.0 MPa with the increase in the pore fluids pressure. In order to prevent the mean skeleton stress from becoming a negative value, the mean skeleton stress was set to be $0.01\times\sigma_{m0}'$ after it reached 0.0 MPa in the simulations; thus, it was kept at a constant value from the 2 hours. The E.C.P gradually recovered with the decreases in pore pressure due to the liquefaction of CO$_2$.

In order to compare the deformation behavior in the experiments and the simulations, the time profiles for the axial strain $\varepsilon_a$ are plotted in Figure 4.25. In the experiments, extensive axial strain was observed when the hydrates dissociated and it became about -0.45 % in both Case-7 and Case-8. The axial strain in the simulations also increased in an extensive direction after the hydrate dissociation, which was the same behavior as in the experiments, while the maximum values became quite a bit larger than those of the experiments. The time when the extensive axial strain began to increase was consistent with the time when the skeleton stress path reached at the failure line, that is, 2 hours in Case-7 and 2.3 hours in Case-8. The material failure led to the large deformation. The axial strain in the simulations became -2.5% in Case-7 and -2.0% in Case-8. The results indicate that as the excess pore pressure increases, the extensive axial strain becomes larger.
4.3 Simulation Results

Figure 4.6 Distributions of the temperature in Case-7, \( S^H = 15.4 \% \)

Figure 4.7 Distributions of the temperature in Case-8, \( S^H = 32.1 \% \)

Figure 4.8 Distributions of CO\(_2\)-hydrate remaining ratio in Case-7, \( S^H = 15.4 \% \)
4.3 Simulation Results

**Figure 4.9** Distributions of CO$_2$-hydrate remaining ratio in Case-8, $S^{H}_{r} = 32.1$ [%]

**Figure 4.10** Distributions of the pore gas pressure in Case-7, $S^{H}_{r} = 15.4$ [%]

**Figure 4.11** Distributions of the pore gas pressure in Case-8, $S^{H}_{r} = 32.1$ [%]
4.3 Simulation Results

Figure 4.12 Distributions of the pore water pressure in Case-7, $S^H = 15.4\%$.

Figure 4.13 Distributions of the pore water pressure in Case-8, $S^H = 32.1\%$.

Figure 4.14 Distributions of the suction in Case-7, $S^H = 15.4\%$.
4.3 Simulation Results

Figure 4.15 Distributions of the suction in Case-8, $S_H^U = 32.1 \%$

Figure 4.16 Distributions of the mean effective stress in Case-7, $S_H^U = 15.4 \%$

Figure 4.17 Distributions of the mean effective stress in Case-8, $S_H^U = 32.1 \%$
4.3 Simulation Results

Figure 4.18 Distributions of the viscoplastic volumetric strain in Case-7, $S^H_r = 15.4\%$

Figure 4.19 Distributions of the viscoplastic volumetric strain in Case-8, $S^H_r = 32.1\%$

Figure 4.20 Distributions of the accumulated viscoplastic deviator strain $\gamma^p$ in Case-7

$S^H_r = 15.4\%$
4.3 Simulation Results

Figure 4.21 Distributions of the accumulated viscoplastic deviator strain $\gamma^{vp}$ in Case-8,

$$S_r^{vp} = 32.1\%$$

Figure 4.22 Skeleton stress path
4.3 Simulation Results

Figure 4.23 Time profiles of the pore pressure in experiments and the average pore pressure in simulations

Figure 4.24 Time profiles of the effective confining pressure in the experiments and the mean skeleton stress in the simulations
4.4 Summary

In this chapter, a numerical simulation of the dissociation tests was conducted on a specimen containing CO$_2$-hydrates using the thermal recovery method under undrained conditions in order to investigate the dissociation and deformation behavior induced by hydrate dissociation. It was performed through a comparison of the experimental data with that of the simulations, using a chemo-thermo-mechanically coupled finite element analysis method. An elasto-viscoplastic constitutive equation was used for unsaturated soil considering structural changes, the effects of suction and hydrate saturation proposed by Kimoto et al., (2007a). The main points obtained from the simulations are summarized as follows:

1. The hydrate dissociation progressed from the surfaces of the specimen in the case of the thermal recovery method.

2. The pore gas pressure and the pore water pressure increased with decreases in the hydrate remaining ration, and its distribution was uniform among each element. The pore gas pressure at the final state became larger than that of the pore water pressure, and these differences were expressed in the results for the suction.

3. The mean skeleton stress decreased to approximately 0.0 MPa due to the hydrate dissociation in almost all the elements. It became a little bit larger around the top and the bottom surfaces than the middle part of the specimen because of the influences of the
boundary conditions.

4. The large viscoplastic volumetric strain and the deviator strain were generated by the large average pore pressure produced by the hydrate dissociation. The more the excess pore pressure was produced by the dissociation, the larger the viscoplastic volumetric strain and the deviator strain became. Those strains were likely to localize in the area where the mean skeleton stress was reduced.

5. When the hydrate dissociation initiated, the mean skeleton stress decreased drastically and moved toward the failure line.

6. The average pore pressure increased drastically when the hydrates dissociated in the simulations, and the increasing process was very similar to that of the experiments. The peak pressure became larger in the simulation results than the experimental one. One possible reason is that the liquefaction of CO₂ gas should occur in the experiments, while it was not considered in the simulations. The differences between ideal gas and real gas were also likely to produce such differences and, in the simulation, the gas was treated as ideal gas.

7. As for the mean skeleton stress and the effective confining pressure, the behavior was qualitatively similar between the simulations and the experiments. Thus, it can be said that the simulation results were able to describe the experimental results.

8. In both the experiments and the simulations, large extensive axial strain was observed. In the simulations, the extensive axial strain increased after the stress path reached the failure line. This means that it is possible for hydrate dissociation under undrained conditions to lead to the material failure and large deformation.
Chapter 5

INSTABILITY ANALYSIS AND NUMERICAL SIMULATION OF DISSOCIATION PROCESS OF METHANE HYDRATE-BEARING SOIL

5.1 Introduction

In this section, a linear stability analysis of methane hydrate-bearing soil considering dissociation is shown. We followed the method by Garcia et al. (2010), and extended it by considering the energy balance and hydrates-reaction process in order to deal with the dissociation phenomenon. The governing equations for the chemo-thermo-mechanically coupled behavior are based on Kimoto et al. (2010), and a viscoplastic constitutive model for the soil skeleton is used. Details of the governing equations for the stability analysis are shown in the following. Then, we conduct a numerical analysis of gas hydrate-bearing soil with dissociation in order to investigate the effect of the parameters on the system. The simulation method used in the present study can describe the chemo-thermo-mechanically coupled behaviors such as phase changes from hydrates to water and gas, changes in temperature, and
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

5.2.1 Governing equations

5.2.1.1 Stress variables

The stress variables are defined in the following one-dimensional form. Total stress \( \sigma \) is obtained from the sum of the partial stresses, namely,

\[
\sigma = \sigma^S + \sigma^H + \sigma^W + \sigma^G
\]

(5.1)

where superscripts \( S, H, W, G \) indicate the soil, hydrate, water, and gas phases, respectively. The partial stresses for the fluid phases can be written as

\[
\sigma^W = -n^W P^W
\]

(5.2)

\[
\sigma^G = -n^G P^G
\]

(5.3)

where \( P^W, P^G, n^W, \) and \( n^G \) are the pore water pressure, the pore gas pressure, the volume fraction of the water phase, and the volume fraction of the gas phase, respectively. Tension is positive for the stresses. For simplicity, we assume that the soil phase and the hydrate phase are in the same phase, namely, the solid phase. Thus, the partial stress of the solid phase is defined as

\[
\sigma^{SH} = \sigma^S + \sigma^H
\]

(5.4)

\[
\sigma^{SH} = \sigma' - n^{SH} P^F
\]

(5.5)

\[
n^{SH} = n^S + n^H
\]

(5.6)

where \( \sigma' \) is called the skeleton stress in the present study; it acts on the solid phase and is used as the stress variable in the constitutive equation. The terms \( n^S \) and \( n^H \) are the
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

volume fractions of the soil phase and the hydrate phase, respectively, and $P^F$ is the average fluid pressure given by

$$ P^F = sP^W + (1-s)P^G $$

(5.7)

where $s$ is the water saturation. Substituting Equations (5.2)–(5.7) into Equation (5.1), the skeleton stress is obtained as:

$$ \sigma' = \sigma + P^F $$

(5.8)

5.2.1.2 Conservation of mass

The conservation of mass for the soil, the water, the gas, and the hydrate phases are given by the following equations:

$$ \frac{D^a}{Dt} \left( n^a \rho^a \right) + n^a \rho^a \frac{\partial v^a}{\partial x} - m^a = 0 \quad (\alpha = S, W, G, H) $$

(5.9)

in which $\rho^\alpha$ is the material density for the $\alpha$ phase, $v^\alpha$ is the velocity vector of each phase, and $m^\alpha$ is the mass-increasing rate per unit volume due to hydrate dissociation. $D^a/Dt$ denotes the material time derivative following the particles of the $\alpha$ phase. Assuming that the densities of the soil, the water, and the hydrate are constant, that is, $\dot{\rho}^S = \dot{\rho}^W = \dot{\rho}^H = 0$, where the superimposed dot denotes the material time derivative, Equation (5.9) yields

$$ n^S \rho^S + n^S \rho^S \frac{\partial v^S}{\partial x} = 0 $$

(5.10)

$$ n^W \rho^W + n^W \rho^W \frac{\partial v^W}{\partial x} - m^W = 0 $$

(5.11)

$$ n^G \rho^G + n^G \dot{\rho}^G + n^G \rho^G \frac{\partial v^G}{\partial x} - \dot{m}^G = 0 $$

(5.12)
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

\[ \dot{n}^H \rho^H + n^H \rho^H \frac{\partial v^H}{\partial x} - \dot{m}^H = 0 \]  

(5.13)

in which the mass-increasing rate for the soil phase is zero, i.e., \( \dot{m}^s = 0 \). We assume that the soil particles and the hydrates move together, namely,

\[ v^s = v^H = v^{SH} \]  

(5.14)

Under the small strain condition, the spatial gradient of velocity for the solid phase is equal to the strain rate.

\[ \frac{\partial v^s}{\partial x} = \frac{\partial v^H}{\partial x} = \frac{\partial v^{SH}}{\partial x} = \dot{\epsilon} \]  

(5.15)

Considering Equations (5.14) and (5.15), Equations (5.10)~(5.13) provide

\[ -\dot{n}^s + (1-n)\dot{\rho}^s \dot{\epsilon} = 0 \]  

(5.16)

\[ \dot{n}^f s \dot{\rho}^w + n^f \dot{s} \rho^w + n^f s \rho^w \frac{\partial v^w}{\partial x} - \dot{m}^w = 0 \]  

(5.17)

\[ \dot{n}^f (1-s) \dot{\rho}^G + n^f \dot{s} \rho^G + n^f (1-s) \dot{\rho}^G + n^f (1-s) \dot{\rho}^G \frac{\partial v^G}{\partial x} - \dot{m}^G = 0 \]  

(5.18)

\[ \dot{n}^H = \frac{\dot{m}^H}{\rho^H} - n^H \dot{\epsilon} \]  

(5.19)

\[ n^f = \frac{V_E}{V} = \frac{V_w + V_G}{V} \]  

(5.20)

where \( V \) is the total volume of the system and \( V_\alpha \) is the volume of the \( \alpha \) phase. The apparent velocities of the water and the gas, with respect to the solid phase, are defined as:

\[ V_\beta = n^\beta (v_\beta - v^{SH}) \quad (\beta = W, G) \]  

(5.21)

In order to describe the changes in gas density, the equation for ideal gas is used, i.e.,
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

\[ \rho^G = \frac{M P^G}{R \theta} \]  

(5.22)

\[ \dot{\rho}^G = \frac{M}{R} \left( \frac{\dot{P}^G}{\theta} - \frac{P^G \dot{\theta}}{\theta^2} \right) \]  

(5.23)

\[ \frac{\dot{\rho}^G}{\rho^G} = \left( \frac{\dot{P}^G}{P^G} - \frac{\dot{\theta}}{\theta} \right) \]  

(5.24)

where \( M \) is the molar mass of gas and \( \theta \) is the temperature. Multiplying Equation (5.16) by \( s \left( \rho^w / \rho^s \right) \), adding Equation (5.17), and considering Equation (5.21), the continuity equation for the water phase can be written as

\[ s \dot{e} + \dot{\dot{n}}^F + \frac{\partial V^w}{\partial x} + \left( \frac{m^H}{\rho^H - \rho^w} \right) = 0 \]  

(5.25)

Similarly, multiplying Equation (5.16) by \( (1-s) \left( \rho^g / \rho^s \right) \), adding Equation (5.18), and considering Equation (5.21), the continuity equation for the gas phase can be obtained as:

\[ (1-s) \dot{e} + \dot{\dot{n}}^F + \frac{\partial V^G}{\partial x} + \left( \frac{m^H}{\rho^H - \rho^G} \right) = 0 \]  

(5.26)

5.2.1.3 Darcy type of law

The Darcy type of law for the flows of the water and the gas can be described as follows:

\[ V^w = -\frac{k^w}{\gamma^w} \left( \frac{\partial P^w}{\partial x} - \rho^w \overline{F} \right), \quad \gamma^w = \rho^w g \]  

(5.27)

\[ V^G = -\frac{k^G}{\gamma^G} \left( \frac{\partial P^G}{\partial x} - \rho^G \overline{F} \right), \quad \gamma^G = \rho^G g \]  

(5.28)
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

in which \( F \) is the gravitational force per unit mass, \( k^W \) and \( k^G \) are the permeability coefficients for the water and the gas phases, respectively, and \( g \) is the gravity acceleration.

5.2.1.4 Equation of equilibrium

The one-dimensional equilibrium equation can be written as:

\[
\frac{\partial \sigma}{\partial x} + \rho F = \frac{\partial \sigma'}{\partial x} + \frac{\partial P^e}{\partial x} + \rho F = 0 \quad (5.29)
\]

5.2.1.5 Conservation of energy

In the present study, we consider heat conductivity and the heat sink rate as being associated with the hydrate dissociation. The one-dimensional equation of energy conservation is written as:

\[
(\rho c) \dot{\theta} = -\frac{\partial h}{\partial x} + \sigma' \dot{\varepsilon} + \dot{Q}^H \quad (5.30)
\]

\[
\rho_c = \sum_{\alpha} n^\alpha \rho^\alpha c^\alpha \quad (\alpha = S, W, G, H) \quad (5.31)
\]

where \( c^\alpha \) is the specific heat of phase \( \alpha \), \( \theta \) is the temperature for all the phases, and \( \dot{Q}^H \) is the time rate of dissociation heat per unit volume due to the hydrate dissociation.

\[
\dot{Q}^H = (a - b \theta) \frac{\dot{N}_H}{V} \quad a = 56599, b = 16.744 \quad (5.32)
\]

Heat flux \( h \) follows Fourier’s law as:

\[
h = -k^\theta \frac{\partial \theta}{\partial x} \quad (5.33)
\]

in which \( k^\theta \) is the thermal conductivity for all phases.

Substituting Equations (5.32) and (5.33) into Equation (5.30), and assuming that the term \( \sigma' \dot{\varepsilon} \), which is related to the viscoplastic work, is very small and negligible, Equation (5.30) can be rewritten as
\[
\dot{\theta} = \frac{1}{\rho c} \left( k^\theta \frac{\partial^2 \theta}{\partial x^2} + (a - b \theta) \frac{\dot{N}_H}{V} \right) \tag{5.34}
\]

### 5.2.1.6 Dissociation rate of methane hydrates

Methane hydrates dissociate into water and methane gas with the reaction expressed in Equation (5.35).

\[
\text{CH}_4 \cdot n\text{H}_2\text{O (hydrate)} \rightarrow \text{CH}_4(\text{gas}) + n\text{H}_2\text{O (water)} \tag{5.35}
\]

where \( n \) is a hydrate number and that is assumed to be equal to 5.75 in natural hydrates. For the methane hydrate dissociation rate, \( \dot{N}_H \), we use Kim-Bishnoi’s equation (Kim et al., 1987), namely,

\[
\dot{N}_H = -D_H \exp \left( -\frac{9400}{\theta} \right) \left( P^e - P^F \right) \frac{1}{N_{H0}^{\frac{1}{3}} N_H^{\frac{2}{3}}} \quad D_H = 5.85 \times 10^{12} \tag{5.36}
\]

\[
P^e = c \exp \left( d - \frac{e}{\theta} \right), \quad c = 1.0 \times 10^{-6} (\text{MPa}), \quad d = 49.3185, \quad e = 9459 \tag{5.37}
\]

where \( N_H \) is the moles of hydrates in the volume \( V \), \( N_{H0} \) is the moles of hydrates in the initial state, \( P^F \) is the average pore pressure, and \( P^e \) is the equilibrium pressure at temperature \( \theta \). When the dissociation occurs, the dissociation rate is negative, i.e., \( \dot{N}_H < 0 \). The generating rates of water and gas are given by

\[
\dot{N}_W = -5.75 \dot{N}_H \tag{5.38}
\]

\[
\dot{N}_G = -\dot{N}_H \tag{5.39}
\]

The increasing mass rates for hydrates and the water and the gas phases, required in Equations (5.25) and (5.26), can be obtained from the above equations:
where $M_H$, $M_W$, and $M_G$ are the molar mass of the methane hydrates, the water, and the methane gas, respectively.

### 5.2.1.7 Simplified viscoplastic constitutive equation

In the analysis, a simplified viscoplastic constitutive model is used. The stress-strain relation can be expressed as:

$$\sigma' = H \varepsilon + \mu \dot{\varepsilon}$$  \hspace{1cm} (5.41)

where $\varepsilon$ is the strain, $\dot{\varepsilon}$ is the strain rate, $H$ is the strain hardening-softening parameter and $\mu$ is the viscoplastic parameter. We ignore the dependency of the hardening-softening parameter $H$ on the skeleton stress $\sigma'$, namely, we assume that strain hardening-softening parameter $H$ is a function of suction $P^C$ and hydrate saturation $S^H_r$ for simplicity. Viscoplastic parameter $\mu$ is a function of the temperature $\theta$.

$$\begin{cases} H = H\left(P^C, S^H_r\right) \\ \mu = \mu(\theta) \end{cases}$$  \hspace{1cm} (5.42)

Suction $P^C$ and the hydrate saturation $S^H_r$ are defined as:

$$P^C = P^G - P^W$$  \hspace{1cm} (5.43)

$$S^H_r = \frac{V^H}{V^V} = \frac{n^H}{n}$$  \hspace{1cm} (5.44)

### 5.2.2 Perturbed governing equations

Next, in order to estimate the instability of the material system, we consider the equilibrium equation, the continuity equation, the energy balance equation, the constitutive equations, and the equation of hydrate dissociation rate in a perturbed configuration. In the above governing
equations, the unknowns are pore water pressure \( P^W \), pore gas pressure \( P^G \), strain \( \varepsilon \), temperature \( \theta \), and the moles of hydrate \( N_H \). For each unknown, we suppose that

\[
\begin{align*}
P^W &= P^{W(0)} + \tilde{P}^W \\
P^G &= P^{G(0)} + \tilde{P}^G \\
\varepsilon &= \varepsilon^{(0)} + \tilde{\varepsilon} \\
\theta &= \theta^{(0)} + \tilde{\theta} \\
N_H &= N_H^{(0)} + \tilde{N}_H
\end{align*}
\] (5.45)

where the first terms on right side in Equation (5.45) indicate the values which satisfy the governing equations and the second terms are the perturbations of each variable. For the perturbations, we assume the following periodic form:

\[
\begin{bmatrix}
\tilde{P}^W \\
\tilde{P}^G \\
\tilde{\varepsilon} \\
\tilde{\theta} \\
\tilde{N}_H
\end{bmatrix} =
\begin{bmatrix}
P^{W*} \\
P^{G*} \\
\varepsilon^* \\
\theta^* \\
N_H^*
\end{bmatrix}
\exp (\omega t + iqx)
\] (5.46)

where \( q \) is the wave number \( (= 2\pi / L \text{ wave length}) \), \( \omega \) is the rate of the fluctuation growth, and superscript \( (\cdot)^* \) indicates the amplitude of each variable.

Disregarding the changes in material density and considering the body force as constant, the perturbation of the equilibrium equation, Equation (5.29), is expressed by

\[
\frac{\partial \tilde{\sigma}}{\partial x} = \frac{\partial \tilde{\sigma}'}{\partial x} - \frac{\partial \tilde{P}^F}{\partial x} = 0
\] (5.47)

where the perturbed variables are indicated by a tilde. From Equation (5.41), the perturbation of the skeleton stress \( \tilde{\sigma}' \) can be written as:

\[
\tilde{\sigma}' = \tilde{H}\varepsilon + \tilde{H}'\tilde{\varepsilon} + \tilde{\mu}\tilde{\varepsilon}' + \mu\tilde{\varepsilon}'
\] (5.48)
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

Since the strain hardening-softening parameter $H$ is a function of the suction and the hydrate saturation, and the viscoplastic parameter $\mu$ is a function of temperature, the perturbations of $H$ and $\mu$ are given as:

$$
\tilde{H} = \frac{\partial H}{\partial P_c} \tilde{P}^c + \frac{\partial H}{\partial S_r^H} \tilde{S}_r^H = H_{pc} \tilde{P}^c + H_{sh} \tilde{S}_r^H 
$$

(5.49)

$$
\tilde{\mu} = \frac{\partial \mu}{\partial \theta} \tilde{\theta} = -\alpha_\mu \tilde{\theta} 
$$

(5.50)

$$
\tilde{P}^c = \tilde{P}^g - \tilde{P}^w 
$$

(5.51)

$$
\tilde{S}_r^H = \frac{\tilde{n}^H}{n} - \frac{n^H}{n^2} \tilde{n} = \frac{M_H}{n p_H \epsilon V} \tilde{N}_H = \frac{n^H}{n^2} \tilde{\varepsilon} 
$$

(5.52)

Parameter $H$ increases with an increase in the suction $P_c$; consequently, the slope of the $H-P_c$ curve is positive, i.e., $H_{pc} > 0$. Similarly, $H$ increases with an increase in hydrate saturation $S_r^H$, i.e., $H_{sh} > 0$. On the other hand, viscoplastic parameter $\mu$ decreases with an increase in the temperature $\theta$ due to thermo-viscoplasticity; hence, $\partial \mu / \partial \theta$ is negative. Consequently, the term $\alpha_\mu (=-\partial \mu / \partial \theta)$ in Equation (5.50) is positive. Using Equation (5.46), and Equations (5.48)~(5.52), and considering $\tilde{\varepsilon} = \omega \tilde{\varepsilon}$, we obtain a spatial gradient of perturbed skeleton stress as

$$
\frac{\partial \tilde{\sigma}'}{\partial x} = \frac{\partial}{\partial x} \left\{ -\varepsilon H_{pc} \tilde{P}^w + \varepsilon H_{pc} \tilde{P}^G + \left( \mu \omega + H - \varepsilon H_{sh} \frac{n^H}{n^2} \right) \tilde{\varepsilon} - \alpha_\mu \varepsilon \tilde{\theta} + \varepsilon H_{sh} \frac{M_H}{n p_H \epsilon V} \tilde{N}_H \right\} 
$$

$$
= \left\{ -\varepsilon H_{pc} P^{w*} + \varepsilon H_{pc} P^{G*} + \left( \mu \omega + H - \varepsilon H_{sh} \frac{n^H}{n^2} \right) \varepsilon^* - \alpha_\mu \varepsilon \tilde{\theta}^* + \varepsilon H_{sh} \frac{M_H}{n p_H \epsilon V} N_H^* \right\} 
$$

$$
\times iq \exp(\omega t + iqx) 
$$

(5.53)

Similarly, the perturbation of average pore pressure $\tilde{P}^F$ can be written by means of Equation (5.7) as
In Equation (5.54), the degree of water saturation \( s \), is a function of the suction \( P^C \); the perturbation of the saturation is given as

\[
\tilde{s} = \frac{\partial s}{\partial P^C} \tilde{P}^C = B_c \tilde{P}^C
\]  

(5.55)

where \( B_c = \frac{\partial s}{\partial P^C} \) indicates the slope of the \( s - P^C \) curve.

Using Equations (5.46), (5.54), and (5.55), we have a gradient of the perturbed average pore pressure as

\[
\frac{\partial \tilde{P}^F}{\partial x} = \left( B_c P^C + s \right) P^{w*} + (1 - s - B_c P^C) P^{G*} \right) + \mu \omega + \frac{H - \varepsilon H \frac{n^H}{n^w}}{\varepsilon} \varepsilon^* - \alpha \omega \frac{\varepsilon}{n^w \rho \varepsilon} \frac{M_H}{n \rho \varepsilon} = 0
\]  

(5.57)

By substituting Equation (5.53) and (5.56) into Equation (5.47), and rearranging the terms, we obtain

\[
\begin{align*}
\frac{\partial \tilde{V}^{w}}{\partial x} + \left( \frac{5.75 M^w}{\rho \varepsilon} - s \right) \left( \frac{M^H}{\rho \varepsilon} \right) \tilde{N}^H = 0
\end{align*}
\]  

(5.58)

The perturbations of the continuity equations for both the water phase and the gas phase are given by

\[
\begin{align*}
(1 - s) \tilde{\varepsilon} - \tilde{s} n^F + \frac{\partial \tilde{V}^G}{\partial x} + (1 - s) \varepsilon \left( \frac{\tilde{P}^G}{\rho \varepsilon} - \frac{\tilde{n}^F}{\rho \varepsilon} \right) \left( \frac{\tilde{M}^G}{\rho \varepsilon} - (1 - s) \frac{M^H}{\rho \varepsilon} \right) \tilde{N}^H = 0
\end{align*}
\]  

(5.59)

The perturbation of the rate of the water saturation can be written as
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

\[ \tilde{s} = \omega B_s \tilde{P}^G \quad (5.60) \]

The perturbations of the spatial gradient of the Darcy type of equations for water and gas are

\[
\begin{align*}
\frac{\partial \tilde{V}^W}{\partial x} &= -\frac{\partial}{\partial x} \left[ \frac{k^W}{\gamma^W} \left( \frac{\partial \tilde{P}^W}{\partial x} - \rho^W \tilde{F} \right) \right] = -\frac{k^W}{\gamma^W} \frac{\partial^2 \tilde{P}^W}{\partial x^2} = -\frac{k^W}{\gamma^W} (i\omega)^2 \tilde{P}^W \quad (5.61) \\
\frac{\partial \tilde{V}^G}{\partial x} &= -\frac{\partial}{\partial x} \left[ \frac{k^G}{\gamma^G} \left( \frac{\partial \tilde{P}^G}{\partial x} - \rho^G \tilde{F} \right) \right] = -\frac{k^G}{\gamma^G} \frac{\partial^2 \tilde{P}^G}{\partial x^2} = -\frac{k^G}{\gamma^G} (i\omega)^2 \tilde{P}^G \quad (5.62)
\end{align*}
\]

By substituting Equations (5.46), (5.60), (5.61), and (5.62) into Equations (5.58) and (5.59), and rearranging the terms, we obtain

\[ -\left( B_n^F \omega - \frac{k^W}{\gamma^W} q^2 \right) P^{W*} + B_n^F \omega \tilde{P}^{G*} + s \omega \varepsilon^* + \left( \frac{5.75 M_w^W}{\rho^W V} - s \frac{M_H^H}{\rho^H V} \right) \omega \varphi^* = 0 \quad (5.63) \]

\[ B_n^F \omega \tilde{P}^{G*} - \left[ B_n^F - \frac{(1-s) n^G}{\theta} \right] \omega - \frac{k^G}{\gamma^G} q^2 \right) P^{G*} + (1-s) \omega \varepsilon^* \]

\[ -\frac{(1-s) n^F}{\theta} \omega \varphi^* + \left( \frac{M_G^G}{\rho^G V} - (1-s) \frac{M_H^H}{\rho^H V} \right) \omega \varphi^* = 0 \quad (5.64) \]

From Equation (5.34), the perturbation of the conservation of energy is given as

\[ \tilde{\theta} + \frac{1}{\rho c} \left[ k^\theta \frac{\partial^2 \tilde{\theta}}{\partial x^2} - (a-b\theta) \frac{\tilde{N}_H}{V} \right] = 0 \quad (5.65) \]

By using Equations (5.46) and (5.65), and rearranging the terms, we obtain

\[ \left( \omega + \frac{k^\theta}{\rho c} q^2 \right) \theta^* - \frac{a-b\theta}{\rho c V} \omega \varphi^* = 0 \quad (5.66) \]
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

Since the dissociation rate of the methane hydrates is a function of temperature $\theta$, average pore pressure $P^F$, and the moles of hydrate $N_H$, the perturbation of the dissociation rate can be written as

$$\tilde{N}_H = \frac{\partial N_H}{\partial \theta} \tilde{\theta} + \frac{\partial N_H}{\partial P^F} \tilde{P} + \frac{\partial N_H}{\partial N_H} \tilde{N}_H = -A_N \tilde{\theta} + B_N \tilde{P} - C_N \tilde{N}_H$$  \hspace{1cm} (5.67)

$$A_N = -\frac{\partial N_H}{\partial \theta} = D_H \exp \left(-\frac{9400}{\theta}\right) \left\{ \left(9400 + e\right) P^e - 9400P^F \right\} \frac{1}{N^3_{H0}} N^2_H$$  \hspace{1cm} (5.68)

$$B_N = \frac{\partial N_H}{\partial P^F} = D_H \exp \left(-\frac{9400}{\theta}\right) \frac{1}{N^3_{H0}} N^2_H$$  \hspace{1cm} (5.69)

$$C_N = -\frac{\partial N_H}{\partial N_H} = \frac{2}{3} D_H \exp \left(-\frac{9400}{\theta}\right) \left( P^e - P^F \right) \left( \frac{N_{H0}}{N_H} \right)^{\frac{1}{3}}$$  \hspace{1cm} (5.70)

By substituting Equations (5.46) and (5.54) into Equation (5.67), we obtain

$$-B_N \left(B_{C^C} + s\right) P^{w^r} + B_N \left(B_{C^C} + s - 1\right) P^{G^r} + A_N \tilde{\theta} + \left(\omega + C_N\right) \tilde{N}_H = 0$$  \hspace{1cm} (5.71)

Finally, we rewrite the perturbed governing equations, Equations (5.57), (5.63), (5.64), (5.66), and (5.71), in matrix form as
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

\[
\begin{bmatrix}
0 & 0 & \omega + \frac{k_h}{\rho_h} q^2 & -\frac{a-b\theta}{\rho_h V} \omega \\
-B_N \left(B_c P_C + s\right) & B_N \left(B_c P_C + s - 1\right) & 0 & \omega + C_N \omega \\
-(\varepsilon H_{PC} + B_c P_C + s) & \varepsilon H_{PC} + B_c P_C + s - 1 & \mu \omega + \left(H - \varepsilon H_{SH} n_H^2 n^2 \right) & -\alpha \mu \omega \\
-B_c n^2 \omega & B_c n^2 \omega & (1-s)\omega & -\frac{n^2 (1-s)}{\omega} \omega \left(m_g - (1-s)m_H \right) \omega \\
-B_c n^2 \omega + \frac{k_w}{\rho_w} q^2 & B_c n^2 \omega & s \omega & 0 & (m_w - sm_H) \omega \\
\end{bmatrix}
\]

\[
\begin{bmatrix}
P^{w*} \\
P^{g*} \\
\varepsilon^* \\
\theta^* \\
N_H^* \\
\end{bmatrix} = [A] \{y\} = \{0\} \quad (5.72)
\]

\[
m_w = \frac{5.75M_w}{\rho_w V}, \quad m_g = \frac{M_g}{\rho_g V}, \quad m_H = \frac{M_H}{\rho_H V} \quad (5.73)
\]

For the non-zero values of \( P^{w*}, P^{g*}, \varepsilon^*, \theta^*, \) and \( N_H^* \) in Equation (5.72), the determinant of the matrix \([A]\) has to be equal to zero. Under the condition that \( \det[A] = 0 \), we have a polynomial function of \( \omega \) as

\[
a_5 \omega^5 + a_4 \omega^4 + a_3 \omega^3 + a_2 \omega^2 + a_1 \omega + a_0 = 0 \quad (5.74)
\]

The details of coefficients \( a_i \ (i = 1 \sim 5) \) are noted as follows:
\[ a_5 = -B_c n^F \mu \left(1-s\right) n^F \frac{1}{\bar{p}^G} \] (5.75)

\[ a_4 = -\mu \left\{ B_c n^F \frac{k^G}{\bar{G}^G} + \left( B_c n^F - \frac{(1-s)n^F}{\bar{G}^G} \right) \frac{k^w}{\bar{w}^w} + B_c n^F \frac{(1-s)n^F k^0}{\rho c} \right\} q^2 \]

\[ -B_c n^F \left[ 1 + \mu B_N (m_w + m_G - m_H) + \frac{\mu (a-b\theta)(1-s)n^F}{\rho c} \left( \frac{A_N}{\bar{p}^G} - \frac{B_N}{\theta} \right) \right] \]

\[ -B_c n^F \left( \frac{(1-s)n^F}{\bar{G}^G} \right) \left\{ \mu C_N + H - \varepsilon H_{sh} \frac{n^H}{n^F} \right\} + \left( \varepsilon H_{pc} + B_c P^c \right) s \left( \frac{(1-s)n^F}{\bar{G}^G} \right) \left( s + \mu B_N (m_w - m_H) \right) \] (5.76)
\[ a_3 = -\mu \left\{ \left( B_c n^F - \frac{(1-s)n^F}{\bar{P}} \right) k^w \frac{k^0}{\rho c} - \frac{k^w}{\gamma^w} \frac{k^G}{\gamma^G} + B_c n^F \right\} q^4 \]

\[ - \left[ \mu C_N + H - \varepsilon H_{\text{sh}} \frac{n^H}{n^2} \right] \left\{ \left( B_c n^F - \frac{(1-s)n^F}{\bar{P}} \right) \frac{k^w}{\gamma^w} + B_c n^F \frac{(1-s)n^F}{\bar{P}} \frac{k^0}{\rho c} + B_c n^F \right\} \]

\[ + \frac{\mu A_N (a-b\theta)}{\rho c} \left\{ \left( B_c n^F - \frac{(1-s)n^F}{\bar{P}} \right) \frac{k^w}{\gamma^w} + B_c n^F \right\} \frac{\mu B_N (a-b\theta) (1-s)n^F}{\rho c} \frac{k^0}{\rho c} \]

\[ - \left( \varepsilon H_{\text{pc}} + B_c P^c + s \right) \left\{ s + \mu B_N \left( m_w - m_{m} \right) \right\} \left\{ \frac{k^G}{\gamma^G} + \frac{(1-s)n^F}{\bar{P}} \frac{k^0}{\rho c} + B_c n^F \left\{ 1 + \mu B_N \left( m_w + m_G - m_{m} \right) \right\} \right\} \frac{k^0}{\rho c} \]

\[ + \left( \varepsilon H_{\text{pc}} + B_c P^c + s - 1 \right) \left\{ (1-s) + \mu B_N \left( m_G - (1-s)m_{m} \right) \right\} \frac{k^0}{\rho c} \]

\[ - B_N \left( H - \varepsilon H_{\text{sh}} \frac{n^H}{n^2} \right) \left\{ B_c n^F \left( m_w + m_G - m_{m} \right) \left( \varepsilon H_{\text{pc}} + B_c P^c + s \right) \frac{(1-s)n^F}{\bar{P}} \right\} \]

\[ - \left\{ B_c n^F - \left( \varepsilon H_{\text{pc}} + B_c P^c + s \right) \frac{(1-s)n^F}{\bar{P}} \right\} \left\{ C_N - B_N \varepsilon H_{\text{sh}} \frac{m_{m}}{n^m} - m_{m} B_N H_{\text{sh}} \frac{n^m}{n} + \frac{(a-b\theta)}{\rho c} \left( A_N + \alpha \varepsilon B_N \right) \right\} \]

\[ - B_c n^F \frac{(a-b\theta)(1-s)n^F}{\rho c} \left( H - \varepsilon H_{\text{sh}} \frac{n^H}{n^2} \right) \left( \frac{A_N}{\bar{P}} - B_N \right) \theta \]
$a_3 = \mu \frac{k_w k_G k^0}{\gamma^w \gamma^G \rho c} q^6$

$$- \left[ \left( \mu C_n + H - \epsilon H_{SH} \frac{n^H}{n^2} \right) \left( B_e F - \frac{(1-s)n^F}{\bar{P}^G} \right) \frac{k_w k^0}{\gamma^w \rho c} + B_e F \left( \frac{k^G}{\gamma^G \rho c} \right) - \mu A_n \left( a - b \theta \right) \frac{k_w k^G}{\gamma^w} \right]$$

$$+ \left( \frac{1-s + \mu B_n \left( m_G - (1-s)m_H \right)}{(1-s)} \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k_w k^0}{\gamma^w \rho c} - \left( \epsilon H_{PC} + B_c P^C + s \right) \left( s + \mu B_n \left( m_w - s m_H \right) \right) \frac{k^G}{\gamma^G \rho c} \right] q^6$$

$$- \left[ \left( \frac{a - b \theta}{\rho c} \right) \left( A_n + \alpha_n \bar{B}_n \right) \left( 1-s \right) \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k_w k^0}{\gamma^w} - \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k^G}{\gamma^G} \right]$$

$$+ \left( C_n - B_n \epsilon H_{SH} \frac{n^H}{n^2} \right) \left( 1-s \right) \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k_w k^0}{\gamma^w} - \left( \epsilon H_{PC} + B_c P^C + s \right) \left( \frac{k^G}{\gamma^G} + \frac{1-s}{\bar{P}^G} \frac{k^0}{\rho c} \right)$$

$$+ \left( C_n + \left( \frac{a - b \theta}{\rho c} \right) \frac{A_n \left( H - \epsilon H_{SH} \frac{n^H}{n^2} \right)}{\left( B_e F - \frac{(1-s)n^F}{\bar{P}^G} \right)} \frac{k_w k^G}{\gamma^G} \right)$$

$$+ B_n \left( H - \epsilon H_{SH} \frac{n^H}{n^2} \right) \left( m_G - (1-s)m_H \right) \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k_w}{\gamma^w} - \left( m_w - s m_H \right) \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k^G}{\gamma^G} + \frac{1-s}{\bar{P}^G} \frac{k^0}{\rho c}$$

$$- B_n \left( H - \epsilon H_{SH} \frac{n^H}{n^2} \right) \left( a - b \theta \right) \frac{(1-s)n^F}{\rho c \theta} \left( \epsilon H_{PC} + B_c P^C + s \right) \frac{k_w}{\gamma^w}$$

$$+ B_c n^F \left\{ C_n - B_n \epsilon H_{SH} \frac{n^H}{n^2} + \left( H - \epsilon H_{SH} \frac{n^H}{n^2} \right) \left[ B_n \left( m_w + m^G - m^H \right) + \frac{1-s}{\bar{P}^G} \frac{C_n}{C_n} \right] \right\} \frac{k^0}{\rho c} \right] q^2$$

(5.78)
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

\[ a_1 = \left( \mu C_N + H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \frac{k^G k^W k^\theta}{\gamma^G \gamma^W \rho_c} q^6 \]

\[ + \left( C_N + \frac{(a-b\theta)}{\rho_c} A_N \right) \left( H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \frac{k^G k^W}{\gamma^G \gamma^W} q^4 \]

\[ - C_N \left( H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \left[ B_c n^F \frac{k^G}{\gamma^G} + \left( B_c n^F - \frac{(1-s)n^F}{\rho_c} \right) \frac{k^W}{\gamma^W} \right] \]

\[ - \left\{ C_N - B_m m_h \left( H + \frac{\varepsilon}{n} (1-S^m) H_{SH} \right) \right\} \left\{ s \left( \varepsilon H_{PC} + B_c P^C + s \right) \frac{k^G}{\gamma^G} - (1-s) \left( \varepsilon H_{PC} + B_c P^C + s - 1 \right) \frac{k^W}{\gamma^W} \right\} \]

\[ + B_n \left( H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \left\{ m^L \left( \varepsilon H_{PC} + B_c P^C + s \right) \frac{k^G}{\gamma^G} - m^G \left( \varepsilon H_{PC} + B_c P^C + s - 1 \right) \frac{k^W}{\gamma^W} \right\} \frac{k^\theta}{\rho_c} q^4 \]

\[ a_0 = C_N \left( H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \frac{k^W}{\gamma^W} \frac{k^G}{\gamma^G} \frac{k^\theta}{\rho_c} q^6 \]
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

Note that the sign for $q$, $s$, $n$, $n^F$, $n^H$, $\mu$, $\alpha$, $H_{PC}$, $H_{SH}$, $k^W$, $k^G$, $k^\theta$, $\gamma^W$, $\gamma^G$, $\rho c$, $m_w$, $m_G$, $m_H$, $A_N$, $B_N$, $C_N$, $a$, $b$, $P^C$, $P^G$, $\theta$, and $V$ in Equations (5.75)–(5.80) is always positive, whereas the sign for $B_C$ and $\dot{N}_H$ is negative. The sign for strain $\varepsilon$ is positive in expansion and negative in compression, and strain rate $\dot{\varepsilon}$ can be positive or negative. Strain softening-hardening parameter $H$ is positive in viscoplastic hardening and negative in viscoplastic softening.

5.2.3 Conditions of onset of material instability

In the following, we discuss the instability of the material system. If the growth rate of the perturbations $\omega$, which is the root of Equation (5.74), has a positive real part, the material system is unstable. On the contrary, if the real part of $\omega$ is negative, the material system is stable. The necessary and sufficient conditions whereby all the roots have negative real parts are given by the Routh-Hurwitz criteria. When the coefficient of the highest order of $\omega$ is positive, that is, $a_5 > 0$, the necessary and sufficient conditions that all the roots of Equation (5.74) have negative real parts are to satisfy all the equations from (i) to (v) expressed below.

\begin{align*}
(i) & \quad a_0 > 0, \quad a_1 > 0, \quad a_2 > 0, \quad a_3 > 0, \quad a_4 > 0 \tag{5.81} \\
(ii) & \quad D_1 = \begin{vmatrix} a_4 & a_2 \\ a_5 & a_3 \end{vmatrix} > 0 \tag{5.82} \\
(iii) & \quad D_2 = \begin{vmatrix} a_4 & a_2 & a_0 \\ a_5 & a_3 & a_1 \\ 0 & a_4 & a_2 \end{vmatrix} > 0 \tag{5.83} 
\end{align*}
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

(iv) \[ D_3 = \begin{vmatrix} a_4 & a_2 & a_0 & 0 \\ a_5 & a_3 & a_1 & 0 \\ 0 & a_4 & a_2 & a_0 \\ 0 & a_5 & a_3 & a_1 \end{vmatrix} > 0 \] (5.84)

(v) \[ D_4 = \begin{vmatrix} a_4 & a_2 & a_0 & 0 & 0 \\ a_5 & a_3 & a_1 & 0 & 0 \\ 0 & a_4 & a_2 & a_0 & 0 \\ 0 & a_5 & a_3 & a_1 & 0 \\ 0 & 0 & a_4 & a_2 & a_0 \end{vmatrix} > 0 \] (5.85)

The material system can be unstable if at least one of the conditions described above is not satisfied.

5.2.3.1 Sign of the coefficients \( a_5 \) and \( a_0 \)

At first, we will compare the sign of coefficients \( a_5 \) and \( a_0 \).

Considering the sign of the terms described above, \( a_5 \) is always positive.

\[ a_5 = -B_c n^F \mu \left(1-s\right)n^F > 0 \] (5.86)

Hence, when \( a_0 \) becomes negative, the first conditions of Routh-Hurwitz criteria is not satisfied, namely, the material system may become unstable. From Equation (5.80), it possible for \( a_0 \) to become negative.

\[ a_0 = C_N \left(H - \varepsilon H_{SH} \frac{n^H}{n^2}\right) k^w k^G k^\theta \rho_c q^6 < 0 \iff H - \varepsilon H_{SH} \frac{n^H}{n^2} < 0 \] (5.87)
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

From Equation (5.87), the material stability depends on strain hardening-softening parameter $H$, $H_{SH}$, strain $\varepsilon$, and the volume fraction of void $n$ and hydrate $n^H$. We will consider two cases; the first case is the compressive strain $\varepsilon < 0$ and the second is the expansive strain $\varepsilon > 0$.

- $\varepsilon < 0$: compressive strain

1) When parameter $H$ is positive, that is, the viscoplastic hardening, the term in $a_0$ is always positive. The sign for $a_0$ always becomes positive.

$$\varepsilon < 0, \ H > 0 \implies a_0 > 0$$

(5.88)

2) When the parameter $H$ is negative, that is, the viscoplastic softening, $a_0$ becomes negative, if it satisfies the following inequality.

$$\begin{cases} 
\varepsilon < 0, \ H < 0 \\
nH < \varepsilon H_{SH} S_r^H < 0, \quad \therefore S_r^H = \frac{n^H}{n} 
\end{cases} \implies a_0 < 0$$

(5.89)

Thus, the onset of the instability of the material system may appear in the following cases.

- Large value for $|H|$. When parameter $H$ is negative, namely, viscoplastic softening, it becomes easier to satisfy Equation (5.89). In other words, $a_0$ becomes negative more easily, and the possibility of instability is higher, when the absolute value for $H$ increases. This means that the larger the degree of softening becomes, the higher the possibility for material instability becomes.

- Large value for $n$. When $H$ is negative, namely, viscoplastic softening, the term $nH$ on the left side becomes negative more easily when the volume fraction of void $n$ increases. At the same time, the hydrate saturation becomes smaller with an
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

increase in the volume fraction of the void, and the possibility of satisfying Equation (5.89) becomes higher.

- Small value for $H_{sh}$. When the term $H_{sh}$ decreases, it becomes easier to satisfy Equation (5.89), namely, the possibility of instability is higher. This indicates that, if the bonding effect of hydrates on the mechanical properties is relatively small, the material system becomes more unstable.

- $\varepsilon > 0$: expansive strain

3) When the parameter $H$ is positive, that is, the viscoplastic hardening, $a_0$ becomes negative, if it satisfies Equation (5.90).

\[
\begin{cases}
\varepsilon < 0, \ H < 0 \\
0 < nH < \varepsilon H_{sh} S_r^H
\end{cases}
\]  

(5.90)

Thus, the onset of the instability of the material system may appear in the following cases:

- Small value for $H$. When the value for $H$ decreases, it becomes easier to satisfy Equation (5.90), namely, $a_0$ becomes negative more easily, and the possibility of instability is higher.

- Small value for $n$. When the volume fraction of void $n$ decreases, the term $nH$ becomes smaller. At the same time, the hydrate saturation becomes larger with a decrease in the volume fraction of the void, and it becomes easier to satisfy Equation (5.90).

- Large value for $H_{sh}$. When the term $H_{sh}$ increases, it becomes easier to satisfy Equation (5.90), namely, the possibility of instability becomes higher. This indicates that the gradient of the parameter $H$, with respect to hydrate saturation $S_r^H$, becomes larger and the material system becomes more unstable.

$\varepsilon > 0$: expansive strain
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

- Large value for expansive strain: \( \varepsilon > 0 \). When the strain is expansive and large, the term \( \varepsilon H_{SH} \varepsilon_r^H \) becomes larger. Consequently, the possibility of instability becomes higher.

4) When parameter \( H \) is negative, that is, viscoplastic softening, the term \( H - \varepsilon H_{SH} n^H / n^2 \) is always negative. Thus, the sign for \( a_0 \) is always negative. This may lead to the material instability, because it does not satisfy the first condition of the Routh-Hurwitz criteria.

\[
\begin{cases}
\varepsilon > 0, & H < 0 \\
H - \varepsilon H_{SH} n^H n^2 < 0 \Rightarrow a_0 < 0 
\end{cases}
\] (5.91)

In general, the material system is likely to be stable when it is in the viscoplastic hardening regions (Garcia et al., 2010; Kimoto et al., 2007b). However, the main point of this analysis is that the material instability may occur even in the viscoplastic hardening regions, in the case of the expansive strain. From the experimental results shown in Chapter 3, the possibility of the expansive strain has been mentioned when the hydrate dissociation occurs.

5.2.3.2 Sign of the coefficients \( a_5 \) and \( a_4 \)

Next, we will discuss the sign for \( a_4 \). Coefficient \( a_4 \) is given in Equation (5.76), and it can be regarded as a second order polynomial of the wave number \( q \).

\[
a_4 = a_{4,2} q^2 + a_{4,0} q^0
\] (5.92)

where \( a_{4,2} \) and \( a_{4,0} \) indicate the coefficients associated with \( q^2 \) and \( q^0 \), respectively. The coefficient of the highest order term of \( a_4 \) is second, namely \( a_{4,2} \), is always positive.
\[ a_{4,2} = -\mu \left\{ B_e n^F \frac{k_G}{\gamma^G} + \left( B_e n^F - \frac{(1-s)n^F}{\rho^G} \right) \frac{k_w}{\gamma^W} + B_e n^F \frac{(1-s)n^F k^G}{\rho c} \right\} > 0 \quad (5.93) \]

When the term \( a_{4,0} q^0 \) is positive, the sign for \( a_4 \) becomes positive. It is difficult, however, to clarify the sign for \( a_{4,0} \) because of complexity. Even when \( a_{4,0} q^0 \) is negative and the term \( a_{4,2} q^2 \) is larger than \( a_{4,0} q^0 \), \( a_4 \) becomes positive, that is,

\[ a_{4,2} q^2 > a_{4,0} q^0 \quad \Rightarrow \quad a_4 > 0 \quad (5.94) \]

Thus, we obtain conditions for which \( a_4 \) can be positive as follows:

- In the case of large values for \( k_w \), \( k_g \), and \( k^G \), \( a_4 \) can become positive more easily.

Since it is rather difficult to discuss the sign of coefficients \( a_i \sim a_4 \) due to the complexity, we will consider the two extreme conditions. The first case involves a large value for the wave number \( q \to \infty \), and the another case involves a small value for the wave number \( q \to 0 \).

When the wave number of fluctuation \( q \) is very large \( (q \to \infty) \), the sign for the coefficients are determined by the highest order of \( q \). In contrast, when the wave number of fluctuation \( q \) is very small \( (q \to 0) \), the sign for the coefficients is dominated by that of the lowest order of \( q \).

### 5.2.3.3 For the large value of the wave number \( q \)

For a large number of \( q \), the sign for coefficients \( a_0 \sim a_5 \) of the polynomial equation, Equation (5.74), should be determined by that of the highest order of \( q \), as shown below:
5.2 Instability Analysis of Methane Hydrate-Bearing Viscoplastic Materials

\[ a_{5,0} = a_5 = -B_c n^F \mu \frac{(1-s)n^E}{\rho G} (> 0) \]  \hspace{1cm} (5.95)

\[ a_{4,2} = -\mu \left\{ B_c n^F \frac{k^G}{\gamma^G} + \left( B_c n^F - \frac{(1-s)n^F}{\rho G} \right) \frac{k^W}{\gamma^W} \frac{k^G}{\gamma^G} \frac{k^G}{\rho c} \right\} q^2 \]  \hspace{1cm} (5.96)

\[ a_{3,4} = -\mu \left\{ \left( B_c n^F - \frac{(1-s)n^F}{\rho G} \right) \frac{k^W}{\gamma^W} \frac{k^G}{\gamma^G} \frac{k^G}{\rho c} + B_c n^F \frac{k^G}{\gamma^G} \frac{k^G}{\rho c} \right\} q^4 \]  \hspace{1cm} (5.97)

\[ a_{2,6} = \mu \frac{k^W}{\gamma^W} \frac{k^G}{\gamma^G} \frac{k^G}{\rho c} q^6 \]  \hspace{1cm} (5.98)

\[ a_{1,6} = \left( \mu C_N + H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \frac{k^G}{\gamma^G} \frac{k^G}{\gamma^G} \frac{k^G}{\rho c} q^6 \]  \hspace{1cm} (5.99)

\[ a_{0,6} = a_0 = C_N \left( H - \varepsilon H_{SH} \frac{n^H}{n^2} \right) \frac{k^G}{\gamma^G} \frac{k^G}{\gamma^G} \frac{k^G}{\rho c} q^6 \]  \hspace{1cm} (5.100)

From Equations (5.95)–(5.98), \( a_{5,0} \sim a_{2,6} \) are always positive, whereas it is possible that \( a_{1,6} \) and \( a_{0,6} \) can be negative. The conditions under which coefficient \( a_{0,6} \) becomes negative have been already discussed in the previous subsection. The condition under which \( a_{1,6} \) can be negative is written in the following:

\[ a_{1,6} < 0 \iff \mu C_N + H - \varepsilon H_{SH} \frac{n^H}{n^2} < 0 \]  \hspace{1cm} (5.101)

In a similar way to the sign for \( a_0 \), we discuss the possibilities that \( a_{1,6} \) becomes negative. Basically, the conditions of the negative \( a_{1,6} \) are the same as the conditions of \( a_0 \) as
expressed in Equation (5.91), except for that the sign depends on the magnitude of a term \( \mu C_N \).

- \( \varepsilon < 0 \): compressive strain

1) When parameter \( H \) is positive, that is, viscoplastic hardening, all the terms in Equation (5.100) are positive. The sign for \( a_{1,6} \) always becomes positive, namely, it is independent on the value of \( \mu C_N \).

\[
\begin{align*}
&\varepsilon < 0, \quad H > 0 \\
&\mu C_N + H - \varepsilon H_{sH} \frac{n^H}{n} > 0 \\
&\Rightarrow a_{1,6} > 0
\end{align*}
\] (5.102)

2) When parameter \( H \) is negative, that is, viscoplastic softening, \( a_{1,6} \) is negative, if it satisfies the following inequality:

\[
\begin{align*}
&\varepsilon < 0, \quad H < 0 \\
&nH < -n\mu C_N + \varepsilon H_{sH} S_{iH} < 0 \\
&\Rightarrow a_{1,6} < 0
\end{align*}
\] (5.103)

Thus, the onset of the instability of the material system may appear in the following cases:

- Small value for \( \mu C_N \). If the magnitude of \( \mu C_N \) decreases, it becomes easier to satisfy Equation (5.103).

- \( \varepsilon > 0 \): expansive strain

3) When parameter \( H \) is positive, that is, viscoplastic hardening, \( a_{1,6} \) becomes negative, if it satisfies Equation (5.101).

\[
\begin{align*}
&\varepsilon > 0, \quad H > 0 \\
&0 < n\left(H + \mu C_N \right) - \varepsilon H_{sH} S_{iH} < 0 \\
&\Rightarrow a_{1,6} < 0
\end{align*}
\] (5.104)
Thus, the onset of the instability of the material system may appear in the following cases:

- Small value for $\mu C_N$. If the magnitude of $\mu C_N$ decreases, it becomes easier to satisfy Equation (5.104).

4) When parameter $H$ is negative, that is, viscoplastic softening, the term $nH - \varepsilon H_{SH} S''_r$ is always negative. Thus, the sign for $a_{1,6}$ becomes negative when it satisfies the following inequality:

$$\left\{ \begin{array}{l} \varepsilon > 0, \quad H < 0 \\ nH - \varepsilon H_{SH} S''_r < -n\mu C_N < 0 \end{array} \right\} \Rightarrow a_{1,6} < 0$$  \hspace{1cm} (5.105)

- Small value for $\mu C_N$. When the magnitude of $\mu C_N$ decreases, it becomes easier to satisfy Equation (5.105), and $a_{1,6}$ becomes negative more easily.

From a previous discussion for the case of a large number of $q \to \infty$, we have obtained the results whereby a small value for $\mu C_N$ may lead to instability regardless of whether the strain is compressive or expansile, and whether the material is viscoplastic hardening or softening.

**Figure 5.1-(a) and (b)** shows the variations in parameter $C_N$ and dissociation rate $\dot{N}_H$ with respect to the value of $N_H/N_{H0}$, respectively. Parameter $N_H/N_{H0}$ means the ratio of the current moles of MHs to the initial moles, and it will be called as the MH remaining ratio. **Figure 5.1-(a) and (b)** can be drawn by using Equations (5.36) and (5.70). When the MH dissociation progresses, the $N_H/N_{H0}$ should be reduced from the initial value, namely, $N_N/N_{N0} = 1.0$. Parameter $C_N$ becomes smaller with an increase in the $N_H/N_{H0}$. By combining the results whereby a smaller value for $C_N$ makes the material system more unstable, it can be said that the material instability is more likely to occur in an early stage of the MH dissociation than in a later one. From **Figure 5.1-(b)**, the dissociation rate $\dot{N}_H$ increases with an increase in the MH remaining ratio. This also indicates the higher possibility for material instability at the beginning of the MH dissociation rather than at a later stage.
5.3 Numerical Simulation of Instability Analysis by an Elasto-viscoplastic Model

In the previous section, we discussed the conditions for the onset of the instability of methane hydrate-bearing sediments by means of an analytical method using a viscoplastic model and linear instability analysis. From the analysis, we found that material instability may occur in the case of both viscoplastic hardening and softening, and that the magnitude of parameters $H$, $H_{SH}$, and the hydrate saturation $S_r^H$ have a significant effect on the onset of the material instability as well as the sign for strain, that is, compression $\varepsilon < 0$ or expansion $\varepsilon > 0$. The permeabilities for water $k^W$ and gas $k^G$ are also essential parameters for material instability. In the case of a large number of $q$, the material system may become more unstable with a decrease in term $\mu C_N$, regardless of whether the strain is compressive or expansive and

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.1.png}
\caption{(a) Variation in parameters $C_N$ and (b) $\dot{N}_H$ with respect to $N_H/N_{H0}$}
\end{figure}

5.2.3.4 For the small value of the wave number $q$

Next, let us discuss the case for a small wave number: $q \to 0$. In the case of a small number for wave number $q$, the sign for each coefficient is determined by that of the lowest order of $q$. It is, however, too complex to discuss the sign for the lowest order of $q$.
whether $H$ is positive or negative.

In this section, the results of various numerical simulations of the dissociation-deformation problem using the one-dimensional finite element mesh will be presented in order to study the material instability by using the chemo-thermo-mechanically coupled model proposed in Chapter 2. The results will be compared to those of the instability analysis.

The effect of material parameters, especially the permeability, should be investigated. This is because in the previous section, it was found that larger permeability makes the material system more stable. The importance of the permeability was also mentioned by the experimental results in Chapter 3. In order to clarify the relationship between the permeability and the hydrate dissociation, a series of parametric studies on the permeability is conducted. The detailed conditions for the numerical simulation are described in the following section.

5.3.1 One-dimensional model and boundary conditions

Figure 5.2 illustrates a schematic drawing of the target area of MH-bearing sediments for the numerical simulations, and the finite element meshes and boundary conditions used in the simulations are shown in Figure 5.3. The seabed ground, at a depth of about 350 m from the top of the seabed ground surface and at a water depth of 1,010 m, is modeled. The MH-bearing layer lies at a depth of 290 m from the top of the ground surface with a thickness of 44 m. We assume the depressurization method for the hydrate dissociation, and the depressurizing source is set at the left boundary of the model from the surface of the seabed ground. For the finite element mesh, a homogeneous soil column in the horizontal direction with a thickness of 1 m is employed, as shown in Figure 5.3.

The left boundary is set to be permeable for water and gas and the adiabatic boundary. The right boundary is also set to be permeable; however, the boundary conditions of the right boundary are set to the isothermal, namely, the temperature is kept constant at 287 K. The initial state of the pore pressure and temperature at the depressurization source are shown in Figure 5.4 with the methane hydrate equilibrium curve. The initial pore water pressure is 13 MPa for all elements, which is the hydrostatic pressure, and the pore water pressure at the depressurization source is linearly reduced to the target pressure. The target value varies with respect to the initial pore pressure at depressurization source $p_{ini}$; the degree of depressurization $\Delta p$ varies from 20% to 80% with increments of 10%, as shown in Figure 5.4. By changing the magnitude of the depressurization, it becomes possible to control the MH
dissociation. The depressurizing rate for each case is the same; thus, the time when the pore pressure at the depressurization source reaches the target value is different among the cases. The total time of the simulation is 100 hours for each case.

The initial conditions and material parameters are listed in Table 5.1 and Table 5.2, respectively. Initial void ratio $e_0$ is 1.00, and the initial hydrate saturation in the voids, $S_{Hr}^0$, is 0.51. The material parameters for the constitutive model are summarized in Table 5.2. The material parameters for the constitutive equation are mainly determined from the results of triaxial tests and its parametric studies, whose samples were obtained from the seabed ground at the Nankai Trough (Oka & Kimoto, 2008).

In order to study the instability of the MH-bearing material system, the different values for the permeability are considered as well as different levels of depressurization. The permeability for water changes from $1.0 \times 10^{-3}$ to $1.0 \times 10^{-8}$ [m/s], and the permeability for gas phases is set to 10 times of the water permeability. The combinations of the permeability and the level of depressurization are listed in Table 5.3. The total number of cases is 42, that is, 6 permeability and 7 depressurization levels. The case name “Case-i-j” indicates that the permeability for the water phase is $1.0 \times 10^{-4}$ [m/s] and the depressurization level is $j$ [%]. In the following section, the results of the numerical simulation and the discussion which intends to show a trend in the material instability will be presented.
Numerical Simulation of Instability Analysis by an Elasto-viscoplastic Model

Figure 5.2 Schematic view of the target area of MH-bearing sediments for the numerical simulations

Modeled area

element
1 2 3

- Depressurizing source
- Permeable for water and gas
- Adiabatic boundary

50m

- Impermeable for water and gas
- Adiabatic boundary

Figure 5.3 Simulation model and boundary conditions

Figure 5.4 Conditions of change in pore pressure change at the depressurization source
5.3 Numerical Simulation of Instability Analysis by an Elasto-viscoplastic Model

Table 5.1 Initial conditions of the soil material

<p>| | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial void ratio</td>
<td>$e_0$</td>
<td>1.00</td>
</tr>
<tr>
<td>Initial water saturation</td>
<td>$s_{r0}$</td>
<td>1.0</td>
</tr>
<tr>
<td>Initial hydrate saturation</td>
<td>$S_{r0}^H$</td>
<td>0.51</td>
</tr>
</tbody>
</table>

Table 5.2 Material parameters for the constitutive equation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compression index</td>
<td>$\lambda$ 0.185</td>
</tr>
<tr>
<td>Swelling index</td>
<td>$\kappa$ 0.012</td>
</tr>
<tr>
<td>Initial shear elastic modulus [kPa]</td>
<td>$G_0$ 53800</td>
</tr>
<tr>
<td>Viscoplastic parameter [1/s]</td>
<td>$C_0$ $1.0\times10^{10}$</td>
</tr>
<tr>
<td>Stress ratio at failure</td>
<td>$M_f^*$ 1.09</td>
</tr>
<tr>
<td>Compression yield stress [kPa]</td>
<td>$\sigma_{mbi}^*$ 1882</td>
</tr>
<tr>
<td>Degradation parameter (\sigma_{wof}^* / \sigma_{mui}^*)</td>
<td>1.0</td>
</tr>
<tr>
<td>Degradation parameter</td>
<td>$\beta$ 0.0</td>
</tr>
<tr>
<td>Parameter for suction effect [kPa]</td>
<td>$p_i^C$ 100</td>
</tr>
<tr>
<td>Parameter for suction effect</td>
<td>$S_I$ 0.2</td>
</tr>
<tr>
<td>Parameter for suction effect</td>
<td>$s_d$ 0.25</td>
</tr>
<tr>
<td>Parameter for hydrate effect</td>
<td>$S_{ri}^H$ 0.51</td>
</tr>
<tr>
<td>Parameter for hydrate effect</td>
<td>$n_m$ 0.6</td>
</tr>
<tr>
<td>Parameter for hydrate effect</td>
<td>$n_d$ 0.75</td>
</tr>
<tr>
<td>Thermo-viscoplastic parameter</td>
<td>$\alpha$ 0.15</td>
</tr>
<tr>
<td>Permeability for water [m/s]</td>
<td>$k^W$ variable</td>
</tr>
<tr>
<td>Permeability for gas [m/s]</td>
<td>$k^G$ $10 \times k^W$</td>
</tr>
</tbody>
</table>
5.4 Numerical Results

5.4.1 Results of the MH dissociation-deformation problem

Figure 5.5 shows the results of the simulation for different values of the permeability and the depressurization levels. In the figure, the solid circles (●) indicates stable simulation results, while the × marks indicates unstable simulation results. The judgment of stable or unstable is given according to whether the calculation runs to the final time, namely, 100 hours.

\[
D_H = 0.585 \times 10^{10} \text{ [1/kPa \cdot sec]}
\]

Figure 5.5 Stable and unstable regions of permeability and the depressurization level during the MH dissociation process
5.4 Numerical Results

From Figure 5.5, it can be said that the material system becomes stable basically with an increase in permeability. That is because large permeability will make the dissipation of the fluid pressure produced by the MH dissociation easier, while a reduction in pore pressure will reach farther away from the depressurization source and more MHs will dissociate due to the large permeability. In the case of relatively lower permeability, that is, $k^w = 1.0 \times 10^{-7}$ and $k^w = 1.0 \times 10^{-8}$, it becomes more stable for the large depressurization levels than in the cases of $k^w = 1.0 \times 10^{-5}$ and $k^w = 1.0 \times 10^{-6}$. The reason why the lower permeability makes the material system more stable is that the low permeability may limit the spreading of the depressurization; consequently, the area where the MHs dissociates becomes smaller and the production of the pore gas pressure is reduced. The balance between the permeability and the depressurization is one of the important factors in material instability. In order to investigate the details for the onset of material instability, several cases are selected, namely, two stable cases and two unstable cases. For the stable cases, we choose Case-4-30 and Case-7-30, which have the same depressurized level and different permeabilities, and for the unstable cases, Case-4-40 and Case-7-40 are chosen. In Case-4-30 and Case-7-30, the depressurization finishes after about 9.4 hours, while in Case-4-40 and Case-7-40, it ends after 12.5 hours.

Figure 5.6-(a)–(d) shows the time profiles of pore gas pressure $P^G$ [MPa] in elements-1, 2, and 3 for each case. The pore gas pressure is calculated in the elements where the MHs begin to dissociate. In Case-4-30, which is illustrated in Figure 5.6-(a), the pore gas pressure decreases with the progress of the depressurization. The production of pore gas pressure in elements-2 and 3 is initiated soon after that in element-1. This is because the depressurization spreads to the next element easily due to the large permeability. After that, the pore gas pressure in each element becomes the same value, which is consistent with the depressurized one. In Case-4-40, on the other hand, the pore gas pressure diverges just after 9.4 hours, and the calculation stops as shown in Figure 5.6-(b). The large depressurization level may enhance gas production, and the permeability is not enough for the pore gas pressure to be allowed to dissipate. The time profiles of the pore gas pressure are the same as Case-4-30 until 9.4 hours; the pore gas pressure in each element consists of the depressurized value due to the larger permeability.

The time profiles of the pore gas pressure in the case of low permeability, that is, Case-7-30 and Case-7-40, are different from those of larger permeability, as illustrated in Figure 5.6-(c) and (d). The pore gas pressure in both element-2 and element-3 is produced behind that of element-1, because it is difficult for the depressurization to spread due to the low permeability. In the case of large permeability, the pore gas pressure in each element decreased at the same level. In Case-7-30 and Case-7-40, however, the pore gas pressure becomes higher in the element farther away from the depressurization source. It sometimes increases rapidly, mainly because it is more difficult for the pore gas pressure to dissipate than in the case of large
5.4 Numerical Results

permeability. In Case-7-40, the pore gas pressure becomes the high pressure level after about 60 hours. Finally, it diverges at 78 hours and the calculation stops, as shown in Figure 5.6-(d). The behavior of the pore gas pressure is unstable. The large degree of the depressurization produces a larger amount of gas than in Case-7-30, and this makes the material unstable.

In order to see the behavior of MH dissociation, the time profiles of the MH remaining ratio are illustrated in Figure 5.7-(a)–(d). The ratio is defined as the percentage of the current moles of MHs with respect to the initial mole, that is,

\[ \text{MH remaining ratio} = 100 \times \frac{N_M}{N_{H_0}} \% \]  

(5.106)

In the case of large permeability, the ratio decreases equally in each element. This means that the effect of the depressurization spreads in a similar extent because of the large permeability. When the depressurization stops at 9.4 hours, the MH remaining ratio becomes constant in Case-4-30. In the case of lower permeability, on the other hand, the behavior of the ratio differs among the elements. As for the MH remaining ratio in element-2 and element-3, the dissociation starts behind that of element-1; the MH dissociation begins after 15 hours in element-2 and after 24 hours in element-3. This is because it is hard for the depressurized area to spread due to the low permeability and the amount of dissociated MHs is lowered. It may become evidence that the material system becomes more stable even in the case of the lower permeability, as shown in Figure 5.5. The reduction of the ratio continues moderately even after finishing the depressurization at 9.4 hours in Case-7-30, meanwhile it becomes constant after 9.4 hours in Case-4-30. This indicates that a lower permeability can reduce the total amount of MH dissociation more than that of a higher permeability. However, the dissociation will continue for a long term. Both Case-4-40 and Case-7-40 are the unstable results; the calculation stops after 9.4 hours in Case-4-40 and after 78 hours in Case-7-40. It can be said that the material instability may occur in the short term, namely, during dissociation in the case of the larger permeability, and in the lower one, the material instability has to be considered over a long span, namely, even after the depressurization.

Figure 5.8-(a)–(d) and Figure 5.9-(a)–(d) show the time profiles of average pore pressure \( P^p \) and mean skeleton stress \( \sigma_m' \), respectively. Average pore pressure \( P^p \) and mean skeleton stress \( \sigma_m' \) are defined by Equations (2.16) and (2.22). The average pore pressure decreases with increases in the depressurization in each case. In Case-4-30 and Case-4-40, it declines linearly, because the pore pressure at the depressurization source is reduced linearly. In
Case-7-30 and Case-7-40, the average pore pressure in element-2 and element-3 decreases behind element-1, and the pressure gradient increases between element-1 and element-2 or element-3. When material instability occurs in Case-4-40 and Case7-40, the average pore pressure diverges in the same manner as the pore gas pressure shown in Figure 5.6. This abrupt increases in pore gas pressure $P^G$ and average pore pressure $P^F$ leads to a drastic decrease in the mean skeleton stress, as illustrated in Figure 5.9. The result whereby large pore fluid pressure produces a great reduction in the mean skeleton stress is the same as that obtained from the experiments. Figure 5.10 indicates the time profiles of total volumetric strain $\varepsilon_v$ for each element. It is worth noting that the volumetric strain is positive in expansion and negative in compression. In comparing Case-4-30 with Case-7-30, both cases are stable and the total volumetric strain in Case-4-30 becomes larger than that in Case-7-30. This is because the amount of drained water in Case-4-30 than that of Case-7-30 due to the large permeability. Finally, the total volumetric strain in elements-1, 2, and 3 become -2.0%, -1.7%, -1.5%, respectively in Case-4-30. In Case-7-30, the total volumetric strain in elements-1, 2, and 3 become -1.2%, -0.4%, and -0.5%, respectively. In Case-4-30, the depressurization stops at 9.4 hours, and the changes in average pore pressure and mean skeleton stress are also small; however, the total volumetric strain keeps increasing until 100 hours. The results indicate that the deformation is likely to continue increasing even after the changes in pore pressure and mean skeleton stress become small. In the unstable cases, that is, Case-4-40 and Case-7-40, the expansive volumetric strain is observed at the time the calculation stops. The large pore fluid pressure leads to the large expansive strain, and the material system becomes unstable. This results in the expansive strain making the material system more easily unstable, consistent with the results of the linear stability analysis.
5.4 Numerical Results

Figure 5.6 Time profiles of the pore gas pressure \( P^G \) [MPa]

(a) Case-4-30, (b) Case-4-40, (c) Case-7-30, (d) Case-7-40
Figure 5.7 Time profiles of the remaining MH ratio \( 100 \frac{N_{H}}{N_{H0}} [\%] \)
(a) Case-4-30, (b) Case-4-40, (c) Case-7-30, (d) Case-7-40
Figure 5.8 Time profiles of the average pore pressure $P^f$ [MPa]

(a) Case-4-30, (b) Case-4-40, (c) Case-7-30, (d) Case-7-40
5.4 Numerical Results

![Diagram of a 1m element with different boundary conditions: Depressuring source, Permeable for water and gas, Adiabatic boundary, Constant temperature (287K), Impermeable for water and gas.]

**Figure 5.9 Time profiles of the mean skeleton stress \[ \sigma'_{m} \] [MPa]**

(a) Case-4-30, (b) Case-4-40, (c) Case-7-30, (d) Case-7-40
5.4 Numerical Results

**Figure 5.10 Time profiles of the volumetric strain** $\varepsilon_v$ [%]

(a) Case-4-30, (b) Case-4-40, (c) Case-7-30, (d) Case-7-40
5.4.2 Results of variations in the coefficients

The constitutive equation used in the linear stability analysis is different from that used in the numerical simulation, namely, a simplified constitutive model was used in the instability analysis as shown in Equation (5.41). Therefore, a correspondence of the constitutive equation between the linear stability analysis and the numerical simulation should be well considered.

From Equation (5.41), the simplified constitutive equation can be rewritten as follows:

$$\dot{\epsilon} = \frac{1}{\mu} (\sigma' - H \dot{\epsilon}) \quad (5.107)$$

On the other hand, the viscoplastic strain rate $\dot{\epsilon}_{ij}^{vp}$ which is used in the FEM simulation can be expressed by considering Equation (2.41), namely,

$$\dot{\epsilon}_{ij}^{vp} = C_{ijkl} \exp \left( m' \left( \bar{\eta}^* + \bar{M}^* \ln \frac{\sigma'_m}{\sigma'_mb} \right) \right) \left\{ \eta^*_j - \eta_{j(0)}^* - \frac{1}{3} \delta_{ij} \left( \bar{M}^* - \frac{\eta_{mn}^* (\eta_{mn}^* - \eta_{mn(0)}^*)}{\bar{\eta}^*} \right) \right\} \quad (5.108)$$

and its second invariant becomes as follows

$$I^{vp} = \sqrt{\dot{\epsilon}_{ij}^{vp} \dot{\epsilon}_{ij}^{vp}} = C \exp \left( m' \left( \bar{\eta}^* + \bar{M}^* \ln \frac{\sigma'_m}{\sigma'_mb} \right) \right) \sqrt{1 + \frac{1}{3} \left( \bar{M}^* - \frac{\eta_{mn}^* (\eta_{mn}^* - \eta_{mn(0)}^*)}{\bar{\eta}^*} \right)^2} \quad (5.109)$$

Next, we will consider the perturbed formulation of Equation (5.109) in the following equation

$$\tilde{I}^{vp} = C \sqrt{1 + \frac{1}{3} \left( \bar{M}^* - \frac{\eta_{mn}^* (\eta_{mn}^* - \eta_{mn(0)}^*)}{\bar{\eta}^*} \right)^2} \cdot \exp \left( m' \left( \bar{\eta}^* + \bar{M}^* \ln \frac{\sigma'_m}{\sigma'_mb} \right) \right) \frac{m'}{\sigma'_mb} \left( \sigma'_{mb} \left( \bar{\eta}^* + \bar{M}^* \ln \frac{\sigma'_m}{\sigma'_mb} \right) - \bar{M}^* \bar{\sigma'}_{mb} \right) \quad (5.110)$$

where $\sigma'_{mb}$ is the hardening parameter, and the following relationships between $\sigma'_{mb}$ and its
increment $d\sigma'_{mb}$ are considered, namely,

$$
\sigma'_{mb} = N_s N_m \sigma'_{ma}(z) \exp\left(\frac{1+e}{\lambda - \kappa} \epsilon^{vp}_{kk}\right) \\
\sigma'_{ma}(z) = \sigma'_{maf} + (\sigma'_{mai} - \sigma'_{maf}) \exp(-\beta z)
$$

(5.111)

$$
\begin{align*}
\left\{ \begin{array}{l}
d\sigma'_{mb} = N_s N_m \sigma'_{ma}(z) \exp\left(\frac{1+e}{\lambda - \kappa} \epsilon^{vp}_{kk}\right) \left[ \frac{1+e}{\lambda - \kappa} d\epsilon^{vp}_{kk} + N_s N_m \exp\left(\frac{1+e}{\lambda - \kappa} \epsilon^{vp}_{kk}\right) d\sigma'_{ma}(z) \right] \\
d\sigma'_{ma}(z) = -\beta \left( \sigma'_{ma}(z) - \sigma'_{maf} \right) \exp(-\beta z) dz = -\beta \left( \sigma'_{ma}(z) - \sigma'_{maf} \right) dz \\
dz = \sqrt{d \epsilon^{vp}_{ij} d \epsilon^{vp}_{ij}}
\end{array} \right.
\end{align*}
$$

(5.112)

Let us assume that the left-hand side of Equation (5.110), namely, $\tilde{I}^{vp}$, is equivalent to $\tilde{e}$. In the analysis of the one-dimensional finite element mesh, the strain $\epsilon$ in Equation (5.107) corresponds to both the axial and the volumetric strain, that is,

$$
\epsilon = \epsilon_x = \epsilon_v
$$

(5.113)

By substituting $d\sigma'_{ma}(z)$ into $d\sigma'_{mb}$ yields

$$
d\sigma'_{mb} = N_s N_m \exp\left(\frac{1+e}{\lambda - \kappa} \epsilon^{vp}_{kk}\right) \left[ \frac{1+e}{\lambda - \kappa} \sigma'_{ma}(z) d\epsilon^{vp}_{kk} - \beta \left( \sigma'_{ma}(z) - \sigma'_{maf} \right) dz \right]
$$

(5.114)

Assuming that $d\sigma'_{mb}$ corresponds to $\tilde{\sigma}'_{mb}$, and $d\epsilon^{vp}_{kk}$ and $dz$ correspond to $\tilde{e}$, respectively, Equation (5.114) yields

$$
\tilde{\sigma}'_{mb} = N_s N_m \exp\left(\frac{1+e}{\lambda - \kappa} \epsilon^{vp}_{kk}\right) \left[ \frac{1+e}{\lambda - \kappa} \sigma'_{ma}(z) - \beta \left( \sigma'_{ma}(z) - \sigma'_{maf} \right) \right] \tilde{e}
$$

(5.115)

Considering that the term $\left\{ \sigma'_{mb} \left( \tilde{\eta}^{*} + \tilde{M}^{*} \ln \sigma'_{m}/\sigma'_{m0} \right) - \tilde{M}^{*} \tilde{\sigma}'_{mb} \right\}$ in the right-hand side of
5.4 Numerical Results

Equation (5.110) is corresponding to the term \((\sigma' - H\varepsilon)\) in the right-hand side of Equation (5.107), we obtain the following relationships, namely,

$$H = \tilde{M}^* N_s N_m \exp \left( \frac{1 + e}{\lambda - \kappa} \varepsilon_{kp}^v \right) \left[ \frac{1 + e}{\lambda - \kappa} \sigma_m'(z) - \beta \left( \sigma_m'(z) - \sigma_{muf}' \right) \right]$$

(5.116)

$$\frac{1}{\mu} = C(\theta) \frac{m'}{\sigma'_{mb}} \exp \left[ m' \left( \eta^* + \tilde{M}^* \ln \frac{\sigma'}{\sigma'_{mb}} \right) \right] \sqrt{1 + \frac{1}{3} \left( \tilde{M}^* - \frac{\eta_{mn}^*}{\eta_{mn(0)}} \right)^2}$$

(5.117)

From Equations (5.116) and (5.117), the parameters \(H_{PC}\), \(H_{SH}\), and \(\alpha_{\mu}\) in the linear stability analysis can be calculated as follows:

$$H_{PC} = \frac{\partial H}{\partial \rho^c}$$

$$= \tilde{M}^* N_s \exp \left( \frac{1 + e}{\lambda - \kappa} \varepsilon_{kp}^v \right) \left[ \frac{1 + e}{\lambda - \kappa} \sigma_m'(z) - \beta \left( \sigma_m'(z) - \sigma_{muf}' \right) \right] \frac{\partial N_s}{\partial \rho^c}$$

(5.118)

$$= \tilde{M}^* N_s \exp \left( \frac{1 + e}{\lambda - \kappa} \varepsilon_{kp}^v \right) \left[ \frac{1 + e}{\lambda - \kappa} \sigma_m'(z) - \beta \left( \sigma_m'(z) - \sigma_{muf}' \right) \right]$$

$$H_{SH} = \frac{\partial H}{\partial \rho^c}$$

$$= \tilde{M}^* N_s \exp \left( \frac{1 + e}{\lambda - \kappa} \varepsilon_{kp}^v \right) \left[ \frac{1 + e}{\lambda - \kappa} \sigma_m'(z) - \beta \left( \sigma_m'(z) - \sigma_{muf}' \right) \right] \frac{\partial N_m}{\partial \rho^c}$$

(5.119)
5.4 Numerical Results

\[
\alpha_{\mu} = -\frac{\partial \mu}{\partial \theta} = \frac{C'}{C(\theta)^2} \frac{\partial C(\theta)}{\partial \theta} = \frac{C'}{C(\theta)^2} \left[ (\alpha m' \tilde{M}'^*) \left( \frac{C(\theta)}{\theta} \right)^{\left( \alpha m' \tilde{M}'^* \right)^{-1}} \right] = \frac{\alpha m' \tilde{M}'^* C'}{\partial C(\theta)} \tag{5.120}
\]

As for the parameter \( B_C \), which is the gradient of the soil-water characteristic curve (SWCC), can be obtained from the equation of the SWCC as expressed in Equation (2.255).

\[
B_C = -\frac{\partial s}{\partial P_C} = -\alpha mn' (s_{\max} - s_{\min}) \left( \alpha P_C^c \right)^{\mu - 1} \left( 1 + \left( \alpha P_C^c \right)^\nu \right)^{-m-1} \tag{5.121}
\]

Using Equations (5.75)–(5.80) and Equations (5.116)–(5.121), the change of the coefficients in the one-dimensional finite element simulation have been calculated. Figure 5.11-(a)–(f), Figure 5.12-(a)–(f), Figure 5.13-(a)–(f), and Figure 5.14-(a)–(f) indicate the time profiles of the coefficients in element-1 in Case-4-30, Case-4-40, Case-7-30, and Case-7-40 with various wave number \( q \), respectively. As previously mentioned, Case-4-30 and Case-7-30 are the stable cases, while Case-4-40 and Case-7-40 are the unstable.

First of all, the time profiles of \( a_3 \) and \( a_4 \) were always 0.0 in all the cases, because all the terms included within \( a_3 \) and \( a_4 \) are related to the suction, the water saturation, and the parameter \( B_C \), and when the suction is 0.0, these terms also become 0.0. In that case, the polynomial equation, Equation (5.74), becomes the third order.

From Figure 5.11-(a)–(f), the coefficient \( a_3 \) in Case-4-30 was positive at first. However, after the beginning of hydrate dissociation, it changed to negative in the case where the wave number \( q \) was 0.01, 0.1, 1, 10, and 100. In the case of the largest wave number of \( q \), that is, \( q = 1000 \), the \( a_3 \) was kept at positive even though it was reduced drastically and came close to 0.0 after the initiation of hydrate dissociation. The coefficient \( a_2 \) was also positive before hydrate dissociation, and it became negative after the dissociation in the case of \( q = 0.01 \, 0.1 \) as shown in Figure 5.11-(a) and (b). Especially, in the case of \( q = 0.01 \), the coefficient \( a_2 \)
was always negative after the dissociation. In the case of \( q = 0.1 \), on the other hand, \( a_2 \) began increasing after about 10 hours, when the depressurization finished. Finally, it recovered to a positive value. The results show that large value of the wave number \( q \) makes the coefficient \( a_2 \) positive more easily, and that is consistent with the results obtained by the linear stability analysis. It will be possible to make sure the correspondence of the viscoplastic constitutive equations used in the numerical simulations with the simplified model used in the linear instability analysis. When the wave number \( q \) became the largest, that is, \( q = 1000 \), all the coefficients in Case-4-30 were always positive even after the hydrate dissociation as show in Figure 5.11-(f).

Let us see the results of Case-4-40, which is unstable case. The calculation stopped at 9.4 hours due to the divergences in pore gas pressure and the sequential drastic reduction of mean skeleton stress and large expansive volumetric strain. The time profiles of the coefficients in Case-4-40 are illustrated in Figure 5.12-(a)–(f). In this case, a drastic increase in the coefficient \( a_4 \) can be observed just before the calculation stopped, while the \( a_5 \) was always zero throughout the simulation. Thus, the polynomial Equation (5.74) became the fourth order temporarily. The nonzero value of \( a_4 \) might strongly affect the onset of the material instability, and it makes the material system more complicated and unstable. As for the other coefficients, the drastic increase or decreases can be found just before stopping the calculation. The behavior of the coefficients was basically similar to that of Case-4-30. The \( a_5 \) became negative after the initiation of hydrate dissociation in the case of \( q = 0.01, 0.1, 1, 10, 100 \). It was only the case that the \( a_5 \) was always positive even after the dissociation when the wave number \( q \) was 1000. In the case of \( q = 1000 \), all the coefficients were positive, and the material system should have been in the stable region from the Routh-Hurwitz criteria. However, the result was unstable. The perturbations with the small wave number, which means the long wave length, dominated the material system more strongly than the perturbations with the large wave number in Case-4-40. In Case-4-30, on the other hand, it was likely that the material stability was more dominated by the perturbations with the large wave number, that is, the short wave length.

Figure 5.13-(a)–(f) shows the time profiles of the coefficients in Case-7-30, which is the stable case. Similar to Case-4-30, the coefficients \( a_5 \) and \( a_4 \) were zero throughout the simulation.
The coefficient $a_3$ was positive at first, and then it changed to negative after the dissociation.

This behavior was the same as that of Case-4-30. However, the value of $a_3$ in Case-7-30 was kept negative even in the case of the larger wave number, that is, $q = 1000$, while in Case-4-30 it became positive throughout the calculation. The result means that the lower permeability can make the material more unstable, and it is consistent with the result obtained by the linear stability analysis. Finally, $a_3$ became always positive when $q$ became more than 1000. The influences of the permeability can be found in the other coefficients, for example $a_2$ and $a_4$. Especially, $a_4$ in Case-4-30 was positive even if the wave number $q$ was the smallest one, that is, $q = 0.01$. In Case-7-30, however, $a_4$ became negative when the hydrate dissociation began and it was kept negative until 100 hours in the case where $q$ was 0.01, 0.1, 1, 100, and 1000. It was similar manner to the behavior of $a_3$. All the coefficients became positive only in the case of $q = 15000$. Both Case-4-30 and Case-7-30 are the stable cases, however, the range of the wave number $q$ where all the coefficients become positive is smaller in Case-7-30 than that in Case-4-30. The time profiles of the coefficients in Case-7-40, which is the unstable case, were basically the same as Case-7-30 as shown in Figure 5.14-(a)–(f). The prominent behavior in Case-7-40 was found the time profile of the coefficient $a_0$. Compare the coefficient $a_0$ in Case-7-40 to that in Case-7-30, the unstable behavior of fluctuation was observed in Case-7-40 after 60 hours. It might be because that the large depressurization level stimulated the hydrate dissociation more than the case of relatively smaller depressurization level, and the dissociation continued for a long time.
5.4 Numerical Results

(a) Case-4-30: $\kappa_0 = 1.0 \times 10^{-4} \text{ [m/s]}$, Depressurizing level=30%, $q=0.01$

(b) Case-4-30: $\kappa_0 = 1.0 \times 10^{-4} \text{ [m/s]}$, Depressurizing level=30%, $q=0.1$

(c) Case-4-30: $\kappa_0 = 1.0 \times 10^{-4} \text{ [m/s]}$, Depressurizing level=30%, $q=1$

Figure 5.11-(a)–(c) Time profiles of the coefficients in Case-4-30 with various wave number $q$
5.4 Numerical Results

(d) Case-4-30: \( k^h=1.0 \times 10^4 \) [m/s], Depressurizing level=30%, \( q=10 \)

(e) Case-4-30: \( k^h=1.0 \times 10^4 \) [m/s], Depressurizing level=30%, \( q=100 \)

(f) Case-4-30: \( k^h=1.0 \times 10^4 \) [m/s], Depressurizing level=30%, \( q=1000 \)

Figure 5.11-(d)–(f) Time profiles of the coefficients in Case-4-30 with various wave number \( q \)
5.4 Numerical Results

(a) Case-4-40: $k^0=1.0 \times 10^4$ [m/s], Depressurizing level=40%, $q=0.01$

(b) Case-4-40: $k^0=1.0 \times 10^4$ [m/s], Depressurizing level=40%, $q=0.1$

(c) Case-4-40: $k^0=1.0 \times 10^4$ [m/s], Depressurizing level=40%, $q=1$

Figure 5.12-(a)–(c) Time profiles of the coefficients in Case-4-40 with various wave number $q$
5.4 Numerical Results

(d) Case-4-40: $k^h=1.0\times10^{-4}$ [m/s], Depressurizing level=40%, $q=10$

(e) Case-4-40: $k^h=1.0\times10^{-4}$ [m/s], Depressurizing level=40%, $q=100$

(f) Case-4-40: $k^h=1.0\times10^{-4}$ [m/s], Depressurizing level=40%, $q=1000$

Figure 5.12-(d)–(f) Time profiles of the coefficients in Case-4-40 with various wave number $q$
5.4 Numerical Results

(a) Case-7-30: $k^h=1.0\times10^7$ [m/s], Depressurizing level=30%, $q=0.01$

(b) Case-7-30: $k^h=1.0\times10^7$ [m/s], Depressurizing level=30%, $q=0.1$

(c) Case-7-30: $k^h=1.0\times10^7$ [m/s], Depressurizing level=30%, $q=1$

Figure 5.13-(a)~(c) Time profiles of the coefficients in Case-7-30 with various wave number $q$
(d) Case-7-30: $k^0=1.0\times10^{-7}$ [m/s], Depressurizing level=30%, $q=100$

(e) Case-7-30: $k^0=1.0\times10^{-7}$ [m/s], Depressurizing level=30%, $q=1000$

(f) Case-7-30: $k^0=1.0\times10^{-7}$ [m/s], Depressurizing level=30%, $q=15000$

Figure 5.13-(d)-(f) Time profiles of the coefficients in Case-7-30 with various wave number $q$
5.4 Numerical Results

(a) Case-7-40: $k^0=1.0\times10^7$ [m/s], Depressurizing level=40%, $q=0.01$

(b) Case-7-40: $k^0=1.0\times10^7$ [m/s], Depressurizing level=40%, $q=0.1$

(c) Case-7-40: $k^0=1.0\times10^7$ [m/s], Depressurizing level=40%, $q=1$

Figure 5.14-(a)–(c) Time profiles of the coefficients in Case-7-40 with various wave number $q$
5.4 Numerical Results

(d) Case-7-40: $k_w=1.0 \times 10^7$ [m/s], Depressurizing level=40%, $q=100$

(e) Case-7-40: $k_w=1.0 \times 10^7$ [m/s], Depressurizing level=40%, $q=1000$

(f) Case-7-40: $k_w=1.0 \times 10^7$ [m/s], Depressurizing level=40%, $q=15000$

Figure 5.14-(d)-(f) Time profiles of the coefficients in Case-7-40 with various wave number $q$
5.5 Summary

In the first part of this chapter, a linear stability analysis was performed in order to investigate the effects of the parameters on the onset of the instability of MH-bearing sediments induced by dissociation. The governing equations of the MH-bearing sediments used in the chapter are based on the chemo-thermo-mechanically coupled model proposed by Kimoto et al. (2010), and for the constitutive equation, we used a linearized viscoplastic constitutive model. The main conclusions obtained in the stability analysis are listed as follows:

1. The parameters which have a significant influence on the material instability are the viscoplastic hardening-softening parameter, its gradient with respect to hydrate saturation, the permeability of water and gas, the strain, and the gradient of the dissociation rate with respect to the moles of MHs.

2. Material instability may occur in both the viscoplastic hardening region and the softening region regardless of whether the strain is compressive or expansive. However, when the strain is expansive, material instability can occur even if it is in the viscoplastic hardening region. The expansive strain makes the possibility of the instability higher in the model.

3. Permeability is one of the most important parameters associated with material instability. The larger the permeability for water and gas become, the more stable the material system becomes. In other words, the lower the permeability is, the higher the possibility is for material instability to occur. These results are consistent with the results obtained from the experimental studies.

4. It was found that the material becomes more unstable in the earlier stage of the dissociation than the later. The initiation of dissociation will make the material system more unstable.

In the second part of the chapter, some examples of the numerical simulation of the dissociation-deformation problem using the one-dimensional finite element mesh were presented in order to study the material instability by using the chemo-thermo-mechanically coupled model proposed in Chapter 2. The effect of the material parameters, especially the permeability, was investigated, because in the previous section, it was found that a larger permeability makes the material system more stable. The importance of the permeability was also mentioned in the experimental results in Chapter 3. In order to clarify the relationship between the permeability and the hydrate dissociation, a series of parametric studies on the permeability was conducted. For the dissociation method, we adopted the depressurization...
method. The main results of the numerical simulations are summarized as follows:

5. Basically the simulation results became more stable with increases in permeability. However, they also became stable in the region of the lower permeability. This was because the depressurized area was limited due to the low permeability; and consequently, the MH dissociation was also reduced.

6. When the calculation became unstable, the pore gas pressure diverged, and then the mean skeleton stress was decreased drastically. The larger expansive volumetric strain was also observed. These results are consistent with those obtained from the experimental studies and the linear stability analysis.

7. In the case of a higher permeability, the divergence occurred during depressurization and MH dissociation. On the other hand, in the case of the lower one, the instability was observed around the end part of the simulation when the MH dissociation almost converged. It is important to consider the material instability over the long term, that is, even after the dissociation calms down.

8. The compressive volumetric strain kept increasing after the depressurization finished and the changes in pore pressure and mean skeleton stress became small. It also proves the importance of considering the long term stability.

9. From the results of signs of the coefficients, in both the stable cases and the unstable cases, the signs of the coefficients became different in the case of small wave number. On the other hand, all the signs became positive in the case of large wave number.

10. Before MHs dissociated, all the coefficients had the same sign, however, after the dissociation, some coefficients changed to negative value. This indicates that MHs dissociation can make the material system more unstable.
5.5 Summary
Chapter 6

CONCLUSIONS AND FUTURE WORKS

6.1 Concluding Remarks

In the present study, the deformation behavior of the gas hydrate-bearing soils subjected to hydrate dissociation was investigated through an experimental study and its reproductive simulation using a chemo-thermo-mechanically coupled method. In addition, a linear instability analysis of methane hydrate bearing sediments considering hydrate dissociation was carried out in order to investigate the onset of the material instability. The conclusions obtained in each chapter are described below.

In Chapter 2, the elasto-viscoplastic constitutive equation for gas hydrate-bearing sediments proposed by Kimoto et al. (2007a) was introduced. The model is based on the overstress-type of viscoplastic theory proposed by Adachi and Oka (1982) with considerations given to the soil structure degradation for saturated soils (Kimoto & Oka, 2005; Kimoto et al., 2004), the effect of suction using the skeleton stress, and the effect of suction on independent stress variables (Oka et al., 2008a). After that, the governing equations for a chemo-thermo-mechanically coupled analysis and the finite element formulations were presented in this chapter.

In Chapter 3, a series of formation and dissociation tests of CO₂-hydrates were performed
using a temperature-controlled, high-pressure triaxial apparatus in order to characterize the mechanical behavior of gas hydrate sediments subjected to hydrate dissociation. The dissociation tests were conducted using a thermal recovery method under undrained conditions for both the pore gas and the pore water by assuming that the local permeability of the sediments was quite low. In forming the CO$_2$-hydrates in the specimens, a drastic drop in pore pressure and the simultaneous consumption of CO$_2$ gas were observed. This was the evidence for the formation of CO$_2$-hydrates. From the results of the dissociation tests, the hydrate dissociation was seen to cause a significant increase in the pore pressure and to result in significant decreases in the effective confining pressure with extensive axial strain under undrained conditions. In real situations, when MHs dissociate inside areas where the local permeability is quite low, the possibility of the sediments being unstable becomes higher.

In Chapter 4, dissociation tests on a specimen containing CO$_2$-hydrates under undrained conditions were simulated by the chemo-thermo-mechanically coupled finite element method introduced in Chapter 2, in order to compare the experimental results with those of the simulation subjected to hydrate dissociation. In addition to the comparative study, the numerical simulation gave the dissociation-deformation behavior in more detail, such as the distributions of volumetric strain, the degree of the CO$_2$-hydrate-dissociated area, and the effective stress inside the specimen. The behavior associated with the changes in pore pressure and mean skeleton stress was very similar to that of the experiments. In the simulation, the data of volumetric strain were obtained, which were not measured in the experiments. The large viscoplastic volumetric strain and the deviator strain were generated by the large average pore pressure and the decrease in effective stress induced by the hydrate dissociation. The more excess pore pressure was generated by the hydrate dissociation, the larger the viscoplastic volumetric strain and the deviator strain became.

In Chapter 5, a linear stability analysis was performed in order to investigate the effects of the parameters on the onset of the instability of MH-bearing sediments induced by dissociation. The parameters which have a significant influence on the material instability are the viscoplastic hardening-softening parameter, its gradient with respect to the hydrate saturation, the permeability for water and gas, the strain, and the gradient of the dissociation rate with respect to the moles of MHs. When strain is expanded, material instability may occur even if the material is in the viscoplastic hardening region. Expansive strain makes the possibility of the instability higher in the model. In addition to the linear stability analysis, some examples of numerical simulation of the dissociation-deformation problem using the one-dimensional finite element mesh were presented in order to study the effect of the material parameters, especially the permeability, was investigated, because in the previous section, it was found that the larger permeability makes the material system more stable. When the calculation became unstable, the
pore gas pressure and the mean skeleton stress diverged drastically. The larger expansive volumetric strain was also observed. These results are consistent with those obtained from the experimental studies and the linear stability analysis. Basically the simulation results became more stable with increases in permeability. However, they also became stable in the region of the lower permeability. This was because the depressurized area was limited due to the low permeability, and consequently, the MH dissociation was also restricted.

From the results of signs of the coefficients, in both the stable cases and the unstable cases, the signs of the coefficients became different in the case of small wave number. On the other hand, all the signs became positive in the case of large wave number. Before MHs dissociated, all the coefficients had the same sign, however, after the dissociation, some coefficients changed to negative value. This indicates that MHs dissociation can make the material system more unstable.

### 6.2 Recommendations for Future Work

Further investigations on the dissociation-deformation behavior of gas hydrate-bearing sediments are required in order to clarify the mechanism of the instability which will lead to material failure and submarine landslides. The topics of research that deserve future investigation are listed below.

In this experimental study, the volumetric strain of the hydrate-bearing specimen associated with hydrate dissociation was unobtainable due to the leakage in the double cell. The volumetric strain should be one of the most important parameters in describing the mechanical properties of unsaturated soils.

Only dissociation tests were conducted in this study. A series of triaxial shear tests would be required, with specimens in various states, that is, without hydrates, before and after dissociation, and during dissociation, in order to know how the strength of the samples changes after the hydrate dissociation. In addition, knowledge of cyclic behavior of gas hydrate-bearing sediments would be necessary for considering the effects of earthquakes on the sediments. The results of such investigations would be useful for evaluating the long term stability.

As for the numerical simulation, the gas was treated as ideal gas. However, in a high pressure environment where gas hydrates form, the gas should be treated as real gas for a more precise
6.2 Recommendations for Future Work

simulation. In addition, it is preferable to take into account the effect of the liquefaction of CO₂ gas in simulating the experimental results.

In the instability analysis, the relationships between the permeability and the depressurization were investigated through a parametric study. The same parametric studies should be carried out on other parameters, such as the strain hardening-softening parameters, and other dissociation methods, for example, the thermal recovery method.


