

Study of the Effectiveness of Microwave Susceptors for Syngas Production from Biomass and Evaluation of Its Economic and Social Acceptability

バイオマスからの合成ガス製造におけるマイクロ波吸収材料の

有効性の研究と経済的・社会的受容性の評価

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Abstract

Chapter 1 describes the significance of biomass utilization, thermochemical conversion, and microwave heating. The present study is positioned within the microwave heating biomass conversion research landscape. It has gained increasing attention due to its potential for enhanced efficiency and sustainability compared to conventional thermochemical processes. Recent advancements in this field have focused on optimizing susceptor materials, improving syngas and hydrogen yields, and assessing the techno-economic feasibility of large-scale applications. Based on this information, the present study investigates the role of magnetite as a susceptor in microwave heating (Chapter 2), the effect of steam feeding on syngas production (Chapter 3), and the economic and environmental feasibility of a microwave biomass conversion system (Chapter 4). An overview of Chapters 2 to 4 is provided below.

Chapter 2 investigated the effect of susceptor addition to lignocellulosic biomass. The present study focused on magnetite as a susceptor and mixed with cellulose for microwave heating at 800 °C. The effects of the magnetite–cellulose weight ratio, cycle number, and microwave heating were investigated. The results revealed that syngas yield increased with the addition of magnetite, having a maximum yield of 19.1 mmol/g-cellulose with a magnetite–cellulose weight ratio of 1. This yield was higher than conventional heating at 800 °C and similar to conventional heating at 1000 °C. This suggests that the magnetite addition induces internal heating, which increases syngas yield and improves energy efficiency. However, the syngas yield of microwave heating decreased by 40 % with increasing cycle number owing to the reduction of magnetite to iron. Thus, steam oxidation treatment was performed to regenerate magnetite. With the treatment, the syngas yield of the regenerated magnetite was similar to that in the 1st cycle. Therefore, steam oxidation is effective for the repeated use of magnetite. This study demonstrated that magnetite addition can effectively improve the syngas yield.

In Chapter 3, Microwave heating (MWH) of lignocellulosic biomass with susceptors has attracted attention for H₂ production. In this study, H₂ is produced from cellulose via MWH at 800 °C in a steam atmosphere with magnetite as the susceptor. The effects of steam feeding and MWH on the product yields are experimentally investigated compared to conventional heating (CH). The H₂ and CO₂ yields are increased by steam feeding for both MWH and CH because the water–gas shift reaction occurs. Without steam feeding, MWH enhances H₂ and CO production,

increasing maximum energy efficiency. With steam feeding, the steam gasification of C is more predominant for CH than for MWH as the reduction in the C content in the char of CH is higher than that of MWH. MWH reduces the formation of secondary char and is, therefore, expected to overcome issues such as the clogging or blockage of gas lines.

Chapter 4 discusses the techno-economic environment assessment of the microwave biomass conversion system. Recently, the thermochemical conversion of biomass using microwave heating has attracted attention because valuable products, such as hydrogen, carbon monoxide, etc., can be obtained more efficiently than conventional heating. However, the usefulness of the microwave biomass conversion system has not been evaluated sufficiently. The present study performed a techno-economic and social acceptability assessment of the microwave biomass conversion system. As a result, the microwave biomass conversion system was not economically feasible when the energy requirement for biomass conversion (E_{conv}) and the microwave heating unit cost (C_{MW}) were 20 MJ/kg-biomass and 100,000 USD/unit, respectively. The sensitivity analysis for the internal rate of return (IRR) and the net present value (NPV) indicated that the E_{conv} and the C_{MW} must be reduced by 10 % and 13 % to make this system economically feasible. Considering the hydrogen sales price in 2030, the microwave biomass conversion system was not economically feasible, although the E_{conv} and the C_{MW} were the minimum values. This system became economically feasible when a quantity discount of 50 % was applied to the initial cost. In addition, the power conditioning ability relative to the pumped storage (A_{power}) and utilization rate of forest residues (U_{residue}) in Kyushu were calculated to evaluate social acceptability. As a result, the microwave biomass conversion system was found to have an A_{power} of 24 to 31 % and improve the U_{residue} by 10.6 %. These results suggested that this system functions as a power conditioning and contributes to improving the utilization of forest residues. The microwave biomass conversion system is expected to suit Kyushu and other regions requiring electrical power conditioning capability and utilization of forest residues.

In Chapter 5, the conclusion and the future works were described. The syngas production method of the microwave biomass conversion system using magnetite was investigated under different temperatures, atmospheres, weight ratios, and cycle properties. Based on the experimental data, a techno-economic assessment of the microwave biomass conversion system was performed. As a result, the microwave biomass conversion system using magnetite is economically feasible if the heat dissipation from the heat insulator is reduced by 80 %. An oil

analysis and temperature measurement inside samples are needed for future work. The usefulness of the magnetite microwave biomass conversion system can be indicated in determining the oil products. Materials under microwave heating need to be observed from macro to micro perspectives. The temperature effects during microwave heating must be obtained in real-time temperature profiles to be discussed more properly. Therefore, establishing a real-time and trans-scale temperature measurement method will deepen the academic knowledge of microwave biomass conversion technology.

1. Introduction

1.1 Climate change

IPCC AR6 reports that human activities cause climate change [1]. Fig. 1.1.1 shows the change in global surface temperature. The simulated global surface temperature (solar and volcanic) remains almost constant. Conversely, the simulated global surface temperature (human and natural) and observed global surface temperature increased from around 1925 and rapidly increased from 2000. Fig. 1.1.2 shows the observed warming driven by carbon dioxide, methane, etc. Carbon dioxide is the main factor in climate change. *The observed warming is driven by emissions from human activities, with greenhouse gas warming partly masked by aerosol cooling* (described in IPCC AR6 WGI SPM p. 7). *In 2019, atmospheric CO₂ concentrations were higher than at any time in at least 2 million years (high confidence), and concentrations of CH₄ and N₂O were higher than at any time in at least 800,000 years (very high confidence). Since 1750, increases in CO₂ (47 %) and CH₄ (156 %) concentrations far exceed – and increases in N₂O (23 %) are similar to – the natural multi-millennial changes between glacial and interglacial periods over at least the past 800,000 years (very high confidence)* (described in IPCC AR6 WGI SPM p.8 A2.2). Therefore, human activities are one of the factors in climate change, and CO₂ is the main factor in increasing the global surface temperature. To mitigate the climate change problem, temperature rise must be reduced.

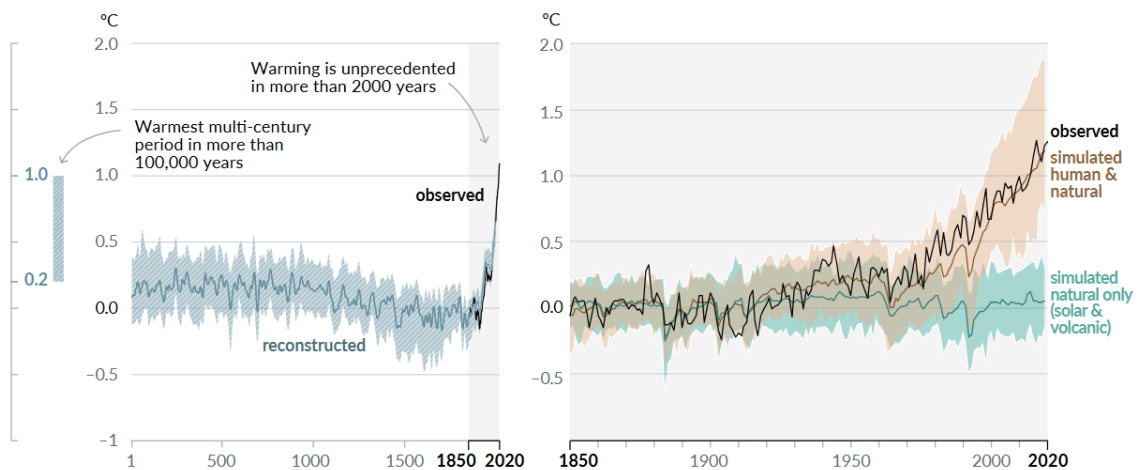


Fig. 1.1.1 Change in global surface temperature (left: decadal average, right: annual average) [1].

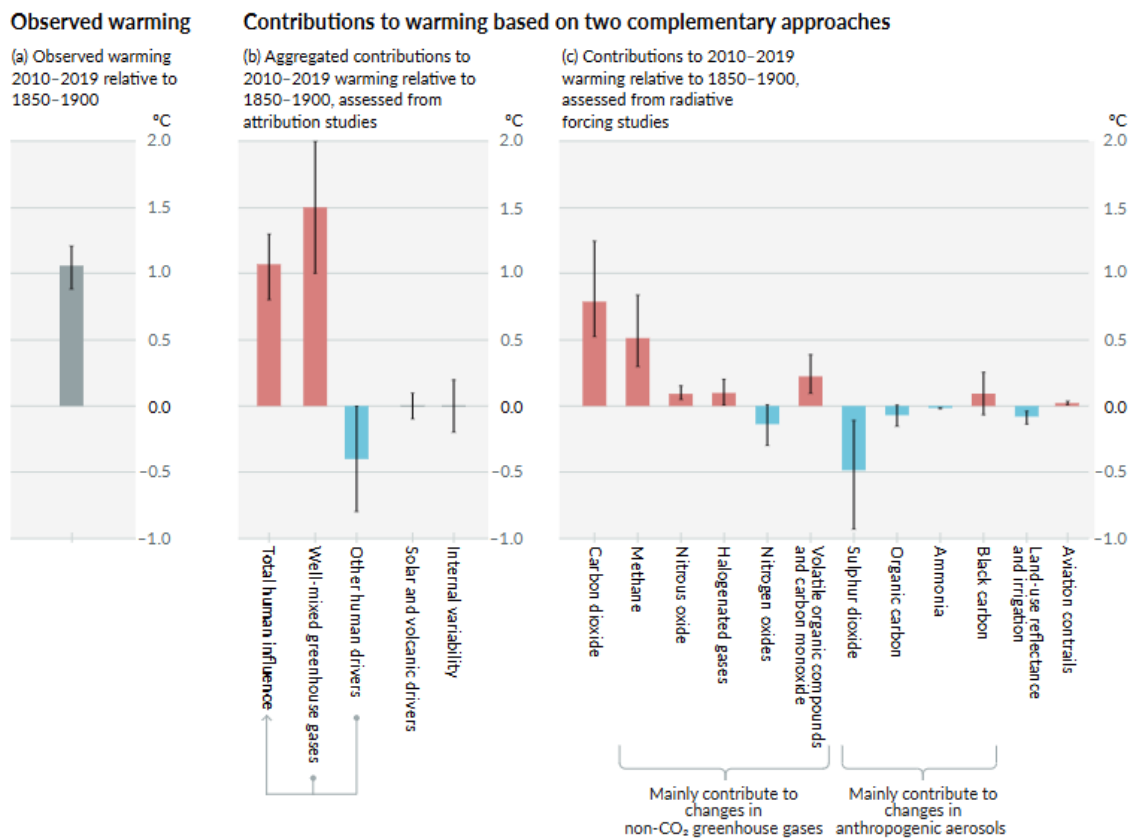


Fig. 1.1.2 Observed warming is driven by each factor [1].

Fig. 1.1.3 shows the greenhouse gas emissions from human activities [2]. CO₂ emissions (GtCO₂-eq/year) from fossil fuels and industry increased from around 1900 and rapidly increased from 1950 (under 10 GtCO₂-eq/year) to 2019 (more than 30 GtCO₂-eq/year). Fig. 1.1.4 shows the carbon budgets and emissions in each scenario. In 2020, the cumulative CO₂ emissions were approximately 2,400 GtCO₂. The carbon budget is 500 GtCO₂ for achieving a goal of the temperature rise until 1.5 °C. In the case of 2.0 °C, the carbon budget is 900 GtCO₂. CO₂ emissions are assumed to reach approximately 500 GtCO₂ when the 2019 CO₂ is emitted constantly. The temperature rise (Warming since 1850—1900) increased with cumulative CO₂ emissions (GtCO₂) since 1850. The temperature rise exceeds the 2.0 °C when the cumulative CO₂ emissions exceed the 4,000 GtCO₂. Climate change is summarized below if the global surface temperature increases by 1.5 and 2.0 °C. To reduce the rise in temperature, CO₂ emissions must be reduced. COP 28 aimed to be independent from fossil fuels to achieve the 1.5 °C scenario. Therefore, humans need to be independent from fossil fuels. However, our energy and chemical products depend on fossil fuels and require alternative resources. Biomass has attracted attention as an alternative to fossil fuels. The next section describes how to utilize biomass.

	1.5 °C	2.0 °C
The population hit by heat waves	9% (0.7 billion people)	28% (2 billion people)
Floods risk	+100%	+170%
Sea level rise	+48 cm until 2100	+56cm until 2100
coral reefs	70% disappearance	Approximately all disappear
Arctic sea ice loss frequency	1 time per 100 years	1 time per 10 years
Water use possibilities	0.35 billion people encounter a lack of water until 2100	0.41 billion people encounter a lack of water until 2100

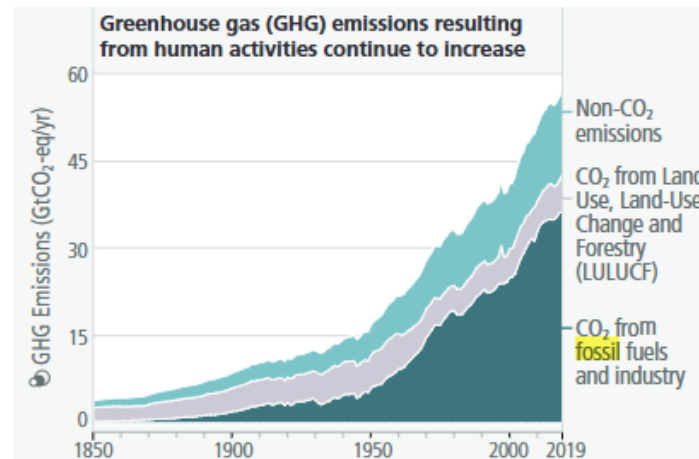


Fig. 1.1.3 Greenhouse gas (GHG) emissions from human activities [2].

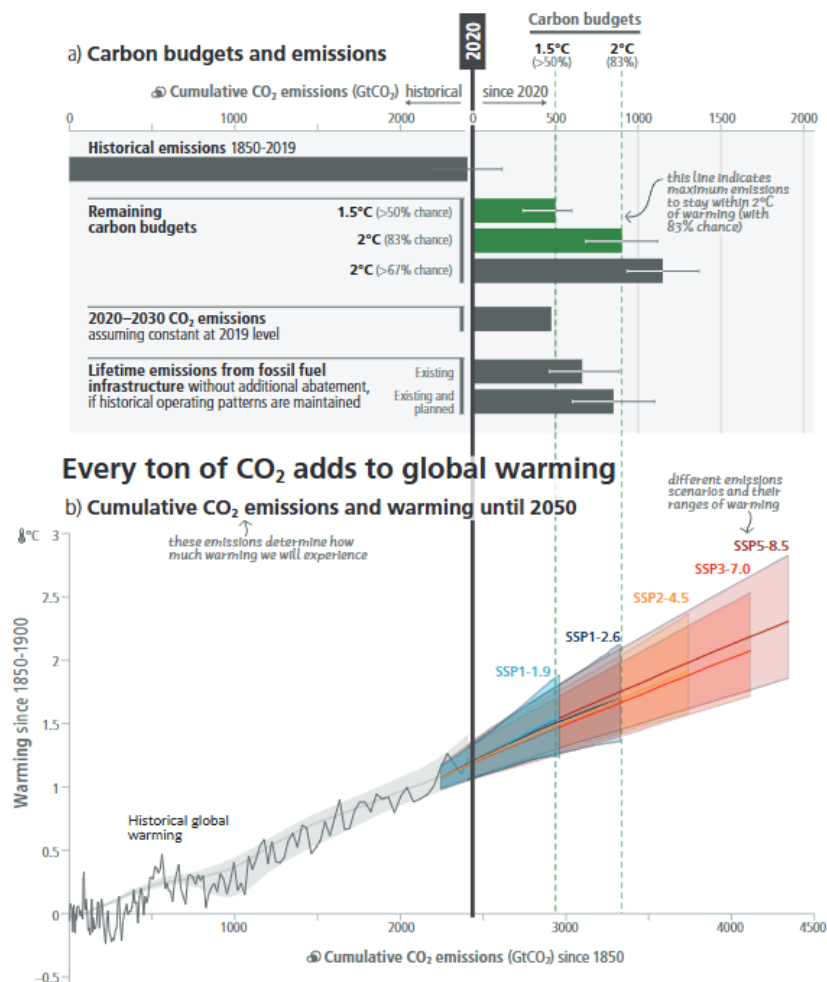


Fig. 1.1.4 Carbon budgets and emissions in each scenario [2].

1.2 Utilization of Biomass

Biomass is an essential resource for becoming independent from fossil fuels. Fig. 1.2.1 shows the global use of bioenergy by sector in the Net Zero Scenario from 2010 to 2030 [3]. The traditional use of biomass refers to traditional cooking methods using basic technologies such as a three-stone fire or basic improved cookstoves. Modern solid bioenergy includes all solid bioenergy except the traditional use of biomass. The traditional use of biomass decreased with the year, and the energy from the traditional use was set to 0 in 2030. Conversely, the energy from modern bioenergy, liquid biofuels, and biogas are expected to increase. Various types of biomasses exist, such as lignocellulosic biomass, herbaceous and agricultural biomass, aquatic biomass, animal and human biomass wastes, contaminated biomass, and industrial biomass wastes (semi-biomass) [4]. Lignocellulosic biomass is an abundant resource that can support the sustainable production of renewable fuels. Forest residues (a kind of lignocellulosic biomass) degrade the forest environment. Therefore, the utilization of forest residues is promoted.

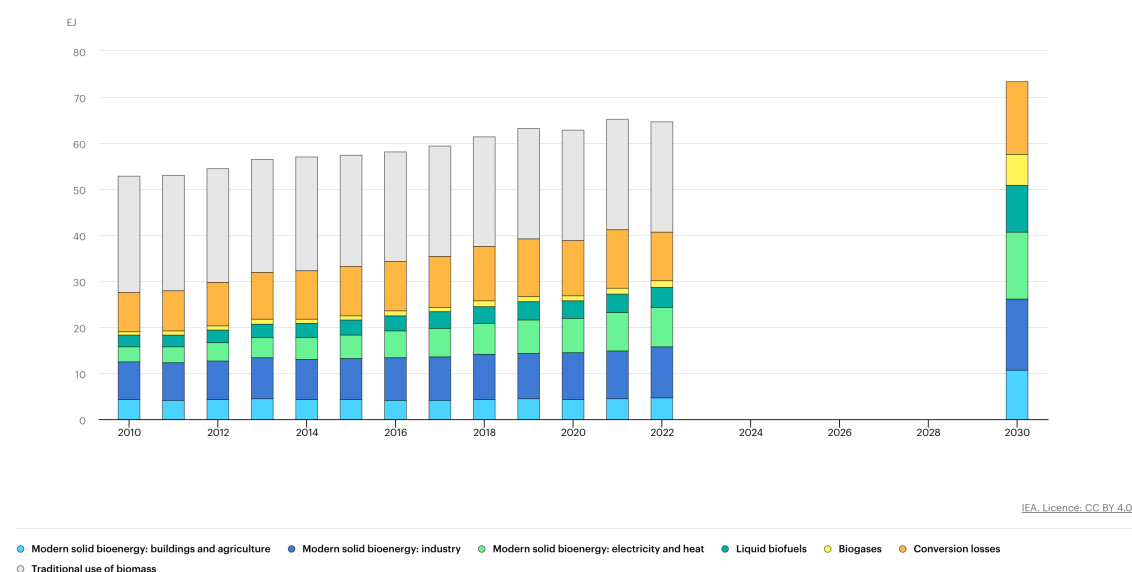


Fig. 1.2.1 Bioenergy use by sector globally in the Net Zero Scenario, 2010-2030 [3].

1.3 Thermochemical conversion of biomass

Biomass can be converted to useful products through physical, thermochemical, and bio-chemical treatment (shown in Fig. 1.3.1) [5]. The thermochemical conversion of biomass is one of the bio-fuel production methods used to obtain syngas. In the thermochemical conversion, several techniques are distinguished by temperature and reaction atmosphere. Fig. 1.3.2 shows the thermochemical conversion process of biomass and products [6,7].

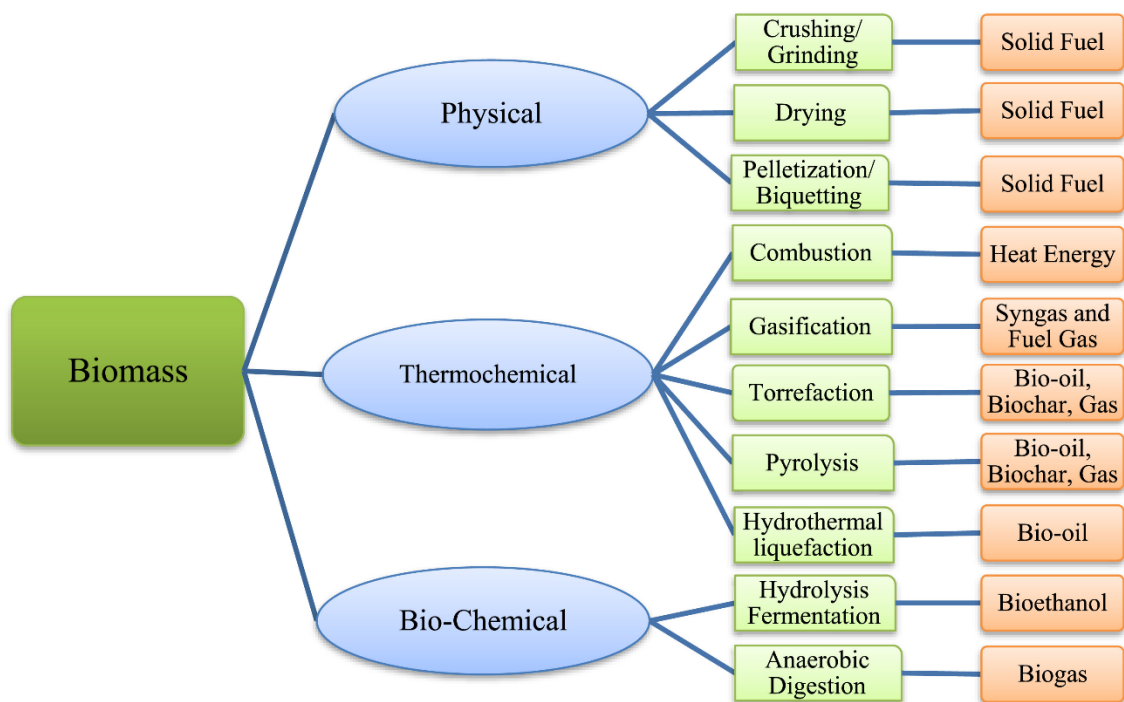
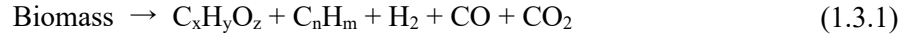
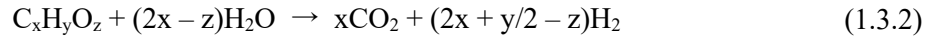


Fig. 1.3.1 Biomass conversion process [5].

Pyrolysis means biomass is heated at 350–700 °C in an oxygen-free atmosphere. The reaction temperature is the main factor that affects pyrolysis products. Generally, the bio-oil is the main product at 500–600 °C [8–10]. Gaseous products are also generated, and the reaction is described as follows:



Liquefaction means that biomass heated at 200–600 °C in a solvent, such as water, ethanol, acetone, glycerol, phenols, and tetralin under high pressure (5–25 MPa) [11]. The liquefaction technique converts biomass into bio-oil (40–60 wt%), biochar (10–12 wt%), and gases (35–45 wt%). Gasification means that biomass heated at 600–1300 °C with the gasification agents such as O₂, steam, and CO₂ [12]. Gasification agents also contribute to following gas shift reactions.



FAST PYROLYSIS PROCESSES FOR BIOMASS

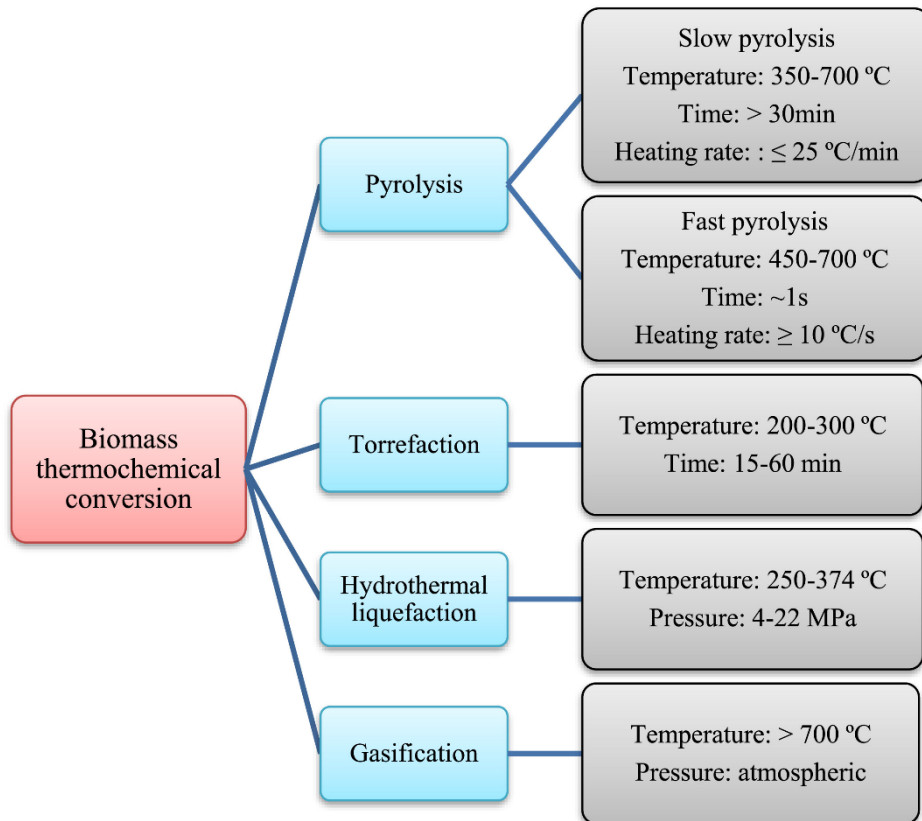
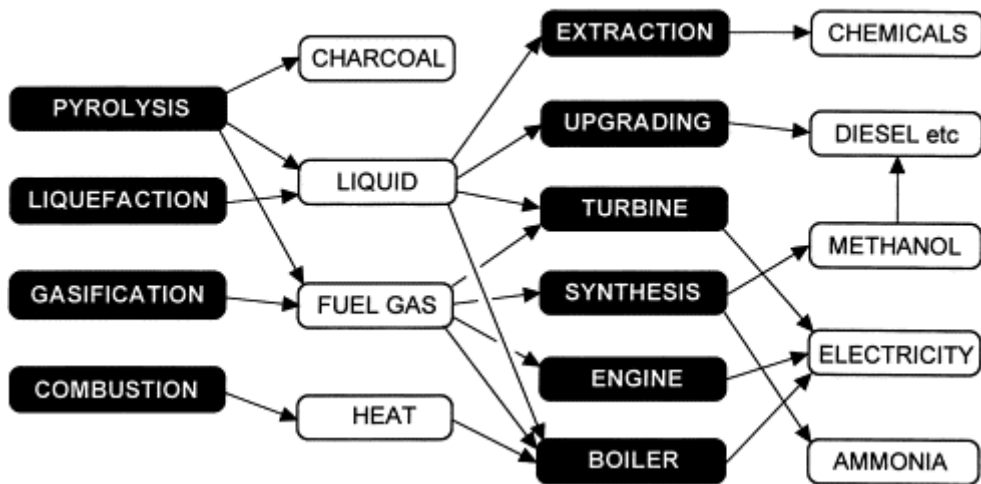


Fig. 1.3.2 Thermochemical biomass processes and products [6,7] .

In conventional heating, heat energy is transferred to the sample section through convection, conduction, and radiation from the surface of the reaction vessel. Thus, a large amount of energy is needed to raise the sample temperature, indicating that conventional heating is inefficient [13]. Moreover, the electrification of materials processing has been needed [14–16]. In response to these situations, microwave heating has attracted attention. Amini et al. indicate that the energy consumption of the microwave process can be reduced compared to the conventional process [17]. Parvez et al. report that the energy efficiency of the microwave process is higher than that of the conventional process [18]. The next section describes the principle of microwave heating and the research trend of microwave biomass conversion.

1.4 Microwave heating of biomass

1.4.1 The principle of microwave heating

Microwave irradiation is electromagnetic irradiation in the range of wavelengths from 0.001 to 1 m and a corresponding frequency range of 0.3 to 300 GHz. Generally, radar transmissions use wavelengths between 0.01 and 0.25 m, and telecommunications use the remaining wavelengths. Microwave ovens operate at 2.45 GHz frequency, which corresponds to a wavelength of 0.122 m. The wavelength of microwaves is equivalent to Wireless Fidelity (Wi-Fi). The leakage must be prevented in microwave heating equipment so as not to interfere with Wi-Fi and other radio waves and avoid adverse effects on the human body. The wavelength of the microwave region in the electromagnetic spectrum is shown in Fig. 1.4.1.

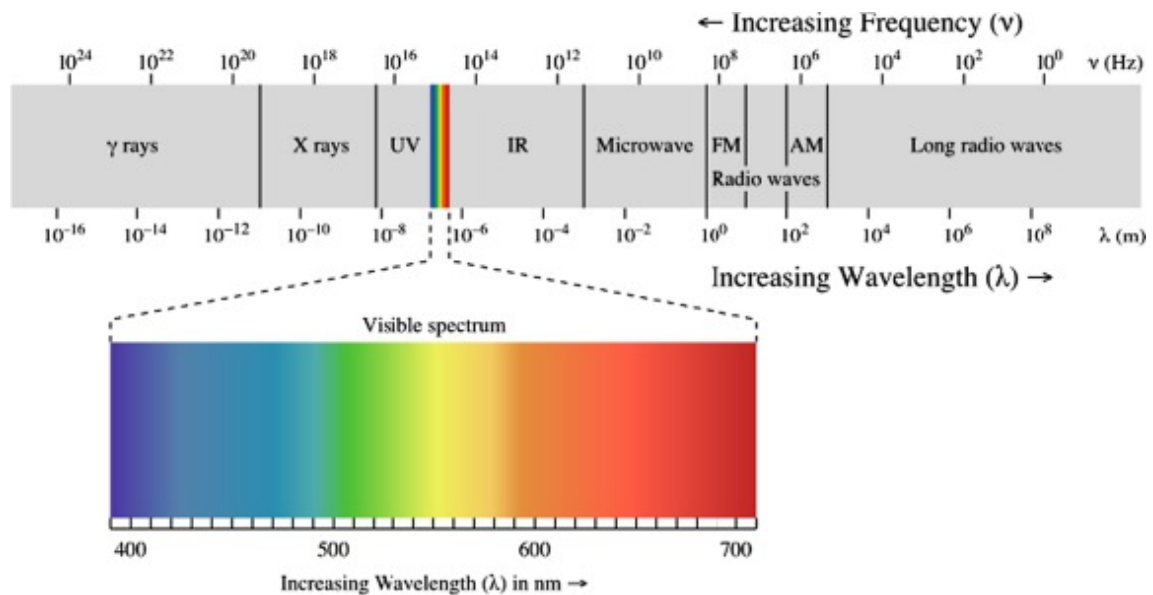


Fig. 1.4.1 The wavelength of each electromagnetic wave [19].

The principle of heat generation in dielectrics is due to the frictional heat generated by polarization. Polarization depends on frequency, and the type of polarization changes with increasing frequency: dipole rotation (orientational polarization), ionic polarization, and electronic polarization. The microwave region responds to orientational polarization. Dielectric material does not contain many free electrons inside but many dipoles with equal positive and negative charges on both sides of the molecule. When the direction of the external electric field changes, dipoles change the axial direction of the array without changing the mean position. In this case, friction and collision cause heat to be generated. If the dipole oscillation does not keep pace with the change in polarity of the electric field, the difference becomes a dielectric loss, inducing a heat-generating effect due to dielectric loss. The microwave energy absorption (P : W/m^3) of material is described in the following equations:

$$P = \pi f \varepsilon_0 \varepsilon_r |E|^2 + \pi f \mu_0 \mu_r |H|^2 + \frac{1}{2} \sigma |E|^2 \quad (1.4.1)$$

Where f , ε_0 , ε_r , E , μ_0 , μ_r , H , and σ are microwave frequency (Hz), vacuum permittivity (F/m), relative dielectric constant, electric field (V/m), vacuum permeability (H/m), relative permeability constant, magnetic field (A/m), and conductivity (S/m). The first, second, and third terms represent dielectric, magnetic, and joule heating, respectively. Considering the dielectric heating, Eq. (1.4.1) can be converted to Eq. (1.4.2) using $\tan\delta$.

$$P = K \varepsilon_r \tan\delta f |E|^2 \quad (1.4.2)$$

Where K and δ are the constant value (0.556×10^{-10}) and dielectric loss angle, respectively. Eq. (1.4.2) indicates that material with a high $\tan\delta$ is easily heated by microwave. Biomass is challenging to heat by microwave due to its low $\tan\delta$. In the microwave heating of biomass, a susceptor is added to it. The $\tan\delta$ of several materials are listed in Table 1.4.1.

Table 1.4.1 Dielectric constant (ϵ'), dielectric loss (ϵ''), and loss tangent ($\tan \delta$) of various materials

Material	ϵ'	ϵ''	$\tan \delta$	Ref.*
Quartz	3.8	0.0001	0.00006	[20]
Lignin	2.6 ± 0.2	0.05 ± 0.01	~ 0.019	[21]
Cellulose	4.5 ± 0.1	0.16 ± 0.02	~ 0.035	[21]
Hemicelluloses	6.4 ± 0.1	0.4 ± 0.03	~ 0.062	[21]
Water	80.4-80.7	9.89-11.9	0.123-0.147	[22]
Carbon black	-	-	0.35-0.83	[23,24]
Bio-solid char	9.60	9.95	1.04	[25]
Activated carbon	-	-	0.57-0.80	[26]
SiC nanotube	-	-	0.58-1.00	[27]
Graphite powder	-	-	0.36-0.67	[28]
Metal oxide	-	-	0.001-0.05	[29]

Microwaves can heat the target material volumetrically. Fig. 1.4.2 shows the difference in the heating mechanism between conventional and microwave heating. The microwave penetration depth is an important factor in evaluating internal heating. The penetration depth of a microwave is defined as the distance from the surface at which the microwave power decreases by $1/e$ relative to the microwave power at the surface. The penetration depth of the microwave power in a material can be calculated below [30].

$$d = \frac{c}{2\pi f \sqrt{2\varepsilon' \{ \sqrt{1 + (\tan\delta)^2} - 1 \}}} \quad (2.26)$$

Where c , f , and ε' are the speed of light (3.0×10^8 m/s), the frequency of a microwave (2.45 GHz), and dielectric constant of the material, respectively.

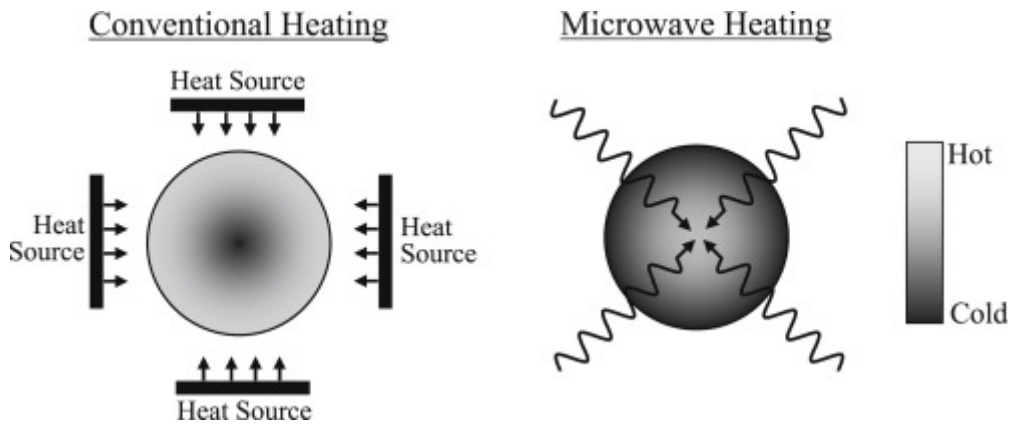


Fig. 1.4.2 Difference of heating mechanism in conventional and microwave heating [31].

1.4.2 Microwave susceptor

Microwave susceptors are heated by microwaves more easily than biomass materials. Thus, microwave susceptors are selectively heated by microwaves and transfer heat energy to biomass. As a result, biomass is heated indirectly at high temperatures. Fig. 1.4.3 shows the diagram of the microwave heating of biomass with susceptors [32]. In the case of without susceptor, biomass absorbs microwave power, but the amount of absorbed energy is quite low due to low $\tan\delta$. On the contrary, the susceptor absorbs microwave power and achieves high temperature. After that, the high temperature susceptor contacts with biomass and transfer heat to biomass. Finally, biomass is heated and decomposed. There are several candidate susceptors, such as carbon-based material, metal oxides, etc.

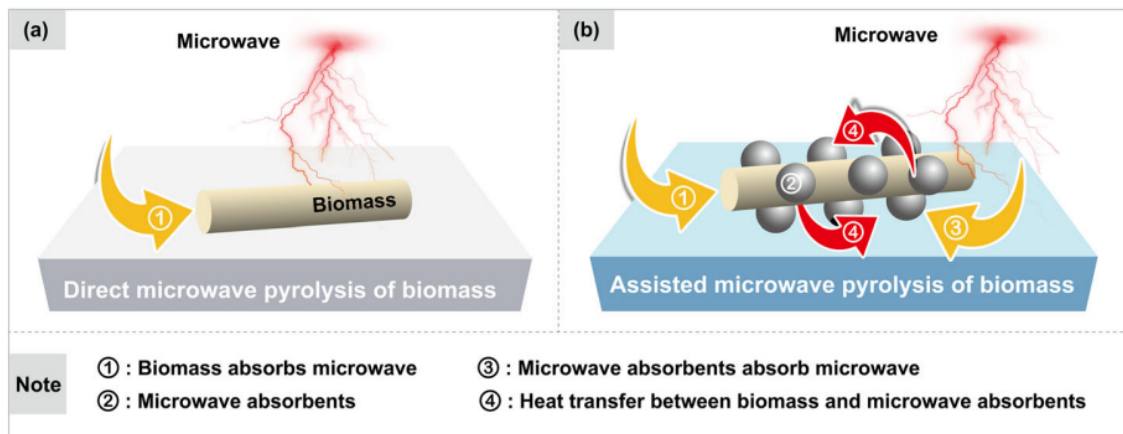


Fig. 1.4.3 Diagram of the microwave heating of biomass (a) without susceptor and (b) with susceptor [32].

1.4.2.1 Carbon-based material

Table 1.4.1 shows that the $\tan\delta$ of carbon-based materials such as carbon black, bio-solid char, activated carbon, SiC nanotube, and graphite powder is higher than that of water. This indicates that carbon-based materials are heated by microwave easily. Ellison et al. investigated the microwave-assisted pyrolysis of biomass (pine sawdust) with biochar (carbon-based material) [33]. A FLIR infrared camera measured the sample temperature through viewports on the top of the waveguide. The sample temperature did not increase, only reaching 200 °C even at 1500 W power when biochar was not added to the pine sawdust. On the other hand, adding biochar increased the sample temperature (600 °C) and accelerated the biomass pyrolysis, increasing non-condensable gas yield. They indicate that adding biochar is effective in promoting biomass pyrolysis. Bu et al. studied the phenol and phenolics production from lignocellulosic biomass by catalytic (activated carbon, GAC830 plus) microwave pyrolysis [34]. An infrared thermometer measured the sample temperature. The high concentrations of phenol (38.9 %) and phenolics (66.9 %) were obtained at the temperature of 316 °C when the catalyst-to-biomass weight ratio of 3:1, indicating the addition of susceptor (activated carbon) is useful to improve the phenol and phenolics yield. This indicates that the susceptor is effective in catalytic pyrolysis of lignocellulosic biomass for phenol and phenolic products. They mentioned that the higher temperature (more than 316 °C) was ineffective in phenol and phenolic production because the gas yield increased with the increase in the temperature. Mamaeva et al. studied the phenolic-rich bio-oil production from lignocellulosic biomass (peanut shell and pine sawdust) with lignite char and activated carbon by microwave-assisted catalytic pyrolysis [35]. The sample temperature was measured by a thermocouple inserted into the sample from the top of the reaction vessel. The sample temperature did not increase, only reaching 200 °C when biochar was not added to the pine sawdust. On the other hand, adding activated carbon or lignite char can increase the sample temperature to 600 °C. They demonstrated that lignite char and activated carbon are useful susceptors to increase the sample temperature.

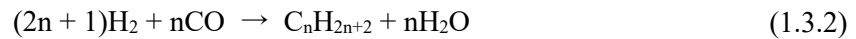
1.4.2.2 Metal oxide

Table 1.4.1 shows that the $\tan\delta$ of metal oxides is lower than that of carbon-based materials. On the other hand, several studies have been conducted on the microwave heating of

biomass with metal oxides. Liu et al. studied the catalytic pyrolysis of biomass (food waste) by microwave heating [36]. MgO, Fe₂O₃, and MnO₂ were used as a susceptor. A thermocouple measured the sample temperature. They found that the maximum temperatures of MgO and MnO₂ were 900 and 700 °C, respectively, higher than those without a susceptor (600 °C). Moreover, MgO and MnO₂ enhance the gas yield and reduce the oil yield because the catalytic cracking of hydrocarbons in bio-oil vapors upon the metal oxide progresses as following reaction [37].



where C_nH_m and C_xH_y represents hydrocarbons in bio-oil vapors and the lighter hydrocarbons, respectively. Longjun et al. investigated the influence of microwave power, metal oxides, and metal salts on the pyrolysis of biomass (algae) [38]. MgO, CaO, and CuO were used as a susceptor. A thermocouple measured the sample temperature. They found that the maximum temperature of CuO was 700 °C higher than that of without a susceptor (600 °C). In addition, CuO increased the gas yield and reduced the solid yield. This indicates that the high temperature would transform micropores into mesopores, making the gas product overflow easier [39]. Huang et al. investigated the catalytic and atmospheric effects on microwave pyrolysis of biomass (corn stover) [40]. NiO, CuO, CaO, and MgO were used as a susceptor. A thermocouple measured the sample temperature. The maximum temperatures of NiO, CuO, and CaO were 510, 500, and 510 °C, respectively, higher than those without a susceptor. Moreover, NiO, CuO, and CaO increase the oil yield and decrease the gas yield. They indicate that the Fischer-Tropsch (as the following reaction) synthesis progresses.

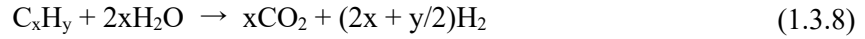
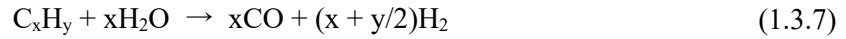
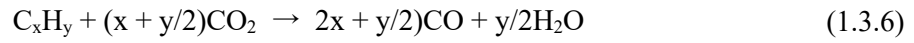
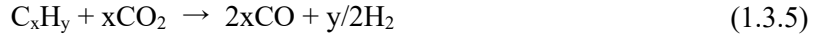


Yu et al. investigated the influence of catalyst types on the microwave-induced pyrolysis of biomass (sewage sludge) [41]. CaO, CaCO₃, NiO, Ni₂O₃, Al₂O₃, and TiO₂ were used as a susceptor. A thermocouple measured the sample temperature. The maximum temperature of each susceptor except for CaO was higher than that of without a susceptor (450 °C). The maximum temperature of CaO was 400 °C lower than that of another susceptor. On the other hand, the H₂ yield of CaO was higher than that of another susceptor. They indicate that CaO enhances the catalytic cracking of hydrocarbons (described following the reaction) and improves the H₂ yield.

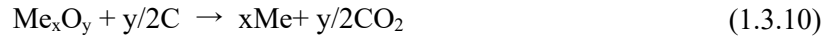


Zhang et al. studied the high-quality syngas production from microwave pyrolysis of biomass

(rice husk) with char-supported metallic catalysts [42]. A rice husk char-supported catalyst (Ni/RHC, Fe/RHC, Cu/RHC) was prepared by incipient wetness impregnation and used as a susceptor. A thermocouple measured the sample temperature. The maximum temperature of susceptor was higher than that of those without a susceptor. Moreover, susceptor increase the gas yield and decrease the solid yield compared to those without a catalyst. They indicate that susceptor enhance the following reactions.



Moreover, metal oxide and carbon reacted to produce CO or CO₂, described the following reactions.



1.4.3 Challenges

1.4.3.1 Temperature effect

Recently, insights into the effect of a susceptor in microwave heating have been deepened. In particular, the catalytic effect of a susceptor has been investigated, as mentioned above. On the other hand, the temperature effect in the microwave biomass conversion has not been sufficiently investigated. Kappe et al. mention that temperature measurement is important when discussing chemical reactions [43]. Microwave heating may induce selective heating, hot spots, etc, indicating that the temperature distribution is not uniform. Pert et al. reported the thermocouple effects during microwave heating of ceramics (zinc oxide) [44]. They found that the edge of the thermocouple absorbed microwave power, inducing the hot spots inside the ceramic pellet. They also showed the appearance of ceramics after microwave heating (shown in Fig. 1.4.4) to indicate the formation of hot spots. Therefore, the non-uniform temperature distribution should be considered when a thermocouple measures the sample temperature during microwave heating. Instead of the thermocouple, there is an infrared thermometer, which can measure the sample temperature indirectly without interference with microwaves. The present study used a fiber-type infrared thermometer (FTKX, Japan Sensor Co., Ltd., Tokyo, Japan) to evaluate the sample temperature. In addition, an electric furnace heating was performed to investigate the temperature effects. The effect of microwave heating will also be verified by comparing microwave heating and electric furnace heating.

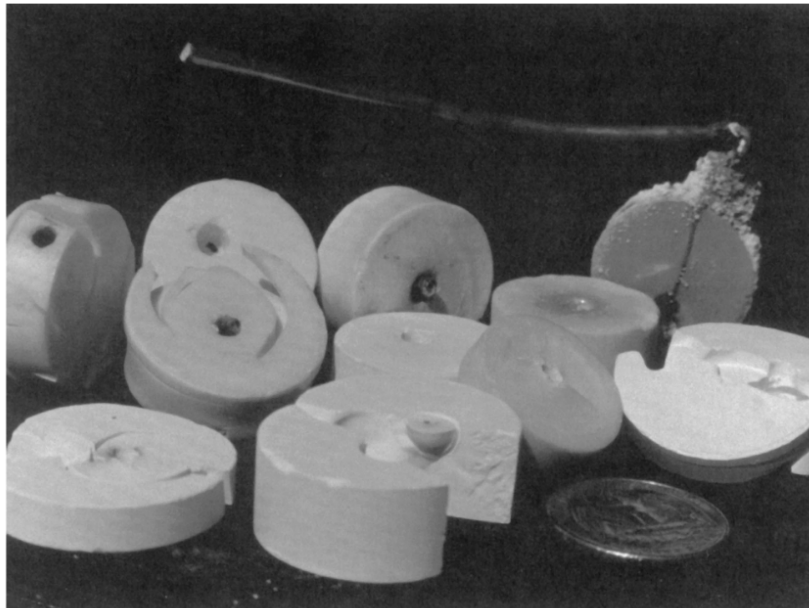


Fig. 1.4.4 Visible evidence of significant problems in zinc oxide samples associated with thermocouples, including carbon spotting, increased densification near the thermocouple tip, and cracking because of large thermal gradients [44].

1.3.3.2 Susceptor for Syngas Production

Studies on microwave biomass conversion using various susceptors have been reported. On the other hand, most studies focus on oil production. Therefore, a susceptor for syngas production has not been sufficiently investigated. The present study focused on magnetite because magnetite can be easily heated by microwave and reach the gasification temperature (more than 600 °C). Moreover, the catalytic effect of magnetite during microwave heating is reported as effective in syngas production [45,46]. However, these studies used the thermocouple to measure the sample temperature (shown in Fig. 1.4.5). Therefore, the discussion on the temperature effect is considered insufficient. In addition, magnetite can be reduced by CO and H₂ from biomass pyrolysis. Thus, the cycle property of magnetite on the syngas production must be considered.

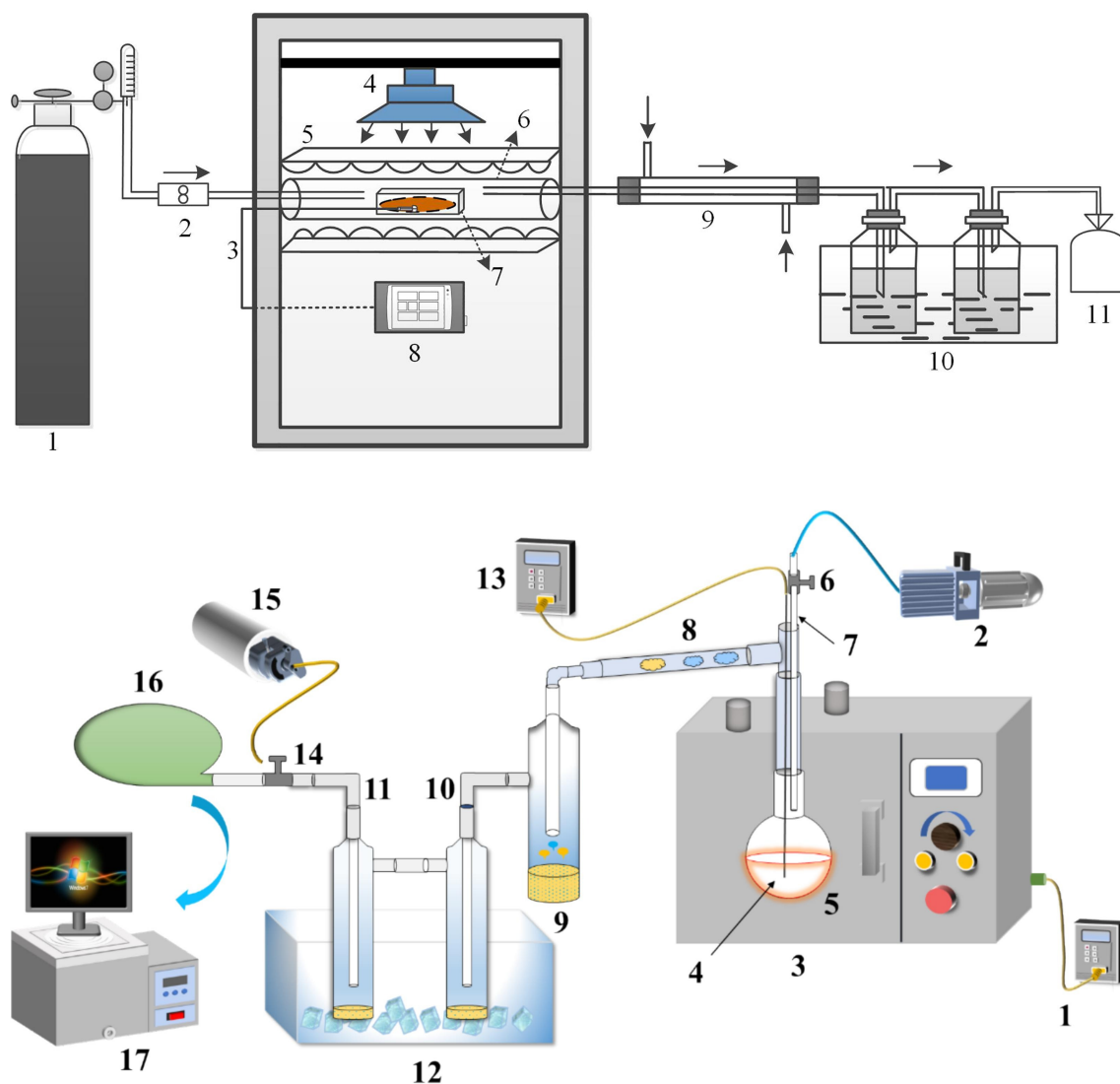


Fig. 1.4.5 The Experiment setup for microwave heating [45,46] (upper side, (1-Carbon dioxide, 2-Gas flowmeter, 3-Thermocouple, 4-Magnetron, 5-Insulation cover, 6-Quartz tube, 7-Samples, 8-Control system, 9-Condensing unit, 10-Liquid product container, 11-Gas collection device)), (lower side, (1-power meter, 2-air compressor, 3-microwave oven, 4-K-type thermocouple, 5-quartz reactor, 6-air flow controller, 7-quartz pipe, 8-straight tubes, (9, 10, and 11-condensing tubes, 12-cooling bath, 13-temperature display, 14-valve, 15-vacuum pump, 16-gas bag, and 17-gas chromatograph)).

1.3.3.3 Techno-economic assessment

Another challenge with the microwave biomass conversion system is that it is still not socially implemented. Therefore, a techno-economic assessment of its social implementation is needed. Moreover, the present study considered the two social problems in Japan. The outline is described in this section. First, a large amount of PV is generated in Kyushu, reducing the frequency quality in the west of Japan. Second, the accumulation of forest residues reduces the quality of the forest environment. The microwave biomass conversion system can utilize excess solar power and treat forest residues. However, the microwave biomass conversion system has not been quantitatively evaluated in terms of how much electricity and forest residue this system can utilize. The present study defines the power conditioning ability, and the utilization rate of forest residues is evaluated when the microwave biomass conversion system is applied in Kyushu, Japan.

1.4 Purpose of this study

The present study aims to clarify the effect of microwaves, a susceptor, and the atmosphere on the microwave heating of biomass for syngas production. Magnetite is used as a susceptor, and cellulose is used to simulate biomass.

Chapter 2 describes the effect of magnetite addition on microwave heating of cellulose for syngas production. The influence of the magnetite-cellulose weight ratio, reusability of magnetite, and microwave heating were experimentally investigated. To evaluate the temperature effect, the electric furnace heating was performed.

Chapter 3 investigates the effect of steam feeding on microwave heating of cellulose for syngas production. To evaluate the temperature effect, the electric furnace heating was performed. The usefulness of microwave heating and electric furnace heating was discussed based on energy efficiency and removing tar,

Chapter 4 performed a techno-economic assessment of the microwave biomass conversion system to verify the feasibility of the investment. In addition, the power conditioning ability and the utilization rate of forest residues are evaluated when the microwave biomass conversion system is applied in Kyushu, Japan.

2. Effect of magnetite addition on microwave heating of cellulose for syngas production

2.1 Introduction

Syngas ($H_2 + CO$) is a key substitute for fossil fuels because it can be converted into various materials, such as ethanol, ammonia, and light hydrocarbons [47–49]. Syngas can also be directly converted into energy using gas turbines [50–52]. Lignocellulosic biomass is a renewable resource used for syngas production and its thermochemical conversion has been widely studied [53–57]. However, conventional heating (CH) processes suffer from challenges such as heat transfer resistance, heat loss to the reaction vessel, non-selective heating, and damage to reactor walls due to external heating [58]. Microwave heating (MWH) has attracted considerable attention to overcome these challenges [32,59–62]. Microwaves can heat the sample section directly, forming a thermal gradient from the center to the outer side [63,64], which is expected to reduce heat loss in the reaction vessel and improve the energy efficiency of syngas production.

Generally, lignocellulosic biomass cannot be easily heated using microwaves because of its low-loss tangent [20]. Thus, a high-loss-tangent material called a susceptor, such as carbon, zeolite, or metal oxide is added. For example, Shang et al. [65] studied the MWH of biomass with susceptors of activated carbon, coke, K_2CO_3 , and NaOH for bio-oil and gas production. They found that the amounts of H_2 , CH_4 , and CO were the highest with an activated carbon susceptor. The activated carbon enhanced the cracking of heavy molecular weight hydrocarbons into small molecular materials. In the case of zeolite, Wang et al. [66] studied the MWH of biomass with zeolite socony mobil-5 (ZSM-5) for phenol and aromatic hydrocarbon production. The yields increased with the addition of ZSM-5, which deoxygenated the oil compounds. Zhang et al. [67] also reported that ZSM-5 mixed with cellulose enhanced decarboxylation and dehydration under MWH, which improved aromatic hydrocarbon and phenol yields. For metal oxides, Huang et al. [68] studied the MWH of biomass with Al_2O_3 for gas production and determined that Al_2O_3 decomposed the product vapor released by biomass pyrolysis into light hydrocarbons (e.g., CH_4 , C_2H_4 , and C_2H_6) and permanent gases (H_2 , CO, and CO_2).

These studies show that microwave susceptors not only function as a heat source, but also enhance biomass decomposition. However, a microwave susceptor that maximizes syngas

production has not yet been identified. Lin et al. [45] reported that the oxygen vacancies in magnetite can decompose volatile organic compounds into H_2 , CO, and light bio-oil during MWH. Magnetite can be easily heated by microwaves because of its high $\tan\delta$ [69]. Therefore, this study focused on using magnetite to improve syngas yield. A limitation of this method is that magnetite is reduced to iron or wüstite when a carbon–magnetite mixture is heated over 800 °C [70,71]. This may cause changes in syngas production properties. Furthermore, the effect of the magnetite amount relative to the biomass amount and MWH on syngas production has not been sufficiently investigated. To this end, this study aimed to clarify the effects of different magnetite–biomass weight ratios and the use of MWH on syngas yield. We also investigate the reusability of magnetite for improved syngas production.

2.2 Materials and Methods

2.2.1 Materials

This study used cellulose as the biomass raw material. Cellulose powder with a particle size of 38 μm was obtained from Fujifilm Wako Pure Chemical Corporation (Osaka, Japan). Its C, H, and O contents were 44.4, 6.2, and 49.3 wt%, respectively. Magnetite powder obtained from Kojundo Chemical Laboratory Co., Ltd., was used as the susceptor. Its particle size was approximately 1 μm , and its purity was $> 98\%$. The powders of cellulose (10 g) and magnetite (1, 5, or 10 g) were mixed in a glass beaker. The mixture was then placed onto quartz glass wool in a tube (inner diameter: 36 mm). Quartz glass balls (40 g, diameter: 5 mm) were mixed with cellulose and magnetite powders to maintain gas flow. These were then microwave-heated to investigate the effects of magnetite.

2.2.2 MWH

Fig. 2.1 shows the schematic of the MWH experiment, which was based on the MWH system of a previous study [72]. In this study, steam was not fed into the quartz glass tube during the MWH of cellulose; however, this was done during the steam oxidation experiment (Section 2.3.5). The quartz glass tube (inner diameter: 36 mm, length: 700 mm) was installed in the microwave cavity, and the sample section was surrounded by a thermal insulator with internal walls coated with SiC. Thus, the sample was heated by microwaves and the radiant heat of SiC. High-purity Ar gas was fed into the quartz glass tube for air purging. The flow rate of Ar gas was adjusted to 500 scc/min using a mass flow controller (Model 3200, KOFLOC Corp.).

Four magnetrons (2.45 GHz, UM-1500CS-A, Micro Denshi Co., Ltd.) and two stirrers were turned on to begin the MWH experiments. A power meter (PM10-DM, Micro Denshi Co., Ltd.) was used to measure the input and reflection power of the microwaves generated by each magnetron. The input power of each magnetron was set at approximately 200 W (the total input power was $200\text{ W} \times 4 = 800\text{ W}$) to increase the sample temperature, which was measured as the surface temperature of the sample using a fiber-type infrared thermometer (FTKX, Japan Sensor Co., Ltd., Tokyo, Japan) with a measurable range of 180–2000 $^{\circ}\text{C}$. After the sample temperature reached 800 $^{\circ}\text{C}$, the output power of each magnetron was maintained below 200 W to maintain

this temperature.

Oil products were condensed in a water-cooling section (5 °C, glass container 1) and two cold traps (−45 °C, glass containers 2 and 3). Gas chromatography (GC) (310C, SRI) analyzed the gaseous products using a Hayesep-D column. The operating conditions were: feeding volume of 1 mL, column temperature of 40 °C, thermal conductivity detector measurement temperature of 150 °C, and carrier gas of high-purity Ar with a flow rate of 20 mL/min. A standard gas mixture (containing 0.1 % H₂, CH₄, CO, and CO₂ in Ar) was used to calibrate H₂, CH₄, CO, and CO₂ in the non-condensable gaseous product.

The microwave heating experiment was performed repeatedly until the syngas yield remained stable to verify the reusability of magnetite. For the second and subsequent cycles, 10 g of cellulose was added to the residue from the previous cycle, and MWH was performed from room temperature (25 °C) to 800 °C for 1 h. For comparison, CH experiments were also performed in this study. Here, the thermal insulator was replaced by a ceramic electric ring furnace (ARF-50KC, Asahi Rikagaku). A K-type thermocouple was used to measure the center temperature of the samples.

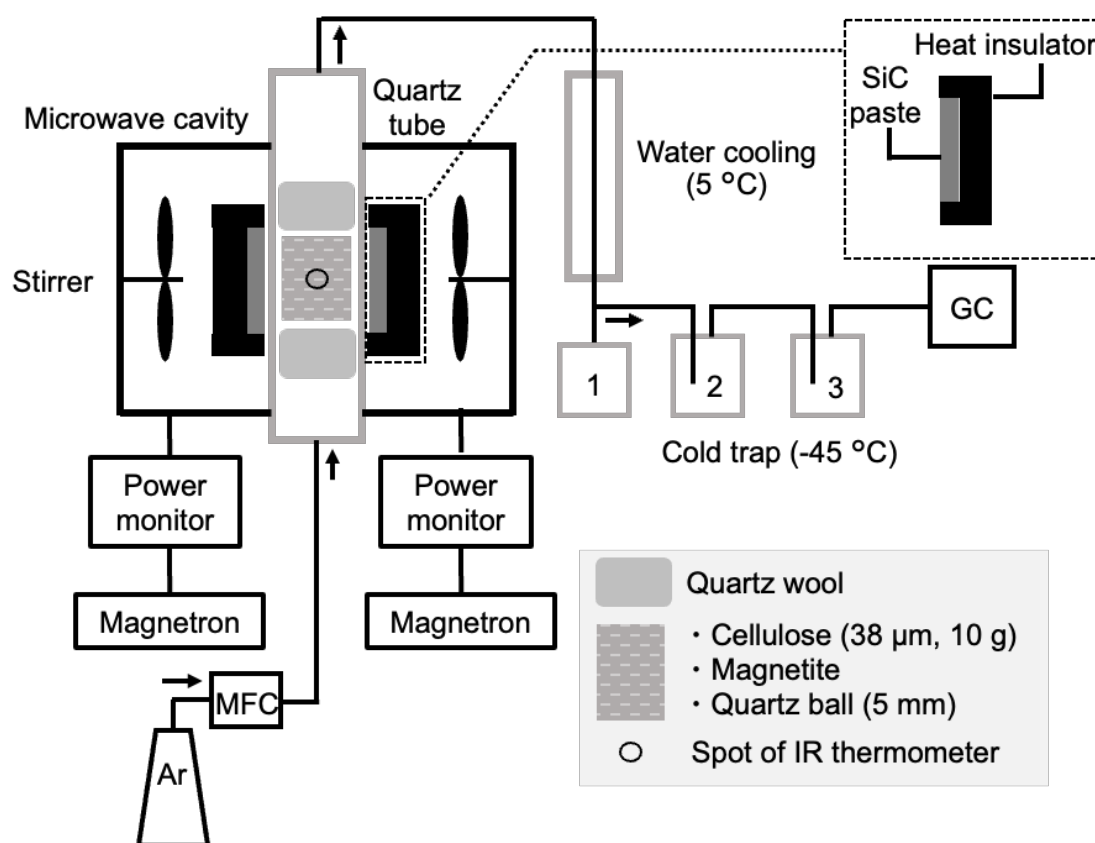


Fig. 2.1 Schematic of the MWH experimental setup. MFC and GC denote the mass flow controller and gas chromatography, respectively.

2.3 Mass balance calculation

The gas (Y_{gas}), solid (Y_{solid}), and oil (Y_{oil}) yields (wt%) were calculated as follows:

$$Y_{\text{gas}} = \frac{M_{\text{gas}}}{M_{\text{cellulose}}} \times 100 \text{ (wt\%)}, \quad (2.1)$$

$$Y_{\text{solid}} = \frac{M_{\text{cellulose}} - M_{\text{diff}}}{M_{\text{cellulose}}} \times 100 \text{ (wt\%)}, \quad (2.2)$$

$$Y_{\text{oil}} = 100 - Y_{\text{gas}} - Y_{\text{solid}} \text{ (wt\%)} \quad (2.3)$$

where M_{gas} and $M_{\text{cellulose}}$ represent the masses of gas and cellulose, respectively, and M_{diff} represents the difference between the weights of the reaction vessel before and after the experiments, considering magnetite reduction. The weight reduction of magnetite is 2.7 g when magnetite was reduced to iron.

2.4 Magnetite powder characterization

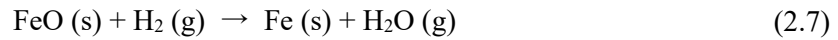
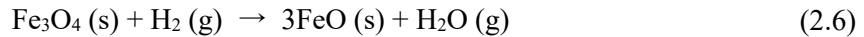
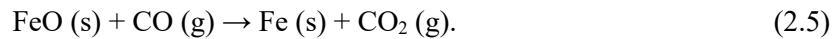
After the experiments, the solid residue was recovered from the quartz glass tube. X-ray diffraction (XRD) analysis of the solid residue was conducted using a RINT TTR-III (Rigaku) instrument with Co-K α characteristic X-ray in the 2θ range of 15–90 ° at intervals of 0.02°.

2.3 Results and Discussion

2.3.1 Effect of magnetite–cellulose weight ratio

Fig. 2.2 shows the mass balance and gas yield for different weight ratios of the mixture. The oil yield decreased by 5.0 %, and the gas yield increased by 3.5 % when the weight ratio of the mix increased from 0 to 0.1. At this time, the magnetite addition caused no significant changes in the syngas yield. In contrast, the solid yield decreased by 10 %, and the gas yield increased by 10 % when the weight ratio of the mixture was increased from 0.1 to 0.5. The CO yield increased by 3.2 mmol/g-cellulose, while CO₂ remained constant.

Fig. 2.3 shows the XRD patterns before and after heating for different weight ratios of the mixture. Magnetite was reduced to iron (α Fe) by the CO and H₂ generated by the pyrolysis of cellulose. The magnetite reduction reaction is as follows:



The CO₂ generation increased with the amount of magnetite added. However, the CO yield increased, whereas CO₂ remained constant when the weight ratio increased from 0.1 to 0.5. These results indicate that the addition of magnetite enhanced the Boudouard reaction (Eq. (2.8)).



Graphitic C forms in the solid when cellulose is pyrolyzed above 600 °C [73]. Therefore, the magnetite addition caused the Boudouard reaction and improved the CO yield when the weight ratio of the mixture was 0.5.

When the magnetite–cellulose weight ratio increased from 0.5 to 1.0, the oil yield decreased by 35 %, and the gas yield increased by 32 % (Fig. 2.2 (a)); the H₂, CO, and CO₂ yields increased by 2.2, 5.9, and 3.2 mmol/g-cellulose, respectively (Fig. 2.2 (b)). Volatiles generated by cellulose pyrolysis are either condensed to produce oil or decomposed to produce light hydrocarbons and permanent gases [74–77]. The addition of magnetite caused the decomposition of volatiles and improved the H₂ and CO yields with a weight ratio of 1.0, in which the maximum value of the syngas yield was 19.1 mmol/g-cellulose. Thus, the subsequent experiments used the

magnetite–cellulose weight ratio 1.0, where syngas yield was the maximum.

The hydrogen (X_H) and carbon (X_C) gasification rates (%) are defined as follows:

$$X_H = \frac{\text{Total mole of H contained in H}_2}{\text{Theoretical mole of H in cellulose}} \times 100 (\%) \quad (2.9)$$

$$X_C = \frac{\text{Total mole of C contained in CH}_4, \text{ CO, and CO}_2}{\text{Theoretical mole of C in cellulose}} \times 100 (\%) \quad (2.10)$$

X_H and X_C were 16.0 % and 63.5 %, respectively, when the weight ratio was 1.0.

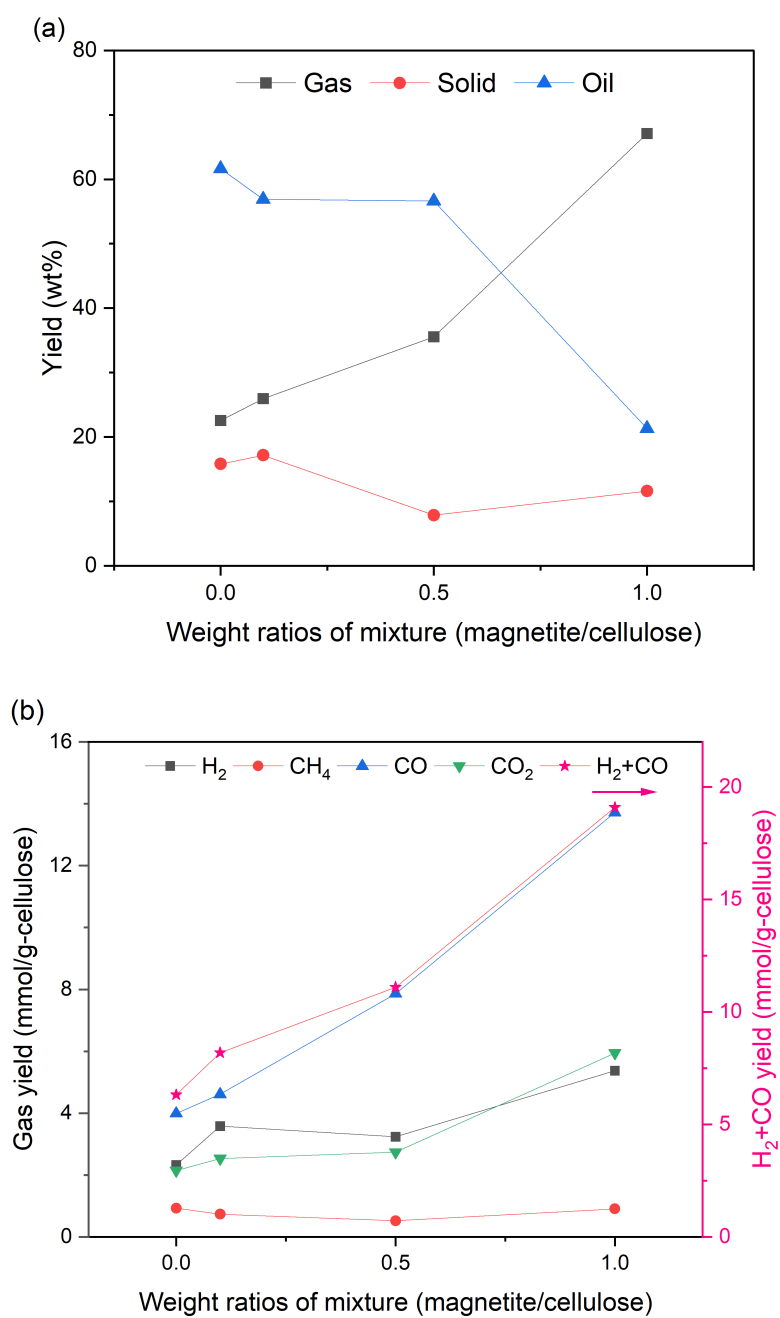


Fig. 2.2 Effect of magnetite–cellulose weight ratio on the (a) mass balance and (b) gas yield of cellulose MWH at 800 °C.

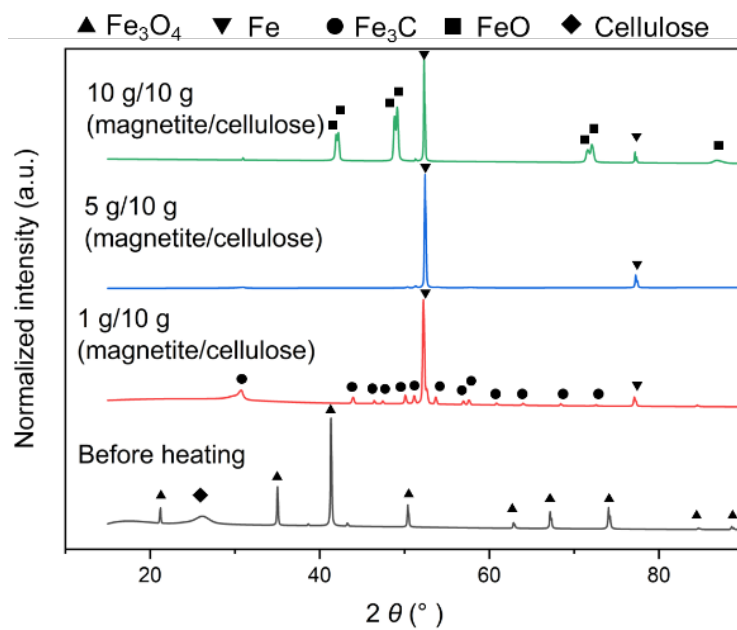


Fig. 2.3 XRD patterns of the samples before and after heating (Co-K α) for different magnetite–cellulose mixing ratios.

2.3.2 Comparison of CH and MWH

Fig. 2.4 (a) shows the gas yields obtained using different heating methods; the left side of the dashed line represents the results of CH, specifically electric furnace heating, and the right side shows those of MWH. CH800 and CH1000 indicate that the temperatures measured by the K-type thermocouple at the center of the sample are 800 °C and 1000 °C, respectively. MWH800 represents that the surface temperature measured by the fiber-type infrared thermometer is 800 °C.

When magnetite was not added to the cellulose, the gas yields of CH800 and MWH800 were similar. On the other hand, when magnetite was added, the H₂, CO, and CO₂ yields of MWH800 were 3.1, 7.2, and 2.9 mmol/g-cellulose, higher than those of CH800, respectively. A region higher than the surface temperature, called internal heating, may occur inside the sample because MWH can induce internal heating. The temperature measurement points differed between CH and MWH. In the case of CH, the K-type thermocouple was used to measure the temperature at the center of the sample. For MWH, the fiber-type infrared thermometer was used to measure the surface temperature of the sample. Therefore, the temperature inside the sample was not measured, and internal heating may have occurred during MWH.

The gas yield of CH1000 was investigated to estimate the internal temperature of MWH800. The analysis revealed similar gas yields for MWH800 and CH1000, which suggests that a temperature region of 1000 °C occurs inside the sample under MWH800. Therefore, the addition of magnetite may have caused internal heating.

Energy efficiency (η_{gas}) was then calculated (Eq. (2.11)).

$$\eta_{\text{gas}} = \frac{H_{t\text{-gas}}}{W_t} \times 100 \quad (\%), \quad (2.11)$$

where H_{gas} (J) and W_t (J) are the lower heating values of the gaseous products (H₂, CH₄, and CO) and input energy for the experiments, respectively. These were integrated up to each GC measurement time. $H_{t\text{-gas}}$ was calculated as follows:

$$H_{t\text{-gas}} = 120 \times M_{\text{H}_2\text{-t}} + 50 \times M_{\text{CH}_4\text{-t}} + 10 \times M_{\text{CO-t}} \quad (\text{MJ}), \quad (2.12)$$

where $M_{\text{H}_2\text{-t}}$, $M_{\text{CH}_4\text{-t}}$, and $M_{\text{CO-t}}$ (g) represent the H₂, CH₄, and CO yields at each GC measurement time considering heating time, respectively. The W_t of the CH ($W_{t\text{-CH}}$) and MWH ($W_{t\text{-MWH}}$) represent the Joule heat for the electric furnace and the difference in the microwave input energy

and reflection energy obtained from the power monitor, respectively.

The energy efficiency of microwave oscillations in a magnetron is 60–90 %. In the present study, the energy efficiency of microwave oscillations in a magnetron was not considered. Figs. 2.4 (b) and 2.4 (c) show that CH1000 and MWH800, whose gas yields were comparable (Fig. 2.4 (a)), were energy efficient when magnetite was added to cellulose. The maximum η_{gas} of CH1000 and MWH800 were 3.4 % and 5.3 %, respectively, in 1842 and 1281 s heating times. Therefore, MWH800 produced syngas more efficiently than CH1000. These results indicate that magnetite addition causes internal heating during MWH, improving the energy efficiency of syngas production.

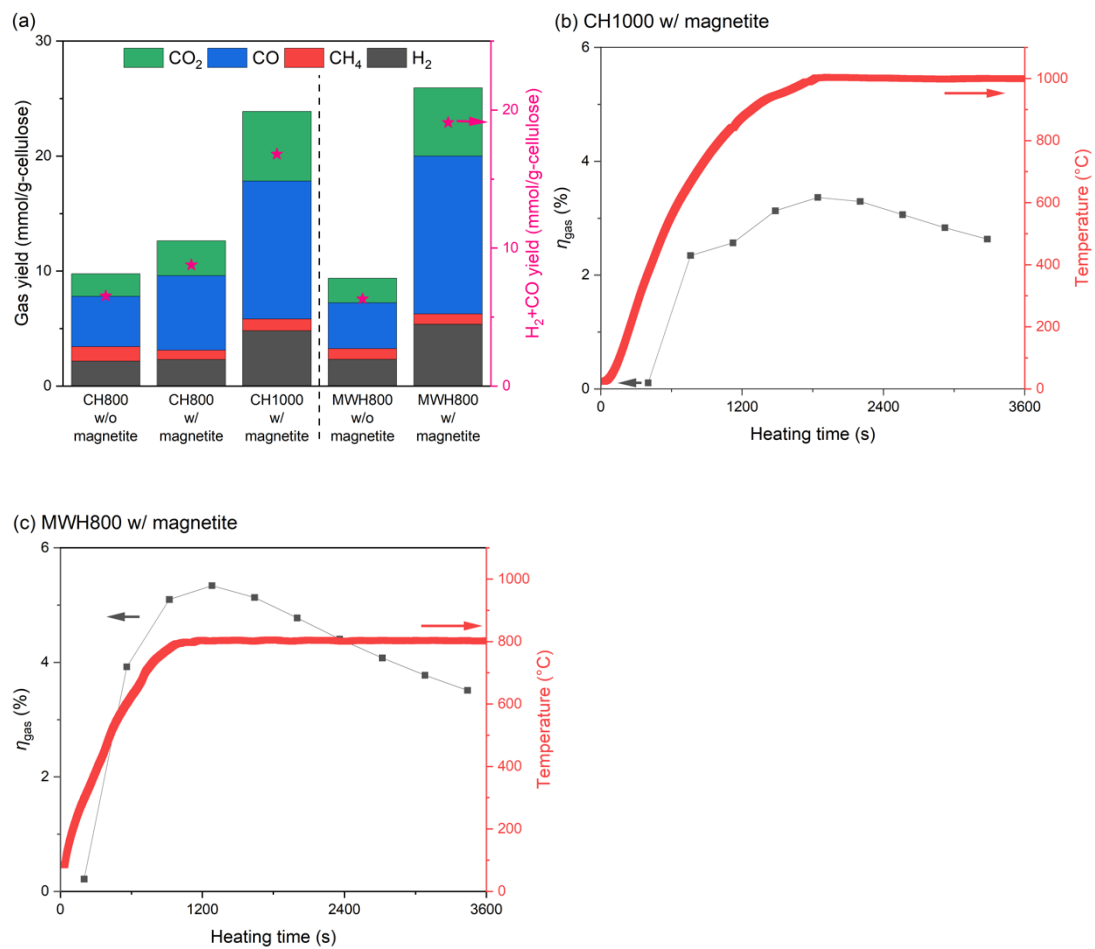


Fig. 2.4 Comparison of (a) CH and MWH gas yields (w/: with, w/o: without). Energy efficiencies of (b) CH1000 and (c) MWH800 with magnetite.

2.3.3 Cycle properties

The syngas yield ($H_2 + CO$) was measured for every cycle to investigate the reusability of the microwave susceptor. Fig. 2.5 shows the mass balance and gas yield for each cycle. The result for the 1st cycle was the same as that for a weight ratio of 1.0, as shown in Fig. 2.2 (b). From the 1st to the 2nd cycle, the oil yield increased by 17.7 %, the gas yield decreased by 14.5, and CO and CO₂ yields decreased by 0.9 and 2.7 mmol/g-cellulose, respectively. These results indicate that the decomposition of volatiles was suppressed. Therefore, volatiles condensed to oil in the cold trap system, increasing the oil yield. From the 2nd to the 3rd cycle, the oil and solid yields increased by 13.8 % and 11.5 %, respectively, whereas the gas yield decreased by 25.3 %. The CO and CO₂ yields declined by 7.9 and 0.7 mmol/g-cellulose, respectively. These results indicate that the decomposition of volatiles and the Boudouard reaction were suppressed. After the 3rd and subsequent cycles, the mass balance and gas yield remained constant.

Fig. 2.6 shows the XRD patterns of the initial samples and those after the 1st to 5th cycles. In the 1st cycle, magnetite was reduced to iron (α Fe) and wüstite (FeO) owing to CO and H₂ (Eq. (2.4)–(2.7)). In the 2nd cycle, the FeO peak disappeared, and only the Fe peak was observed, indicating that FeO was reduced by CO and H₂ (Eq. (2.5) and (2.7)). In 3rd cycle, the peak of Fe₃C was observed. This suggests that Fe and graphitic C reacted to produce Fe₃C. In the subsequent cycles, the XRD patterns remained relatively constant. The syngas yield decreased after the 2nd cycle (Fig. 2.5 (b)) likely because the internal heating effect caused to magnetite was not achieved in the subsequent cycles as iron does not absorb microwaves. Amini et al. [78] studied the hydrogen reduction behavior of magnetite under MWH. They found that the formation of metallic iron results in less microwave absorption, which leads to a decrease in the sample temperature during MHW. This means that iron cannot function as a susceptor. Iron should be oxidized to magnetite to obtain a stable syngas yield, as in the 1st cycle. Instead, the cumulative amount of residue increased with the number of cycles. This residue contains graphitic C, which could function as a susceptor because of its high $\tan\delta$. However, the syngas yield of the 5th cycle was similar to that of CH. This suggests that graphitic C does not form an internal heating equivalent to magnetite.

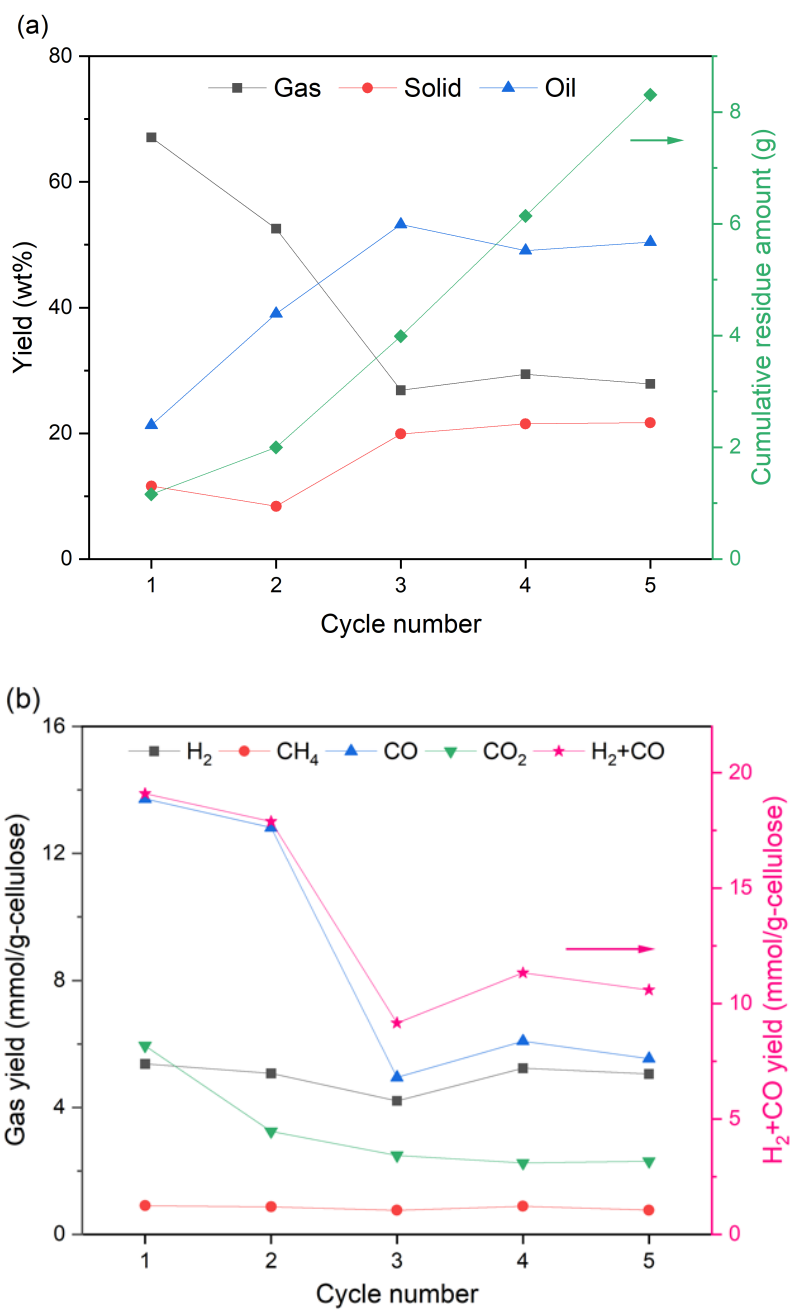


Fig. 2.5 Cycle properties of the (a) product distribution and (b) gas yield for MWH800.

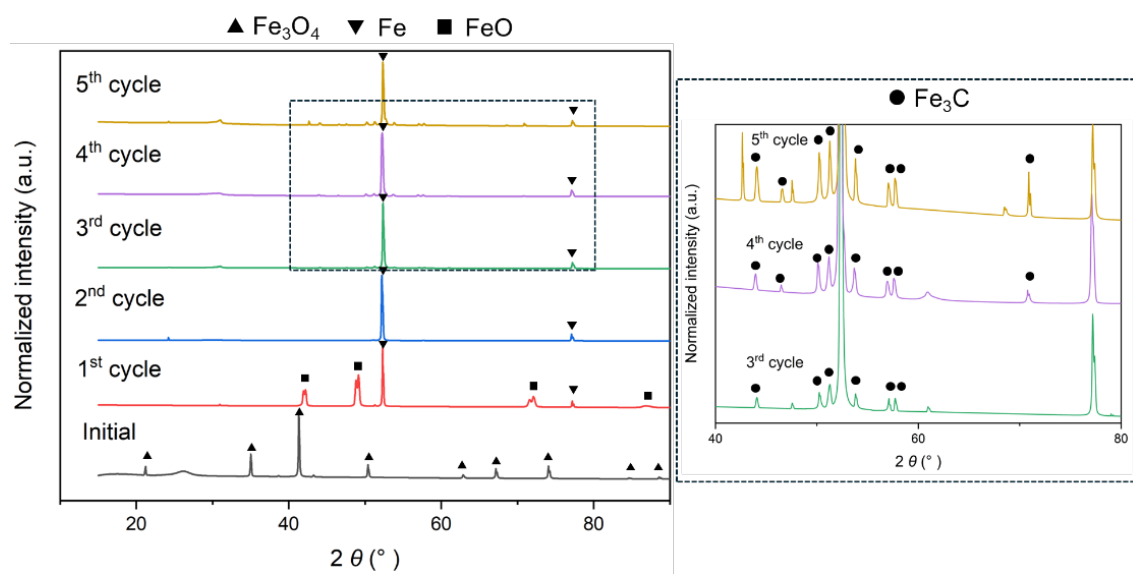


Fig. 2.6 XRD patterns of the initial and subsequent samples after each cycle (Co-K α).

2.3.4 Comparison of magnetite and graphite

The results in Section 2.3.3 suggest that graphitic C functioned as a susceptor after the 2nd cycle. This section verifies the effect of graphite as a susceptor by comparing the syngas yield of MWH800 with graphite and magnetite. Fig. 2.7 (a) compares the yields obtained using different heating methods. The H₂, CH₄, CO, and CO₂ yields were 5.2, 1.1, 5.9, and 3.1 mmol/g-cellulose when graphite (1 μ m, 10 g) and cellulose (38 μ m, 10 g) mixture was microwave heated at 800 °C (MWH800 graphite). These values were similar to the gas yield of the 5th cycle, suggesting that iron no longer absorbs microwaves, whereas graphitic C functions as a susceptor after the 3rd cycle. Moreover, the syngas yield of MWH800 with graphite (hereafter MWH800 graphite) was higher than that of MWH800 without magnetite. This suggests that internal heating may have occurred when the graphite–cellulose mixture was microwave heated. The syngas yields of MWH800 magnetite (5th cycle) and MWH800 graphite were lower than that of MWH800 magnetite (1st cycle).

Saito et al. [79] reported that the internal temperature was lower than the surface temperature when a microwave-heated mixture contained graphite. The results of this study suggest that the internal temperature of the graphite and cellulose mixture was lower than that of the magnetite and cellulose mixture during MWH. Figs. 2.7 (b) and 2.7 (c) show the changes in the energy efficiencies of MWH800 magnetite (5th cycle) and MWH800 graphite over time. The maximum η_{gas} of MWH800 magnetite (5th cycle) and MWH800 graphite were 3.8 % and 3.1 %, respectively, lower than those of MWH800 magnetite (1st cycle). Therefore, the magnetite during the 1st cycle was a better susceptor than graphite in syngas production.

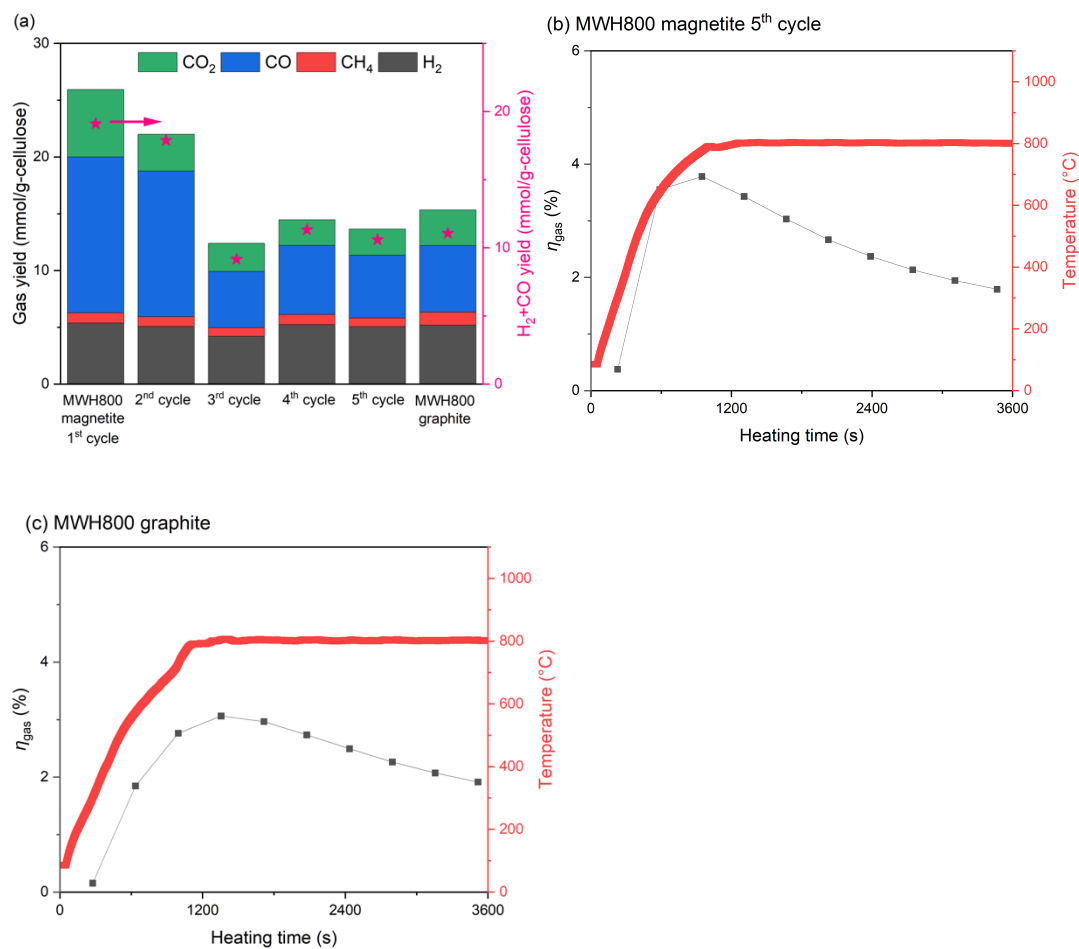


Fig. 2.7 Comparison of (a) CH and MWH gas yields. Energy efficiencies of (b) MWH800 magnetite (5th cycle) and (c) MWH800 graphite.

2.3.5 Reusability of magnetite

Steam oxidation of the 5th cycle residue was performed to regenerate iron into magnetite. The weight of the 5th cycle residue was measured, and its value was 8.25 g. The sample loss of Fe and graphitic C in the cycling experiment occurred at a 1:1 weight ratio, the weight of iron was 3.35 g (60 mmol), and the weight of graphitic C was 4.90 g (408 mmol). This was microwave heated at 800 °C under Ar (30 %) and steam (70 %) for 2 h. Subsequently, the same steam oxidation treatment was applied for another 2 h. The steam oxidation of Fe is expressed as follows:

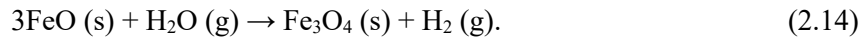
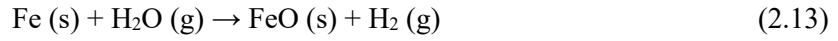


Fig. 2.8 shows the XRD patterns of the initial and steam-oxidized residue. Iron was oxidized to magnetite by steam.

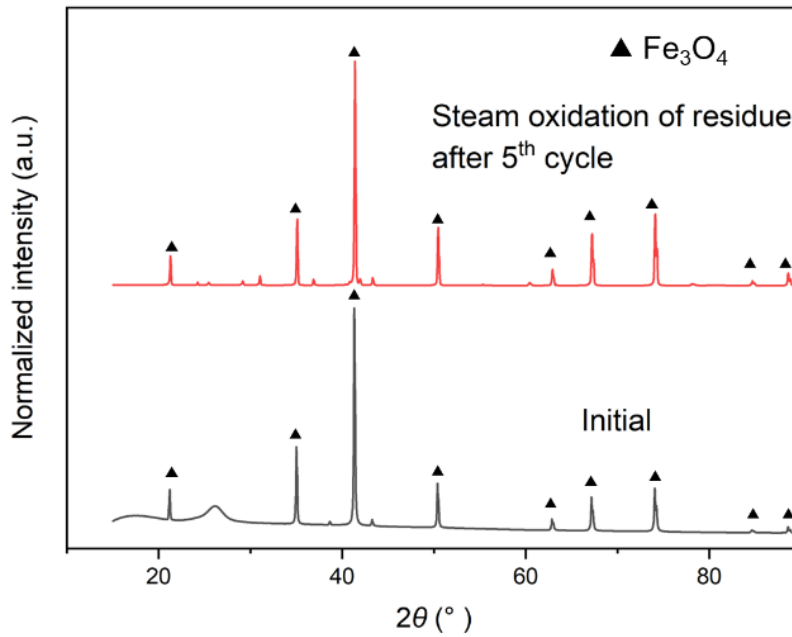
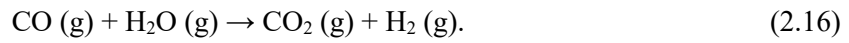
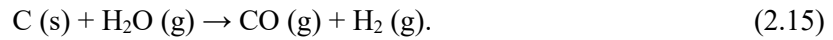


Fig. 2.8 Comparison of the initial sample and steam-oxidized residue after the 5th cycle and as-received.

Fig. 2.9 shows the H₂, CH₄, CO, and CO₂ concentrations in the non-condensable gaseous products under heating. Pineau et al. [80] reported that magnetite forms when $H_2 / (H_2 + H_2O) < 0.3$ at 800 °C. $H_2 / (H_2 + H_2O)$ changed under 0.3, as shown in Fig. 2.9 (c). This indicated that the iron was completely oxidized by steam. The total H₂, CO, and CO₂ yields after steam oxidation of the residue were 544, 196, and 209 mmol, respectively. The total CO and CO₂ were 415 mmol, indicating that the 408 mmol of graphitic C was gasified. Therefore, 600 mmol of H₂ was produced by the steam oxidation of carbon (Eq. (2.15)) and water-gas shift reaction (Eq. (2.16)).



If the graphitic C is gasified, the residue is considered magnetite. After the steam oxidation treatment of the 5th cycle residue, 5 g of magnetite was obtained, and this was used as a susceptor (oxidized magnetite) to investigate the syngas yield.

The syngas yield was the highest when the amount of the susceptor relative to cellulose was 1.0 (Section 2.3.1). Thus, 5.0 g of cellulose was mixed with 5 g of oxidized magnetite. Fig. 2.10 shows that the syngas yield of the 1st cycle magnetite and oxidized magnetite were similar. This suggests that the oxidized magnetite occurs through internal heating to obtain a syngas yield similar to that of the 1st cycle magnetite. Therefore, steam oxidation treatment is effective for reusing magnetite.

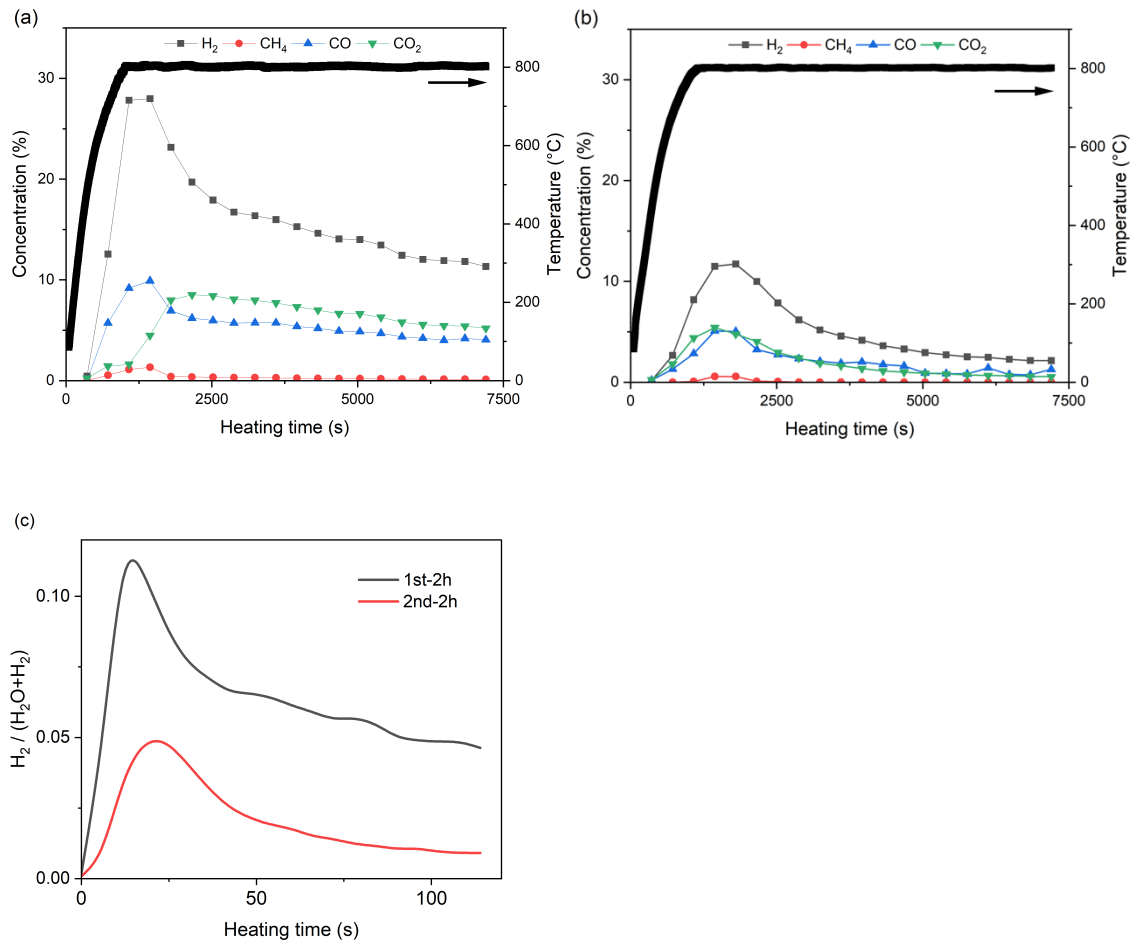


Fig. 2.9 Released gas concentration relative to Ar changes over time of the (a) first and (b) following 2 h of steam oxidation treatment.

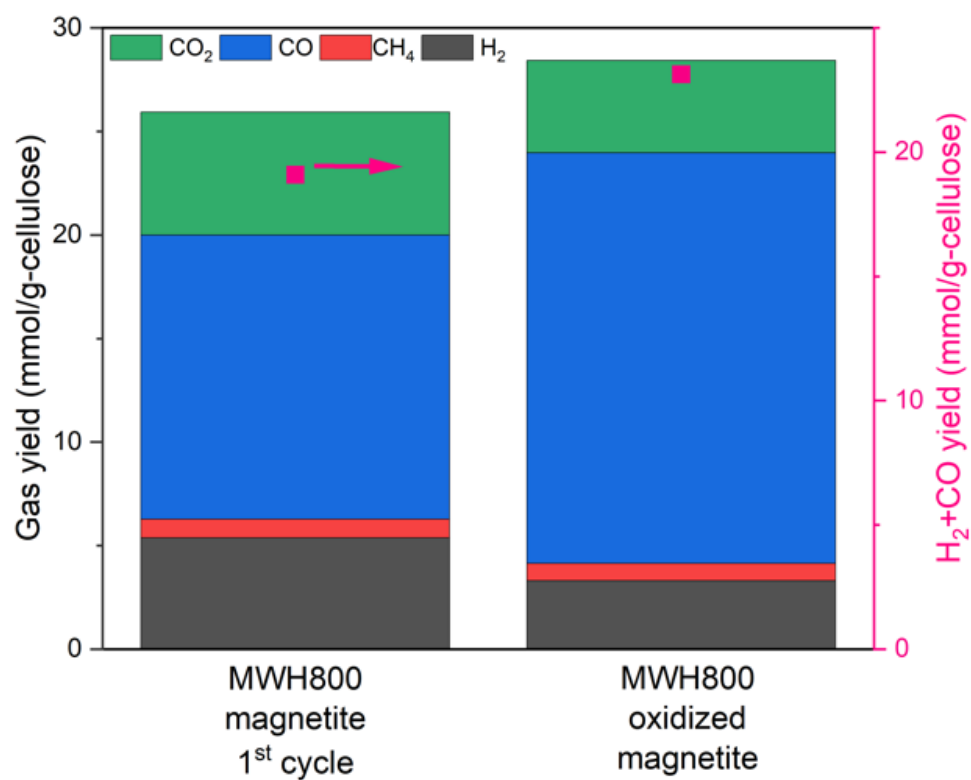


Fig. 2.10 Comparison of the gas yields of MWH800 magnetite (1st cycle) and oxidized magnetite.

2.3.6 Possibility of internal heating

2.3.6.1 Heat transfer equation

The quartz glass tube was used as a reaction vessel. Thus, the heat transfer equation for the cylindrical is considered as below.

$$\rho c \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial r^2} + \frac{k}{r} \frac{\partial T}{\partial r} + Q - q \quad (2.17)$$

where T , ρ , c , k , Q , and q are temperature distribution (K), mass density (kg/m^3), specific heat capacity ($\text{J}/(\text{kg} \cdot \text{K})$), thermal conductivity ($\text{W}/(\text{m} \cdot \text{K})$), volumetric heat (W/m^3), and radiant heat conduction inside the cylindrical material (W/m^3). Fig. 2.11 shows the mixed model of magnetite, graphite, and quartz glass ball after cellulose is pyrolyzed into graphite. Models A and B represent magnetite and graphite used as susceptors, respectively. Graphite after cellulose pyrolysis is considered to avoid overestimation of cylinder center temperature. The packing fraction of the mixture is 50 % based on the actual experimental condition. Fig. 2.12 shows the heat transfer calculation model for estimating temperature distribution inside a radial direction of a cylinder. The heat transfer equation is discretized using the radial finite difference method as below.

$$\frac{T_{i,n+1} - T_{i,n}}{\Delta t} = \frac{k}{\rho c} \left(\frac{T_{i+1,n} - 2T_{i,n} + T_{i-1,n}}{\Delta r^2} + \frac{1}{r_i} \frac{T_{i+1,n} - T_{i-1,n}}{2\Delta r} \right) + \frac{Q}{\rho c} - \frac{q}{\rho c} \quad (2.18)$$

$$q = \frac{\epsilon \sigma}{2\Delta r} (T_{i-1}^4 - T_{i+1}^4) \quad (\text{W}/\text{m}^3) \quad (2.19)$$

where ϵ , σ , and Δr are emissivity, Stefan-Boltzmann constant ($5.67 \times 10^{-8} \text{ W}/(\text{m}^2 \cdot \text{K}^4)$), and mesh size of the radius direction, respectively. In the present study, quartz glass balls whose diameter is 5 mm are used, indicating the radiant heat conduction is on the order of 5 mm. Therefore, Δr is set to 0.006 mm to divide 0.018 m into 4 parts. Δt is the time interval (10 s), and the calculation range of time is from 0 s to 100 s. The initial condition $T(r, 0)$ is 1073 K. The boundary conditions are described as follows:

$$\frac{T_{0,n+1} - T_{0,n}}{\Delta t} = 2 \times \frac{k}{\rho c} \frac{T_{1,n} - T_{0,n}}{\Delta r^2} + \frac{Q}{\rho c} - 2 \times \frac{q}{\rho c} \quad (2.20)$$

$$T(R, t) = 1073 \text{ (K)} \quad (2.21)$$

$$q_{\text{surface}} = \frac{\epsilon \sigma}{\Delta r} (T_{\text{surface}}^4 - T_{\text{env}}^4) \quad (\text{W}/\text{m}^3) \quad (2.22)$$

Where q_{surface} , T_{surface} , and T_{env} are heat radiation from the surface of the cylindrical material, surface temperature of the cylindrical material, and outside temperature (293 K). The volumetric heat of Q is calculated as below.

$$Q = \frac{P}{V} \quad (2.23)$$

Where P and V are the input power into the cylindrical material (W) and the volume of the sample section regarding the packing fraction 50 % (m^3).

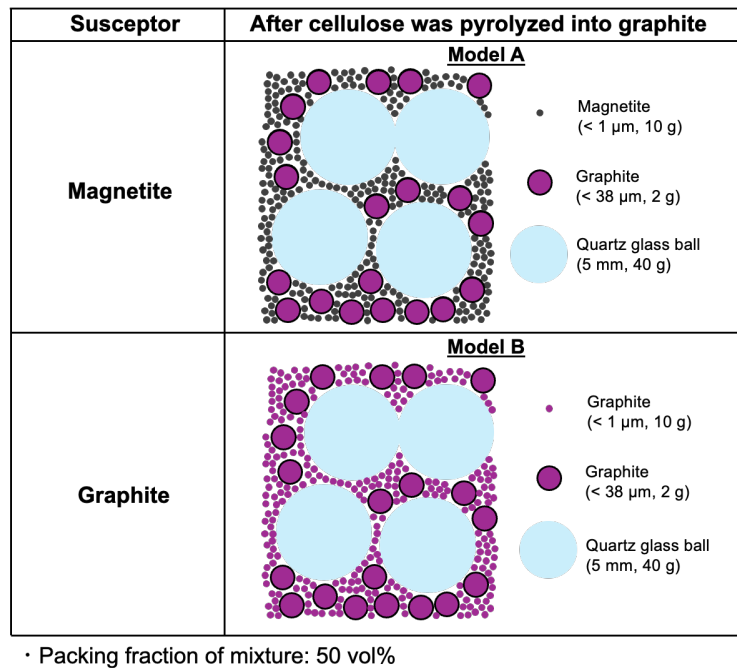


Fig. 2.11 Mixed model of magnetite, graphite and quartz glass ball after cellulose is pyrolyzed into graphite (Model A: Magnetite, Model B: Graphite).

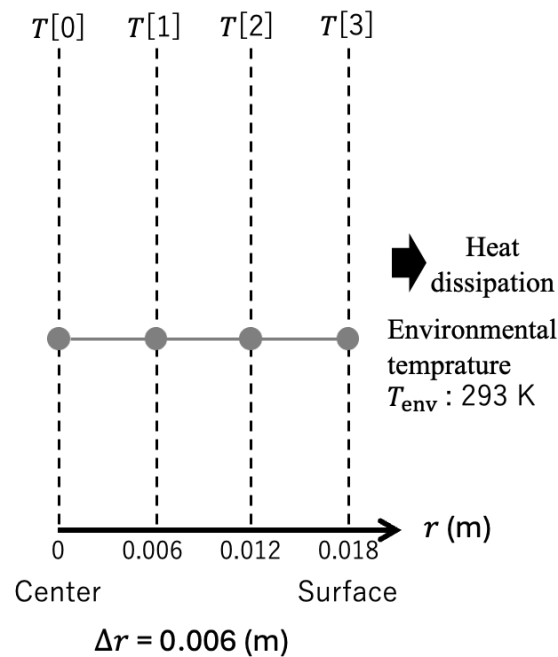


Fig. 2.12 Heat transfer calculation model for estimating temperature distribution inside a radial direction of a cylinder.

2.3.6.2 Defenition of physical peroperty

Table 2.1 summarizes the physical properties of each material, a mixture of Model A and Model B. The thermal conductivity of the mixture is defined as that of graphite because graphite powder ($< 38 \mu\text{m}$) is assumed to be uniformly filled in the voids between the quartz glass balls (5 mm). Therefore, the thermal conductivity of the mixture is obtained by multiplying the thermal conductivity of graphite by the volume fraction of graphite. The specific heat capacity of the mixture is obtained by the following equation.

$$c = \frac{\sum_i m_i \times c_i}{\sum_i m_i} \text{ (J/(kg} \cdot \text{K))} \quad (2.24)$$

Table 2.1 Physical property of each material, a mixture of Model A, and Model B for the calculation of the heat transfer equation.

Model A	Weight m_i (kg)	Mass density ρ_i (kg/cm ³)	Volume (cm ³)	Volume fraction f_i (%)	Thermal conductivity k_i (J/(m · K))	Specific heat capacity c_i (J/(kg · K))
Quartz glass ball	0.040	2.65	15.1	38	2	1210
Graphite	0.002	5.00	0.4	1	400	720
Magnetite	0.010	2.27	4.4	11	5	862
Mixture	0.052		19.9	50	4.00	1124
Model B						
Quartz glass ball	0.040	2.65	15.1	43	2	1210
Graphite*	0.012	5.00	2.4	7	400	720
Mixture	0.052		17.5	50	28.0	1097

*Graphite contains a susceptor (10 g) and residue after cellulose pyrolysis (2 g).

2.3.6.3 Calculation results of the heat transfer equation

The input power of the microwave was adjusted to 500 W to maintain the measurement temperature of a fiber-type infrared thermometer at 800 °C. Thus, the input power into the cylindrical material was set to 500 W in Eq. (2.23). Fig. 2.13 shows that the temperature distribution in the radial direction of the cylinder changes over time. Fig. 2.13 (a) shows that the temperature difference between the center (Radius: 0 m) and surface (Radius: 0.0180 m) formed when magnetite was used as a susceptor, and the input power was 500 W. The center temperature increased with time and reached 990 °C at 100 s. Therefore, the maximum temperature difference between the center and the surface was 190 °C. When graphite was used as a susceptor, and the input power was 500 W, the maximum temperature difference between the center and the surface was 50 °C, as shown in Fig. 2.13 (b). However, the center temperature of Model B was lower than that of Model A. This indicates that magnetite has the advantage of inducing internal heating. The thermal conductivity of magnetite is lower than that of graphite, as described in Table 2.1. The low thermal conductivity improves the effect of internal heating. A higher temperature is suitable for obtaining a high syngas yield. Therefore, magnetite is more effective than graphite in terms of biomass gasification because the internal heating effect of magnetite is higher than that of graphite. Haneishi reports that hot spots form at the contact point between SiC particles (diameter: 2.38, 3.18, and 3.97 mm) [81]. The present study can not consider the hot spots at the contact point between magnetite powder. The heat transfer calculation model for the hot spots is needed to estimate the temperature distribution more precisely.

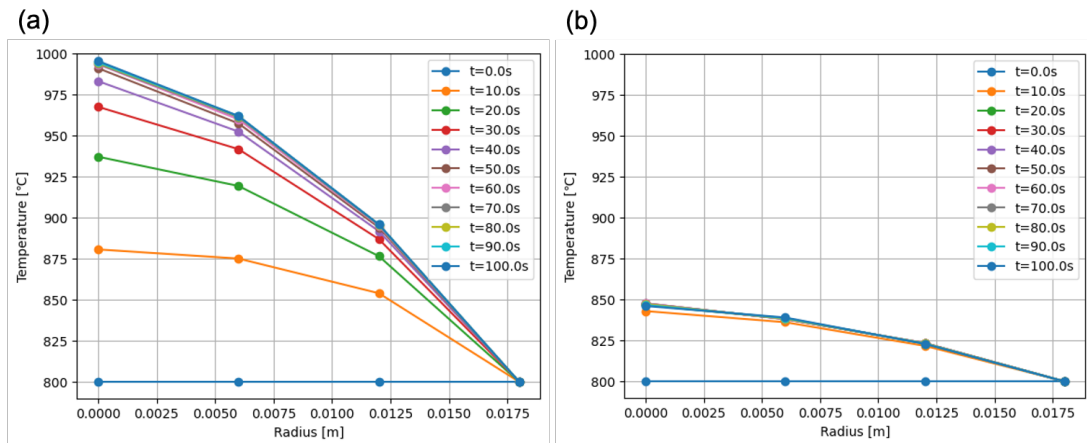


Fig. 2.13 The temperature distribution in the radial direction of the cylinder changes over time ((a) Model A: susceptor: magnetite, $P = 500$ W, (b) Model B: susceptor: graphite, $P = 500$ W)

2.3.6.4 Calculation of microwave penetration depth

The heat transfer equation calculation assumed that the sample section was heated volumetrically. Here, the microwave penetration depth into the sample section is calculated to evaluate the potential for volumetric heating. The penetration depth of a microwave is defined as the distance from the surface at which the microwave power decreases by $1/e$ relative to the microwave power at the surface. The penetration depth of the microwave power in a material can be calculated below.

$$d = \frac{c}{2\pi f \sqrt{2\varepsilon' \{ \sqrt{1 + (\tan\delta)^2} - 1 \}}} \quad (\text{m}) \quad (2.25)$$

Where c , f , and ε' are the speed of light (3.0×10^8 m/s), the microwave frequency (2.45 GHz), and the dielectric constant of the material, respectively. The dielectric constant ε' and $\tan\delta$ is obtained from the following equations.

$$\varepsilon' = \varepsilon_b + \frac{\frac{1}{3} \sum_{i=1}^n f_i (\varepsilon_i - \varepsilon_b) \sum_{k=1}^3 \frac{\varepsilon_b}{\varepsilon_b + N_{ik}(\varepsilon_i - \varepsilon_b)}}{1 - \sum_{k=1}^3 \frac{\varepsilon_b}{\varepsilon_b + N_{ik}(\varepsilon_i - \varepsilon_b)}} \quad (2.26)$$

$$\tan\delta = \frac{\sum_i f_i \tan\delta_i}{\sum_i f_i} \quad (2.27)$$

Eq. (2.26) is called Maxwell-Garnett mixing rule. Where ε_b , ε_i , f_i , and N_{ik} are the dielectric constant of argon (the difference due to gaseous products is negligible because the dielectric property of Ar and gaseous products are similar), the dielectric constant of material of i , and the volume ratio of material of i ($i = 1$: cellulose, $i = 2$: quartz glass balls, $i = 3$: magnetite), and anti-electric field coefficient, respectively. $N_{ik} = 1/3$ when the shape of each material is assumed to be a sphere. The mixture model is shown in Fig. 2.11. Table 2.2 summarizes the dielectric constant and the $\tan\delta$ of the mixture. As a result, ε' and $\tan\delta$ were 1.36 and 0.34, respectively, and the penetration depth d was 0.050 m. The microwave's penetration depth exceeded the cylinder radius (0.018 m). Therefore, microwaves can heat the mixture volumetrically, indicating the formation of internal heating. It is noted that the $\tan\delta$ of magnetite increases with increasing temperature [69]. Hotta et al. report that the $\tan\delta$ of magnetite is 3 at 600 °C. In the present study, the temperature was 800 °C, suggesting that the $\tan\delta$ may increase and decrease the penetration depth. More precise measurement of $\tan\delta$ is necessary in the future. In Model B (graphite), ε' and $\tan\delta$ were 1.36 and 0.035, respectively, and the penetration depth d was 0.090 m. Thus, microwave heats the mixture of Model B volumetrically. The assumption that the volumetric heat in the

sample section is reasonable in heat transfer calculations because microwaves penetrate inside the sample section sufficiently.

Table 2.2 Physical property of each material, a mixture of Model A, and Model B for the calculation of the penetration depth.

Model A	Volume fraction f_i (%)	ε'	$\tan\delta$
Quartz glass ball	38	3.8	0.00006
Graphite	1	12	0.5
Magnetite	11	7.0*	3
Argon	50	1.0	0
Mixture	50	1.34	0.007
Model B			
Quartz glass ball	43	3.8	0.00006
Graphite	7	12	0.5
Argon	50	1.0	0
Mixture	50	1.36	0.035

2.4 Conclusion

This study investigated the effects of magnetite addition to cellulose MWH on syngas production and reached the following conclusions.

- The syngas yield increased with the addition of magnetite when the magnetite–cellulose weight ratio ranged from 0 to 1.0, reaching a maximum yield of 19.1 mmol/g-cellulose at a weight ratio of 1.0. Therefore, the magnetite–cellulose weight ratio is important for improving syngas yield.
- The syngas yield of MWH800 with magnetite is similar to that of CH1000 with magnetite. Moreover, the heat transfer calculation shows that the center temperature of the sample reaches 990 °C when magnetite is used as a susceptor. These results indicate that a temperature region may have occurred inside the sample during MWH800 with magnetite. Thus, magnetite addition may induce internal heating, which improves syngas yield and energy efficiency.
- Cellulose MWH was repeated five times to investigate the effects of magnetite cycle properties on syngas production. The syngas yield decreases with cycling because of the reduction of magnetite to iron. Thus, magnetite cannot be repeatedly used to obtain syngas with a yield similar to that from the 1st cycle.
- Steam oxidation was performed in the 5th cycle to regenerate the magnetite. Iron was oxidized to magnetite by steam, and the regenerated magnetite was mixed with cellulose and microwave heated at 800 °C. The syngas yield of the oxidized magnetite was similar to that in the 1st cycle. Therefore, steam oxidation treatment is effective for regenerating magnetite.

The results of this study suggest that the addition of magnetite caused internal heating. However, the internal temperature was not directly measured in the experiment. Therefore, further research is required to determine the internal temperature using coupled electromagnetic thermal analysis and a fiber temperature sensor. This study demonstrates that the MWH with magnetite can produce syngas from cellulose more efficiently than the CH on the lab scale. The sample feeding system and the microwave heating cavity should be designed for continuous scale-up in the future.

3. Effect of steam feeding on microwave heating of cellulose with magnetite used as a susceptor

3.1 Introduction

Lignocellulosic biomass is a renewable resource used for H₂ production, and this process has been widely investigated [82–84]. Thermochemical conversion is an attractive method to convert lignocellulosic biomass into H₂ for developing low-cost and high-efficiency processes [85]. It involves combustion, pyrolysis, and gasification. The combustion of lignocellulosic biomass mainly produces carbon monoxide and carbon dioxide because lignocellulosic biomass reacts with O₂ [86,87]. Therefore, the combustion process is not suitable for H₂ production. Pyrolysis and gasification are commonly used for H₂ production because little or no O₂ is present [88]. The pyrolysis and gasification of lignocellulosic biomass also produce gases (CH₄, CO, and CO₂), oil, and char [89,90]. However, the pyrolysis and gasification processes require a large amount of thermal energy; thus, considerable energy loss occurs when using conventional methods such as external heating [91].

In recent years, microwave heating (MWH) has been effective for increasing the energy efficiency of pyrolysis because of the internal heating of the sample section [31,92]. However, lignocellulosic biomass is not easily heated using microwaves, owing to its low loss tangent [58,93]. Therefore, it is combined with a high-loss-tangent material. Herein, the high-loss-tangent material is referred to as a susceptor. Metal oxides and carbonaceous materials are generally used as susceptors [94–97]. For example, MgO enhances gaseous production, and both NiO and CuO enhance oil production relative to the case without metal oxide when sugarcane bagasse is pyrolyzed at 500 °C for 30 min [98]. Although various studies have been conducted, the optimal susceptor has not yet been determined. Further research on susceptors is needed to optimize H₂ production [93]. In this study, magnetite was used as a susceptor because of its high-loss-tangent and catalytic activity [45,69].

Steam feeding is an effective method for improving H₂ production because steam reacts with char, oil, and carbon monoxide to generate H₂ [99]. However, the effect of steam on the

MWH of lignocellulosic biomass with magnetite has not been sufficiently investigated. Reactions between magnetite and steam during MWH of lignocellulosic biomass have rarely been investigated. In the present study, the effects of steam feeding on the product yield under MWH are examined. In addition, the effects of MWH are evaluated and compared with those of conventional heating (CH). This study provides valuable insights into the MWH of cellulose with magnetite and serves as a foundation for the optimization of H₂ production from lignocellulosic biomass.

3.2 Methods

3.2.1 Materials

Cellulose powder was obtained from Fujifilm Wako Pure Chemical Corporation (Osaka, Japan). It had a particle size of 38 μm . The contents of C, H, and O in the cellulose powder were 44.4, 6.2, and 49.3 wt% respectively. Magnetite powder obtained from Kojundo Chemical Laboratory Co., Ltd. was used as the susceptor. Its particle size was approximately 1 μm , and its purity was >98 %. The cellulose powder (10 g) and magnetite powder (10 g) were mixed in a glass beaker. The mixture was then placed on quartz glass wool in a quartz glass tube with an inner diameter of 36 mm. Quartz glass balls were mixed with cellulose and magnetite powders to maintain the gas flow.

3.2.2 Experimental procedures

3.2.2.1 Microwave and conventional heating

Fig. 3.1 shows a schematic of the MWH experiment. When the quartz glass tube was installed in the microwave cavity, the sample section was surrounded by a thermal insulator whose internal wall was coated with SiC. High-purity Ar gas was injected into the quartz glass tube for air purging. The flow rate of Ar gas was adjusted to 500 mL/min using a mass flow controller. When the MWH experiments without steam were conducted, the deionized water bottle was empty. During the steam-feeding experiment, deionized water was heated using a mantle heater. V1 and V2 were closed and open, respectively. When the temperature of the deionized water reached 100 °C (monitored by TC1), the electric heater was turned on to heat the steam to 150 °C (monitored by TC2). In the steam-feeding experiment, an Ar (30 vol%) and steam (70 vol%) gas mixture was used. Then, V2 was closed, V1 was opened, and 4 magnetrons (2.45 GHz, UM-1500CS-A, Micro Denshi Co., Ltd.) and stirrer were turned on to begin the MWH experiments. A power meter (PM10-DM, Micro Denshi Co., Ltd.) was used to measure the input and reflection powers of the microwaves generated by each magnetron. The input power of each magnetron was set at approximately 200 W (the total input power was $200\text{ W} \times 4 = 800\text{ W}$) to increase the sample temperature, which was measured using a fiber-type infrared thermometer (FTKX, Japan Sensor

Co., Ltd., Tokyo, Japan). The sample temperature ranged from 180 to 2000 °C. After the sample temperature reached 800 °C, the output power of each magnetron was controlled below 200 W to maintain the sample temperature at 800 °C. Oil products were condensed in the water-cooling section (5 °C, glass container 1) and two cold traps (−45 °C, glass containers 2 and 3). The gaseous products were analyzed via gas chromatography (310C, SRI) using a Hayesep-D column. The operating conditions of gas chromatography were as follows: feeding volume: 1 mL; column temperature: 40 °C; thermal conductivity detector (TCD) measurement temperature: 150 °C; carrier gas: high-purity Ar with a flow rate of 20 mL/min. A standard gas mixture (containing 0.1 vol% H₂, CH₄, CO, and CO₂ in Ar) was used to determine the concentrations of H₂, CH₄, CO, and CO₂ in the non-condensable gaseous product. The MWH experiments were performed for 2 h. The steam feeding was stopped before the magnetron was stopped. CH experiments were also conducted. The thermal insulator was replaced with a ceramic electric ring furnace (ARF-50KC, Asahi Rikagaku). A K-type thermocouple was used to measure the internal temperatures of the samples.

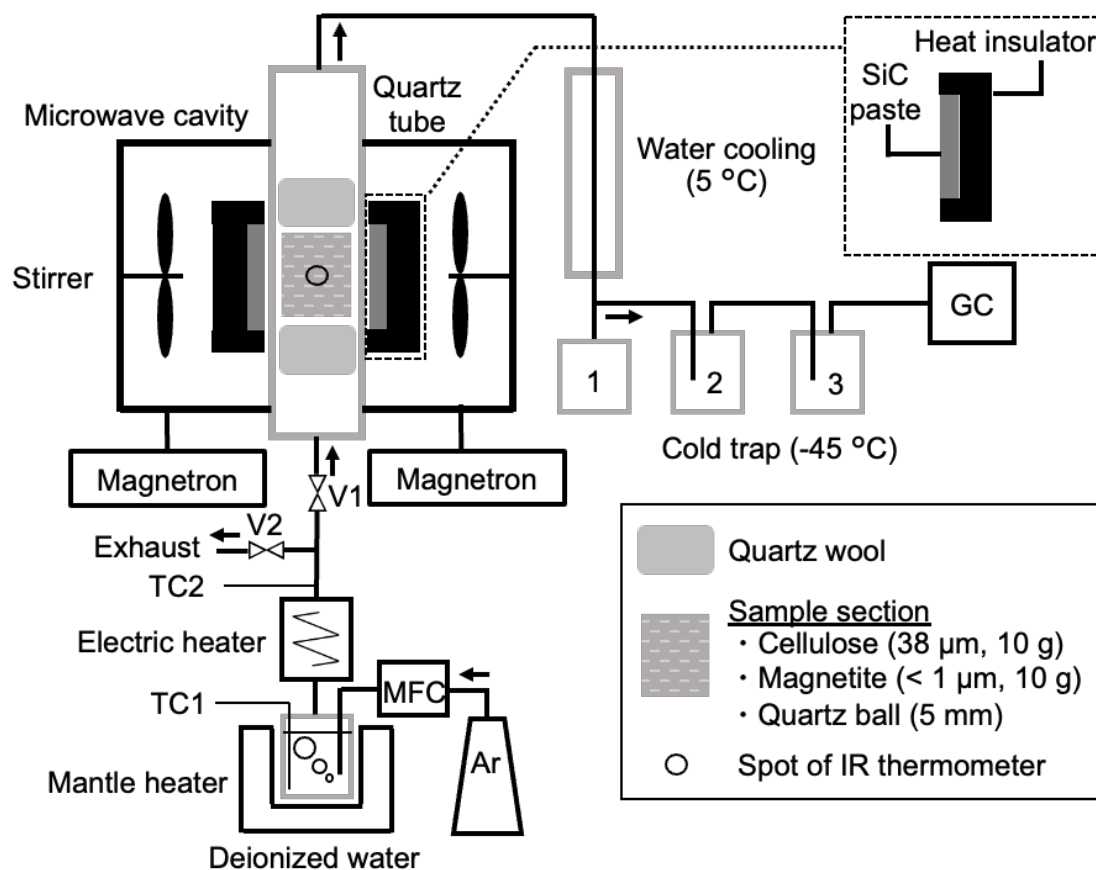


Fig. 3.1 Schematic of the setup for the MWH experiment, wherein MFC, TC, V, and GC denote the mass flow controller, the thermocouples, the valves, and gas chromatography, respectively.

3.2.2.2 Calculation of C content in products

The C content in the gas (CH_4 , CO , and CO_2) X_{gas} was calculated as follows:

$$X_{\text{gas}} = \frac{C_{\text{gas}}}{C_{\text{raw}}} \times 100 \quad (\%), \quad (3.1)$$

C_{gas} (g) and C_{raw} (g) represent the molar amounts of C in the gas and the cellulose, respectively.

The C content of the char X_{char} was calculated as follows:

$$X_{\text{char}} = \frac{C_{\text{char}}}{C_{\text{raw}}} \times 100 \quad (\%), \quad (3.2)$$

where C_{char} (g) represents the difference between the weights of the reaction vessel before and after the experiment. C_{char} was calculated using the following equation when steam was not fed into the reaction vessel:

$$C_{\text{char}} = C_{\text{char}'} - M_{\text{O}}, \quad (3.3)$$

$C_{\text{char}'}$ and M_{O} represent the differences in the reaction vessel weight and molar amount of O atoms in Fe before and after the experiments, respectively. In the experiments without steam feeding, it was assumed that magnetite (Fe_3O_4) was reduced to Fe because the main peak in the X-ray diffraction (XRD) patterns corresponded to Fe. The C content in oil X_{oil} was calculated as:

$$X_{\text{oil}} = 100 - X_{\text{char}} - X_{\text{gas}}, \quad (3.4)$$

3.2.3 Characterization of magnetite powder

XRD analysis of the solid residue was conducted using a SmartLab SE (Rigaku) instrument with $\text{Cu-K}\alpha$ radiation in the 2θ range of $15\text{--}90^\circ$ at intervals of 0.02° . After the experiments, the solid residue was recovered from the quartz glass tube. The microstructure of solid residues was observed by field emission scanning electron microscopy (FESEM, Zeiss Ultra 55). Element mapping was conducted using an energy-dispersive X-ray (EDX) spectrometer of solid residues.

3.3 Results and Discussion

3.3.1 Temperature profiles

The products obtained from cellulose pyrolysis depend on the heating rate. For example, char is the main product when the heating rate is 0.1–1 °C/s (slow pyrolysis), and the amount of oil increases with a heating rate of 10–1000 °C/s (fast pyrolysis) [100]. Fig. 3.2 shows the temperature over time for the MWH and CH. For the CH experiments, the temperature refers to the internal sample temperature measured using the K-type thermocouple. The heating rates were adjusted to be as close as possible to compare the products under each condition. The heating rates for each condition are presented in Table 3.1. The experiments performed in this study involved slow pyrolysis.

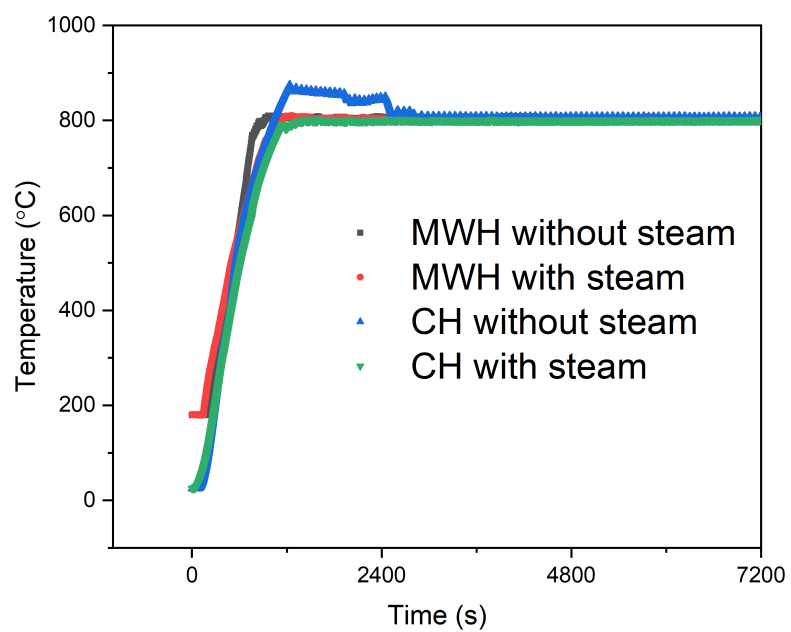


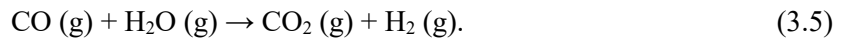
Fig. 3.2 Temperature profiles for MWH and CH.

Table 3.1 Heating rates of MWH and CH.

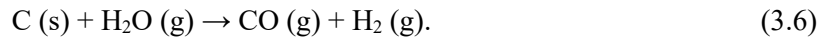
Method	Heating rate (°C/s)
MWH without steam	0.9
MWH with steam	0.6
CH without steam	0.7
CH with steam	0.7

3.3.2 Quantities of gaseous products

Fig. 3.3 shows the gas yields obtained under different conditions. The main gaseous products were H₂, CH₄, CO, and CO₂ under all the conditions. In the MWH experiments, the H₂ and CO₂ yield were increased by 11.1 and 4.0 mmol/g-cellulose, respectively, by steam feeding. Conversely, the CO yield was reduced by 7.5 mmol/g-cellulose. These results indicate the water–gas shift reaction (Eq. (3.5)) occurred.



In the CH experiments, the H₂ and CO₂ yields were increased by 17.9 and 9.5 mmol/g-cellulose, respectively, by steam feeding. The CO yield was reduced by 0.8 mmol/g-cellulose; i.e., it hardly changed. The increases in the H₂ and CO₂ yields of CH were 6.8 and 5.5 mmol/g-cellulose larger than those of MWH. In addition, the reduction in the CO yield of CH was 6.7 mmol/g-cellulose smaller than that of MWH. Therefore, the steam gasification of C occurred during the CH experiments. This phenomenon has been confirmed previously and is expressed as follows [101]:



The CO generated by the reaction of Eq. (3.6) and steam reacted to produce H₂ and CO₂ according to Eq. (3.5), and steam gasification of C enhanced the production of H₂ and CO₂. Without steam feeding, the H₂, CO, and CO₂ yields of MWH were 1.9, 6.1, and 2.0 mmol/g-cellulose higher than those of CH, respectively. Generally, the rate of non-condensable gas production tends to increase with the pyrolysis of cellulose [76]. Li et al. reported that the formation of CO is significantly affected by the secondary reactions of low-molecular-weight products during the conventional heating at 800 °C [75]. Moreover, Mao et al. noted that hot spots in the catalyst may be generated by microwave heating at 800 °C, which enhance the Boudouard reaction (Eq. (3.7)) and increase the CO yield [96].



Without steam feeding, hot spots may be generated in magnetite during MWH and the CO yield increased. In the present study, the CO yield of MWH was significantly higher than that of CH; thus, the pyrolysis of cellulose proceeded to a greater extent for MWH than for CH. From the results of the gas yield in MWH, the H₂ and CO₂ yield increased 11 and 4 mmol/g-cellulose by the steam feeding, respectively. Conversely, the CO yield decreased by 7 mmol/g-cellulose. It is noted that the increased amount of the H₂ yield was higher than the decreased amount of the CO

yield. Moreover, the increased amount of the CO_2 yield was lower than the decreased amount of the CO yield. Therefore, changes in the H_2 , CO, and CO_2 yield were not explained by only the water-gas shift reaction (Eq. (5)). Lin et al. reported that the oxygen vacancy formed during the MWH and CO_2 could be reduced to CO in oxygen vacancies [45]. In addition, steam (H_2O) also reduced in oxygen vacancies and produced H_2 [102]. These insights suggest that oxygen vacancies are formed by the MWH, and the reduction of CO_2 and steam (H_2O) affects the gas yield.

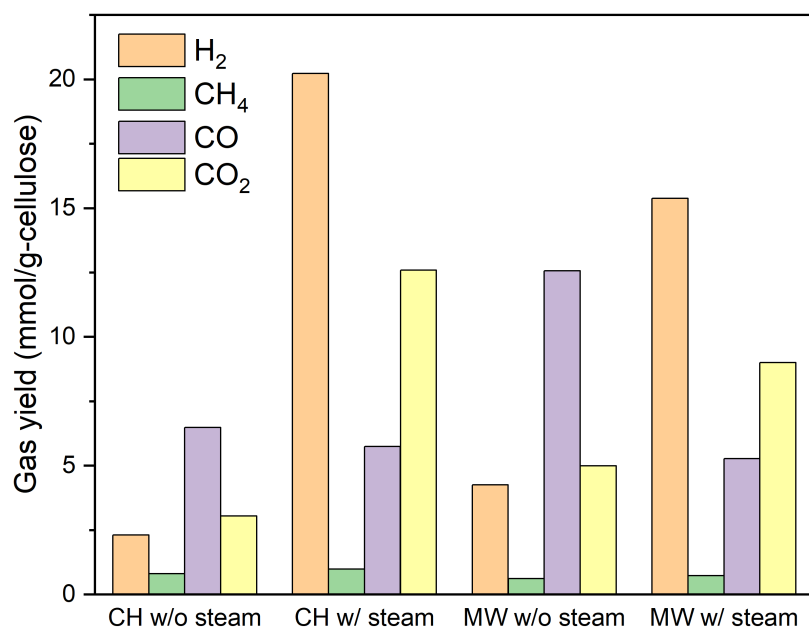


Fig. 3.3 Effect of steam feeding on the gas yield for CH and MWH at 800 °C (w/o: without, w/: with).

3.3.3 Carbon contents of products

Fig. 3.4 shows the C contents of the gas, oil, and char products. When steam was not fed into the reaction vessel, the C content of the MWH char was 20 mol% lower than that for CH. Conversely, the C content of the MWH gas was 25 mol% higher than that for CH. These results suggest that char was selectively heated by microwaves, which enhanced the pyrolysis reaction to a greater extent than CH. In the CH experiments, the C content of the char decreased from 51 to 36 mol%. The C content of the gas increased from 32 to 60 mol%. Generally, C in the char forms graphitic C when cellulose is pyrolyzed above the temperature of 300 °C [73,103]. Therefore, the C in the char and steam reacted to produce H₂ and CO, respectively (Eq. (3.5)), in the CH experiment. This is consistent with the results of increasing the H₂ and CO₂ yields (Section 3.2). In the MWH experiments, the C content of the char decreased from 31 to 28 mol%, and that of the gas decreased from 56 to 46 mol%. The C content of the gas was lower for MWH than for CH, and the reduction in the C content of the char was smaller for MWH than for CH when steam was fed into the reaction vessel. These results indicate that the steam gasification of C was more predominant for CH than for MWH. However, with steam, the C content of the oil was approximately 20 mol% higher for MWH than for CH. This indicates that the MWH produces oil more easily than char when steam is fed into the reaction vessel.

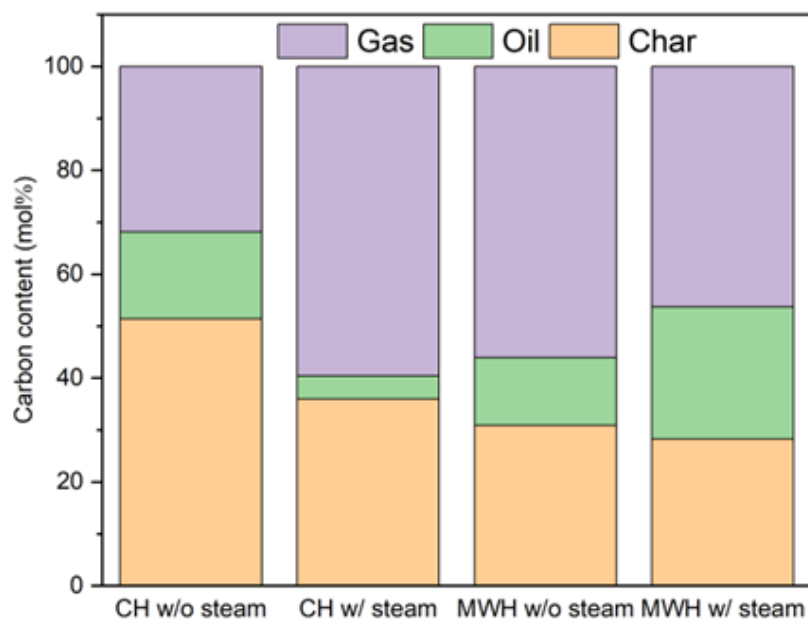


Fig. 3.4 C contents of gas, oil, and char products for CH and MWH (w/o: without, w/: with).

3.3.4 Appearance of reaction vessels

Fig. 3.5 shows the reaction vessels after the CH and MWH experiments. A secondary char was observed downstream of the gas flow section after the CH experiments. Hosoya et al. reported that secondary char was produced at 800 °C under an N₂ atmosphere [104]. Secondary char is formed by the condensation of vapor-phase carbonaceous materials such as tar, which causes problems such as clogging or blockage in gas lines [105,106]. Conversely, the secondary char barely adhered to the downstream gas flow section after the MWH experiments. This suggests that tar decomposed into materials other than secondary char during the MWH. From the results of C content in products (Fig. 3.4), the C content in char of MWH without steam and with steam was 31 and 28 mol%, respectively. The C content in char of CH without steam and with steam was 51 and 36 mol%, respectively. Hence, the C content in the char of MWH is lower than that of CH. This result indicates that MWH reduces secondary char formation because the C content in char includes the secondary char. In studies by Chen et al. and Li et al., tar was decomposed into hydrocarbons such as benzene via MWH [107,108]. Hence, MWH may contribute to the decomposition of tar, reducing the formation of secondary char. The secondary char and oil must be characterized in future studies to clarify the tar decomposition reactions in MWH and CH.

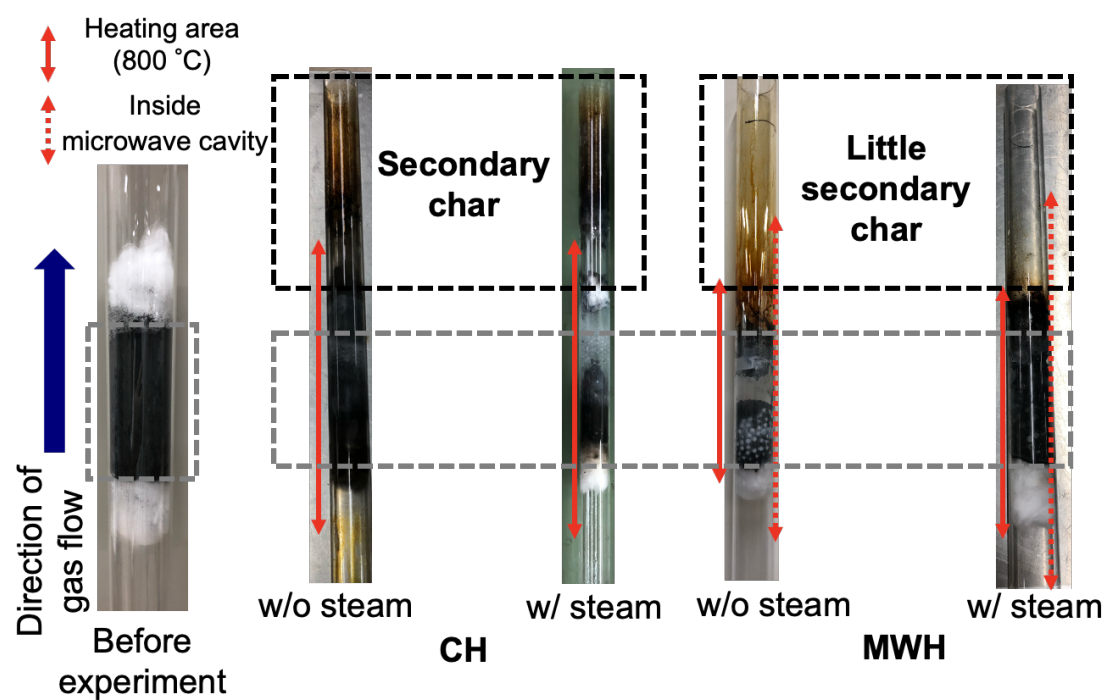
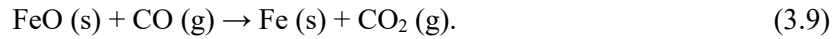


Fig. 3.5 Appearance of reaction vessels after CH and MWH (w/o: without, w/ with).

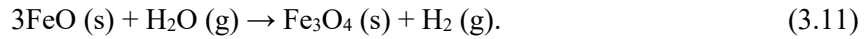
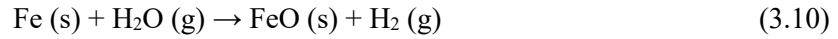
3.3.5 XRD patterns of magnetite powder

Fig. 3.6 shows the XRD patterns obtained before and after heating. The cellulose peak was absent after the MWH and CH experiments. The magnetite (Fe_3O_4) peaks also disappeared, and wüstite (FeO) and Fe peaks appeared when steam was not fed into the reaction vessel. This indicates that magnetite was reduced via the following reactions:



The reactions of Eqs. (3.8) and (3.9) occurred because of the CO generated by the pyrolysis of cellulose. When steam was fed into the reaction vessel in the MWH and CH experiments, the peak patterns of magnetite remained almost unchanged. CO was also produced by the pyrolysis of cellulose in the steam atmosphere; however, because of the water–gas shift reaction (Eq. (3.5)), the reduction reaction of magnetite did not occur.

From another perspective, even after magnetite was reduced, wüstite (FeO) and iron (Fe) were oxidized by reacting with steam (Eqs. (3.10) and (3.11)).



These reactions may enhance the H_2 production with steam feeding. Yoshikawa et al. reported that hot spots formed in the phase boundary between magnetite and wüstite during microwave heating [109]. Without steam feeding, the XRD patterns of the sample after the MWH showed peaks of wüstite. Therefore, the phase boundary between magnetite and wüstite may appear during microwave heating. The hot spots formed by microwave heating may cause an improvement in the CO yield.

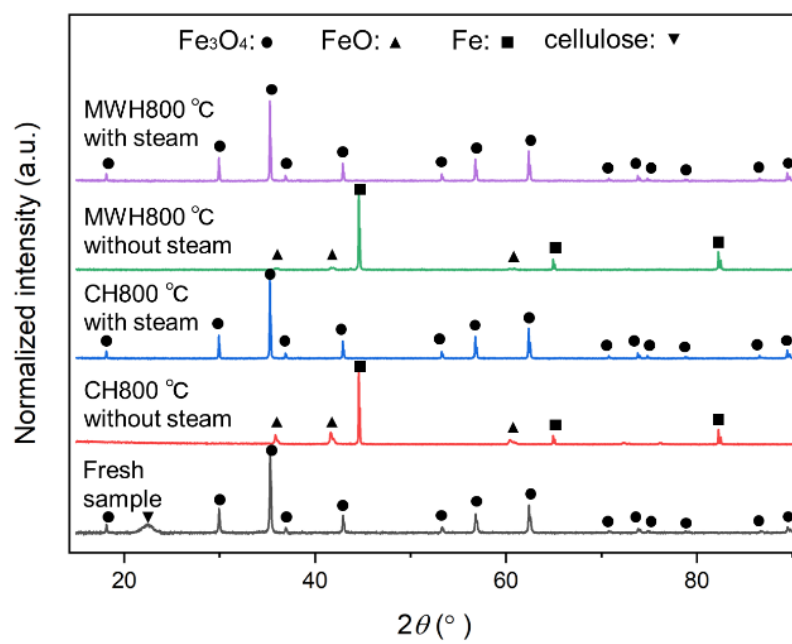


Fig. 3.6 XRD patterns of the samples before and after heating (Cu-K α).

3.3.6 Microstructural observations

Fig. 3.7 shows the SEM images and EDX element mappings of C, O, and Fe in Fe₃O₄. Fig. 3.7 (a) is the fresh sample (Fe₃O₄). The main elements from the mapping analysis were O and Fe. Part of the C element was detected because samples were fixed on carbon tape. Fig. 3.7 (b) and d show the result of the CH without steam and the MWH without steam, respectively. The darker regions of the C mapping were confirmed than in the fresh sample. This indicated that the residue of cellulose pyrolysis appeared. The Fe mapping was distributed uniformly, but undetected regions of the O atom were confirmed. The decrease of O atom relative to the fresh sample (X) was defined as follows:

$$X = \frac{Y}{Y_{\text{fresh}}}, \quad (3.12)$$

Y and Y_{fresh} are the count ratio of O to Fe obtained from EDX quantitative analysis in the used and the fresh sample, respectively. Table 3.2 displays the value of X in each condition. Without steam feeding, O atom in the used sample decreased compared to the fresh sample. These results showed that Fe₃O₄ reduced to Fe, consistent with the results of the XRD analysis. Fig. 3.7 (c) and (e) show the results of the CH with steam and MWH with steam, respectively. The O mapping was distributed more uniformly than for the without steam experiment. Moreover, X of the with steam experiments were higher than those without steam experiments. Therefore, Fe₃O₄ was hardly reduced to Fe. Inside the dashed line rectangle in Fig. 3.7 (d), Fe, whose size is more than 1 μm , was formed, and the O mapping was not detected. It is suggested that Fe particles were melted by the MWH and solidified during cooling. The melting point of pig iron (carbon content 3.8–4.7 %) is 1147 °C. Therefore, hot spots (more than 1147 °C) were formed in the sample section. 2.3.2 showed that the gas yield of MWH800 °C and CH1000 °C were similar. This result suggests that the hot spots achieve more than 1147 °C. However, the results of XRD patterns after MWH800 °C (Fig. 3.6) showed that the peak of Fe₃C was not formed. This suggests that the particle size of Fe₃C is not enough to be detected by the XRD instrument.

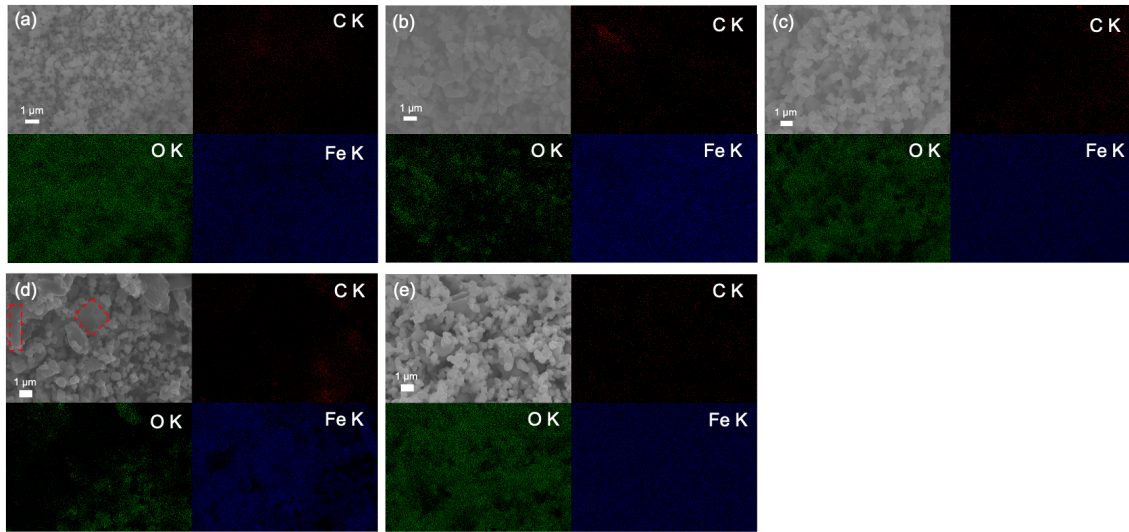


Fig. 3.7 SEM images and EDX element mappings of C, O, and Fe in magnetite. (a) Fresh, (b) CH without steam, (c) CH with steam, (d) MWH without steam, and (e) MWH with steam.

Table 3.2 Decrease ratio of O atom relative to the fresh sample (X) obtained from EDX quantitative analysis under different conditions.

Conditions	X
(b) CH w/o steam	0.14
(c) CH w/ steam	0.81
(d) MWH w/o steam	0.16
(e) MWH w/ steam	0.92

3.3.7 Energy efficiency of gas production

The energy efficiency (η_{gas}) of gas production was evaluated as follows:

$$\eta_{\text{gas}} = \frac{H_{t-\text{gas}}}{W_t} \times 100 \quad (\%), \quad (3.13)$$

where $H_{t-\text{gas}}$ (MJ) and W_t (MJ) are the lower heating values of gaseous products (H_2 , CH_4 , and CO) and the input energy for the experiments, which are integrated up to an arbitrary time t . $H_{t-\text{gas}}$ is obtained as follows:

$$H_{t-\text{gas}} = 120 \times M_{\text{H}_2-t} + 50 \times M_{\text{CH}_4-t} + 10 \times M_{\text{CO}-t} \quad (\text{MJ}), \quad (3.14)$$

where M_{H_2-t} , M_{CH_4-t} , and $M_{\text{CO}-t}$ (g) represent the H_2 , CH_4 , and CO yields at arbitrary time t . The W_t values of the CH ($W_{t-\text{CH}}$) and MWH ($W_{t-\text{MWH}}$) experiments were calculated using the following equations:

$$W_{t-\text{CH}} = W_{t-\text{furnace}} + W_{t-\text{steam}} \quad (\text{MJ}) \quad (3.15)$$

$$W_{t-\text{MWH}} = W_{t-\text{magnetron}} + W_{t-\text{steam}} \quad (\text{MJ}), \quad (3.16)$$

where $W_{t-\text{furnace}}$, $W_{t-\text{steam}}$, and $W_{t-\text{magnetron}}$ represent the Joule heat for the electric furnace, the heating energy for steam (including mantle heater and electric heater) and Ar, and the difference in microwave input energy and reflection energy, respectively. When steam was not fed into the reaction vessel, $W_{t-\text{steam}}$ was 0 J. Fig. 3.8 shows the time series of the energy efficiency (η_{gas}) for MWH and CH. In the experiments without steam feeding (Fig. 3.8 (a)), the η_{gas} of CH increased until 4000 s and reached a maximum value of 3 %. Subsequently, it remained relatively constant. The η_{gas} of MWH increased until 840 s and reached a maximum value of 5 %. Thereafter, it decreased. Until 2500 s, the η_{gas} of MWH was higher than that of CH. Conversely, the η_{gas} of MWH was lower than that of CH after 2500 s. These results indicated that the gasification reaction proceeded more rapidly for MWH than for CH. In the steam-feeding experiments (Fig. 3.8 (b)), the η_{gas} of MWH and CH increased until 2300 and 2500 s, respectively, and reached maximum values of 4.3 % and 3.8 %, respectively. After reaching the maximum values, the η_{gas} of MWH and CH decreased. For the CH method, the η_{gas} were increased by the steam feeding. In addition, the η_{gas} of CH was higher than that of MWH after 1500 s (shown in Fig. 3.8 (b)). Therefore, CH was more efficient than MWH for gas production because the steam gasification of C was more effective.

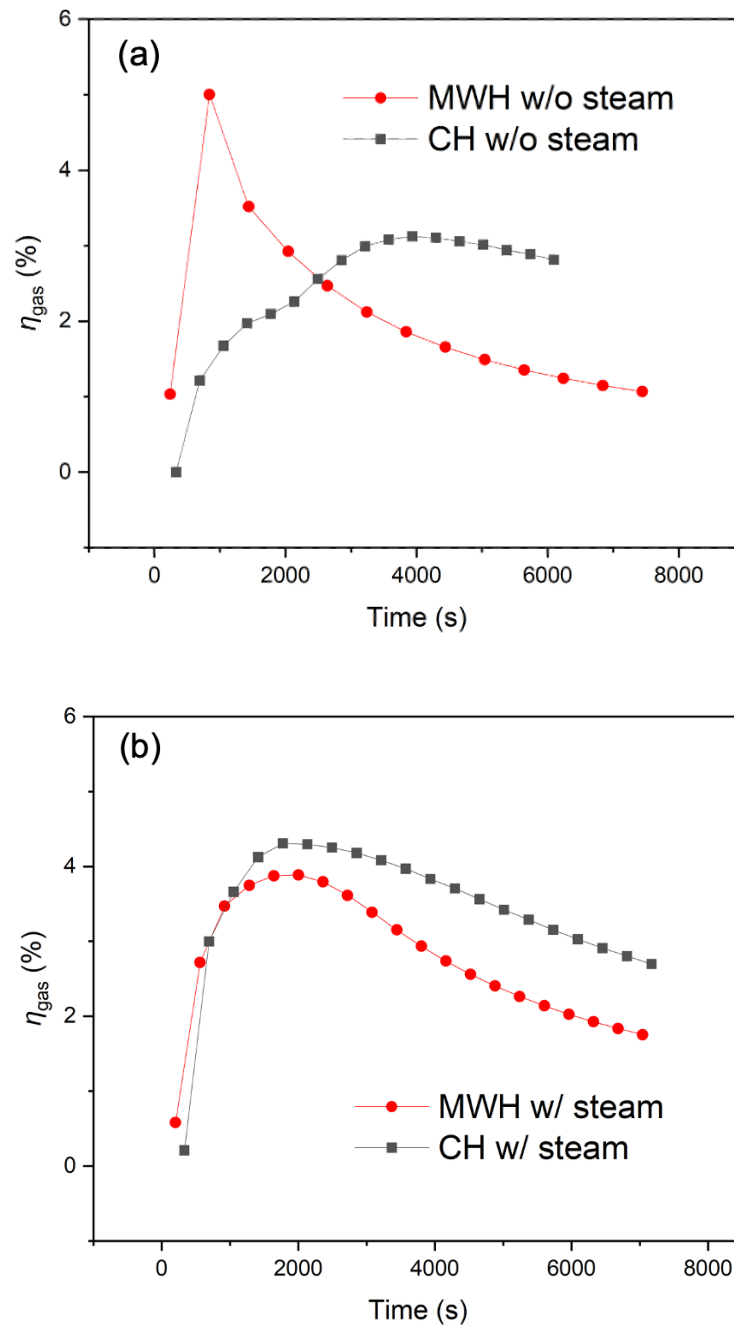


Fig. 3.8 Energy efficiency of H_2 , CH_4 , and CO production for MWH and CH: (a) without steam; (b) with steam.

3.3.8 Estimation of heat dissipation

In the present study, heat dissipation from the heat insulator surrounding the quartz glass tube is confirmed due to the limitation of the experiment setup. Fig. 3.9 shows (a) the top view of the heat insulator and (b) the surfaces of the top and the bottom. Table 3.3 summarizes the details of the heat insulator before and after improvement. The size of the heat insulator is changed to reduce heat dissipation.

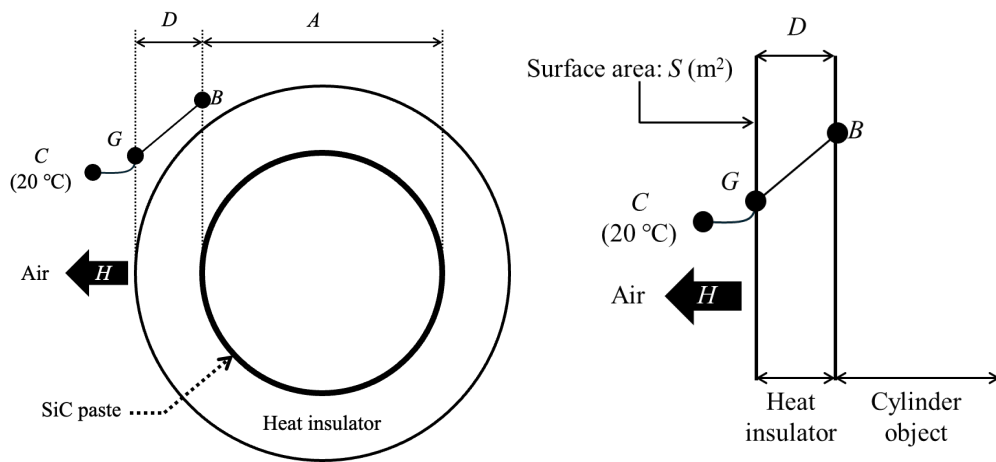


Fig. 3.9 (a) The top view of the heat insulator and (b) the surface of the top and the bottom.

Table 3.3 Details of the heat insulator.

Item	Before	After
The outer diameter of the cylindrical object: A (m)	0.110	0.040
The surface area of the top and the bottom: S (m ²)	0.018	0.002
The temperature of the cylindrical object: B (°C)		800
The outside air temperature: C (°C)		20
The thickness of the heat insulator: D (m)	0.020	0.200
The thermal conductivity of the heat insulator: E (W/m/K)		0.3 ^{*1}
The surface emissivity of the heat insulator: F		0.8
The convective heat transfer coefficient to the outside air: J (W/m ² /K)		10

*1 The thermal conductivity of the heat insulator is referred to the the isolite (JIS A series Insulating Firebrick) [110].

3.3.8.1 Heat dissipation from the side of the insulated cylinder per 1 m

When a cylindrical object of outer diameter A (m) and temperature B (°C) is insulated by a heat insulator of thickness D (m) and thermal conductivity E (W/m/K), the heat dissipation to the air at temperature C (°C) is H (W/m) per meter of cylinder length. The heat dissipation is calculated based on a cylinder length of 0.1 m because the length of the SiC paste is 0.1 m. Here, the heat dissipation from the top, bottom, and sides of the heat insulator was calculated for simplicity.

$$\alpha = (A + 2 \times D) \times \pi \quad (3.17)$$

$$K = (G - C) \times J \times \alpha \quad (3.18)$$

$$L = ((G + 273.15)^4 - (C + 273.15)^4) \times F \times \sigma \times \alpha \quad (3.19)$$

$$H = K + L = \frac{2 \times \pi \times E \times (B - G)}{\ln\left(\frac{A + 2 \times D}{A}\right)} \quad (3.20)$$

Where α , J , F , K , and L are surface area per meter, convective heat transfer coefficient to the outside air (W/m²/K), surface emissivity, convective heat radiation per meter (W/m), and radiative heat radiation per meter (W/m), respectively. The surface temperature G (°C) is determined at which $K + L$ equals the amount of heat passing through the insulation.

3.3.8.2 Heat dissipation from the top and bottom of insulated flat plate

When a flat plate with surface area S (m²) and temperature B (°C) is kept warm by a heat insulator with thickness D (m) and thermal conductivity E (W/m/K), the heat dissipation to the air at temperature C (°C) is H (W).

$$\alpha = (G - C) \times J \quad (3.21)$$

$$\beta = ((G + 273.15)^4 - (C + 273.15)^4) \times F \times \sigma \quad (3.22)$$

$$\frac{E}{D}(B - G) = \alpha + \beta \quad (3.23)$$

$$K = \alpha \times S \quad (3.24)$$

$$L = \beta \times S \quad (3.25)$$

$$H = K + L \quad (3.26)$$

Where α , β , J , and F are convective heat radiation per unit area (W/m^2), radiative heat radiation per unit area (W/m^2), convective heat transfer coefficient to the outside air ($\text{W}/\text{m}^2/\text{K}$), and surface emissivity, respectively. The surface temperature G ($^{\circ}\text{C}$) is determined at which $\alpha + \beta$ equals the amount of heat passing through the insulation.

3.3.8.3 Calculation result

Table 3.4 shows the heat dissipation from the insulator's side, top, and bottom before and after improvement. By changing the outer diameter of the cylindrical object, the surface area of the top and the bottom, and the thickness of the heat insulator (shown in Table 3.3), the total heat dissipation was reduced by approximately 90 %.

Table 3.4 Heat dissipation from the insulator's side, top, and bottom.

Item	Before	After
The side of the heat insulator (W)	314	45
The top and bottom of the heat insulator (W)	142	2
Total heat dissipation (W)	456	47

3.3.9 Real biomass experiment

The feasibility of applying the microwave heating system to real biomass was investigated. The product yield and the appearance of the reaction vessel after the experiments. The present study used cellulose to simulate biomass. On the other hand, experimental data on real biomass is needed for social implementation. Therefore, the product yield of real biomass and the appearance of the reaction vessel after the experiment are investigated. Japanese cypress sawdust whose diameter < 5 mm was used as real biomass and dried at 100 °C for 24 hours. 5 g Japanese cypress sawdust was mixed with 5 g magnetite, as shown in Fig. 3.1. The experimental procedure is the same as 3.2.2. The sample was heated at 800 °C in a steam atmosphere for 2 hours. Fig. 3.10 shows the gas yield of Japanese cypress sawdust changes over heating time. Gas yields were approximately constant at 3600 s. As a result, the H₂, CH₄, CO, and CO₂ yield was 17.7, 1.3, 7.7, and 10.4 mmol/g-Japanese cypress sawdust. The char yield was calculated from the difference in the weight of the quartz glass tube (including the sample) before and after the experiment. The oil yield calculated the difference between the total weight of the biomass and the weight of gas and char. Table 3.5 summarizes the yield of each product from cellulose and Japanese cypress sawdust. The products from Japanese cypress sawdust and cellulose were similar. Fig. 3.11 shows the appearance of the reaction vessel after the experiment. The picture on the right-most side shows the reaction vessel after microwave heating Japanese cypress sawdust. The secondary char barely adhered to the downstream gas flow section. This suggests that the combination of MWH and steam feeding reduces the formation of secondary char. Therefore, the microwave biomass conversion system is expected to be effective for the continuous process.

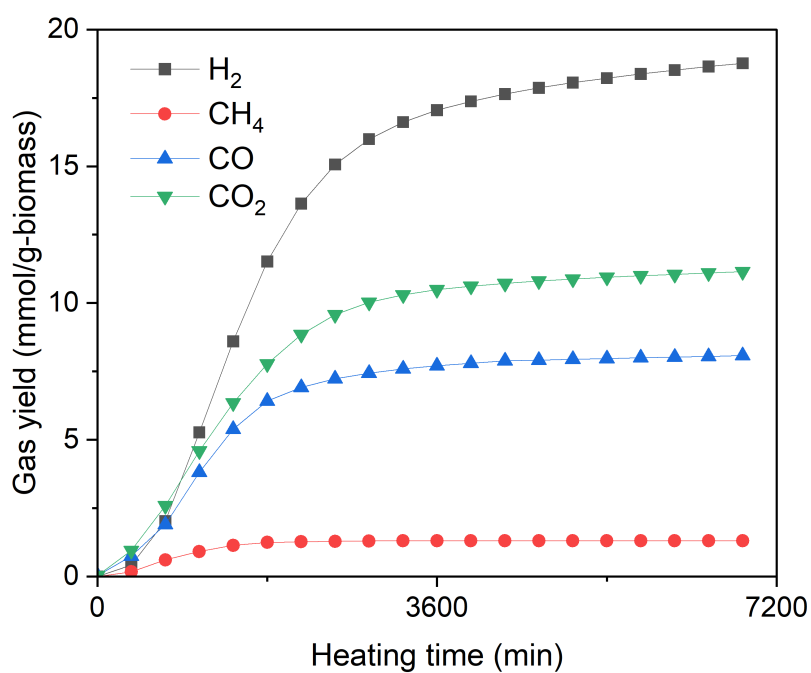


Fig. 3.10 The gas yield changes over heating time.

Table 3.5 Yield of each product from cellulose and Japanese cypress sawdust

Product	Yield of product (kg/kg-cellulose)	Yield of product (kg/kg-Japanese cypress sawdust)
H ₂	0.031	0.034
CH ₄	0.011	0.021
CO	0.148	0.216
Char	0.11	0.09
Oil	0.30	0.17

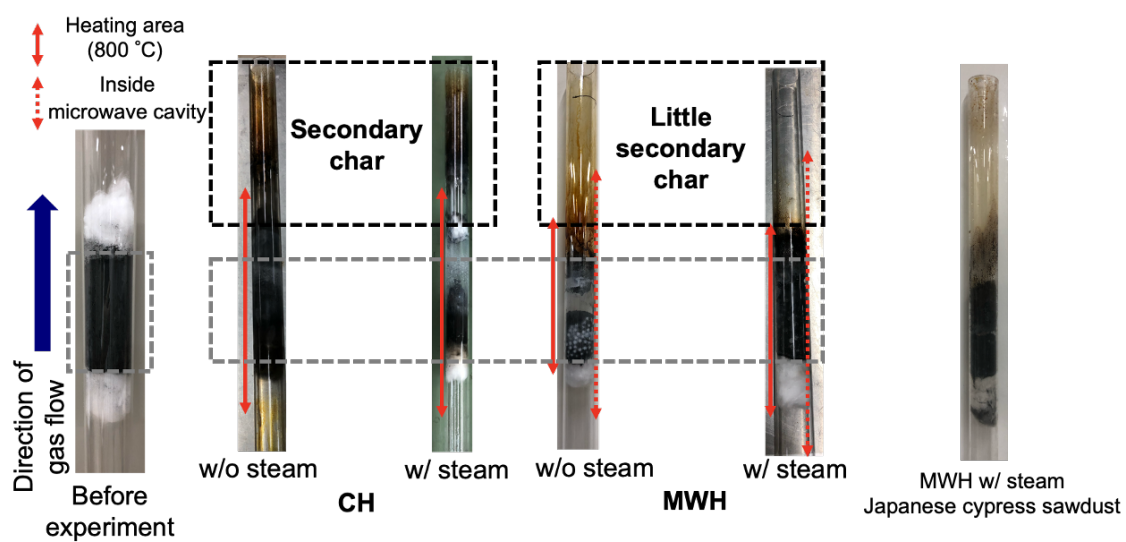


Fig. 3.11 The appearance of reaction vessels before and after the experiment.

3.4 Conclusions

The effects of steam feeding and MWH on the gas production, crystal structure of magnetite, and energy efficiency, when microwaves heated a cellulose and magnetite mixture under a steam atmosphere, were investigated. For MWH and CH, the H_2 and CO_2 yields were increased by steam feeding because the water–gas shift reaction occurred. According to the C content in the char, the steam gasification of C was more predominant for CH than for MWH, leading to higher H_2 and CO yields. According to the XRD patterns of the samples before and after heating, magnetite was reduced by CO, and wüstite and Fe were produced when steam was not fed into the reaction vessel. The XRD patterns of magnetite remained almost unchanged with steam feeding, indicating that the water–gas shift reaction was preferable for the reduction of magnetite. For the CH method, the energy efficiency was increased by steam feeding. Therefore, steam feeding via CH is effective for gas production. The H_2 , CO, and CO_2 yields of MWH exceeded those of CH, and the C content in the char was lower for MWH than for CH, indicating that the pyrolysis of cellulose proceeded to a greater extent in the case of MWH. According to the appearance of the reaction vessels, the formation of secondary char was reduced by MWH. Therefore, MWH can overcome problems such as the clogging or blockage of gas lines. However, further research is needed to determine the secondary char and oil products to elucidate the formation mechanism of secondary char and oil products under MWH. The results of XRD and SEM indicate that magnetite can be used repeatedly when MWH with steam feeding is applied. Moreover, the combination of MWH and steam feeding is expected to solve the tar problems. Therefore, the microwave biomass conversion system with steam feeding is expected to use for continuous biomass processing. The following chapter evaluates the economic feasibility and social acceptability of the microwave biomass conversion system. Conventional heating is not studied because continuous processing is difficult due to tar formation.

4. Techno-economic assessment of microwave biomass conversion system: The case of Kyushu, Japan

4.1 Introduction

The accumulation of forest residue has become a problem in Japan. If forest residues are not correctly used, the forest environment will degrade [111,112]. Therefore, they need to be utilized. In response, the Ministry of Agriculture, Forestry and Fisheries promotes energy use from forest residues [113]. This has increased the utilization ratio of forest residues (the utilization amount of forest residues relative to the amount generated) from 9 % (2014) to 38 % (2022). However, forest residues are still accumulating, and utilization rates must be increased.

There is another social issue in Japan. The Committee on Adjustment Capacity and Supply-Demand Balance Evaluation announced that frequency quality decreased in the west of Japan in September 2024 [114]. During the light load period (spring and fall), frequency quality is significantly reduced because controlling the demand and the supply becomes difficult. Solar excess power is considered as one of the factors contributing to the frequency fluctuation problem. In addition, solar output in Kyushu is the most significant excess in Japan at 1 billion kWh per year. The decline of frequency quality is a concern in the west of Japan. The adjustment amount must be secured by increasing the installation of solar power generation facilities in the future. Pumped storage is useful to adjust the power. Still, it is not easy to introduce pumped storage power plants due to cost and land capacity. Therefore, another adjustment method is needed. The microwave biomass conversion system can solve the above two problems because electricity-generated microwaves can convert biomass to useful products such as H_2 , CH_4 , CO , char, and oil [115,116].

Recently, a techno-economic assessment of microwave biomass conversion has been researched because it has attracted attention as a highly energy-efficient biomass conversion method. Neha et al. conducted a techno-economic analysis of the microwave pyrolysis of biomass (food waste and polyethylene) to verify the economic feasibility of the system when biochar and bio-oil are used as products [117]. They indicate that the system is economical because the internal rate of return (IRR) is higher than the discount rate (7 %), and the payback time is 4 years less than the system's lifetime. Lam et al. studied a techno-economic analysis of microwave vacuum

pyrolysis of biomass (waste plastic and used cooking oil) for oil production based on the experimental data of the lab scale [118]. They indicate that the cost of oil generated by the lab-scale system (2.02 USD/L) is higher than the diesel fuel price in Malaysia (0.52 USD/L, August 2019). The low treatment amount of the lab-scale system affects the low annual production and high cost of oil. Therefore, they investigate a techno-economic analysis of the scale-up system. As a result, the cost of oil generated by the scale-up system (0.25 USD/L) is lower than the diesel fuel price in Malaysia. Abomohra et al. verified the economic feasibility of the microwave vacuum co-pyrolysis of biomass (waste plastic and seaweeds) for enhanced bio-oil [119]. They show that the net present value of the system is 23.17 million USD for the 20-year lifetime plant. These studies indicate that the microwave biomass conversion system for oil production is economically feasible. However, the economic characteristics of microwave biomass conversion considering gaseous products have not been sufficiently investigated. Moreover, the power conditioning ability and the utilization rate of forest residues in the microwave biomass conversion system have not been evaluated. Therefore, the present study performed a techno-economic assessment of the microwave biomass conversion system based on the experimental data of the previous study to investigate the feasibility of the investment [120]. In addition, the power conditioning ability and the utilization rate of forest residues are evaluated when the microwave biomass conversion system is applied in Kyushu, Japan.

4.2 Methods

4.2.1 Evaluation model

Fig. 4.1 shows the schematic of the microwave biomass conversion system. Biomass is converted to useful products such as H_2 , CH_4 , CO , char, and oil in a microwave biomass gasifier. Biomass has a low microwave absorption [121]. Thus, a high microwave absorption material called a susceptor is added [122–124]. Magnetite is one of the useful susceptors because of its high microwave absorption and catalytic ability [46,120,125]. Magnetite is initially loaded in the gasifier. Based on the experimental data of the previous study, the syngas yield obtains the maximum value when the weight ratio of biomass and magnetite is 1:1 [126]. Moreover, magnetite can be reused by steam feeding into the gasifier.

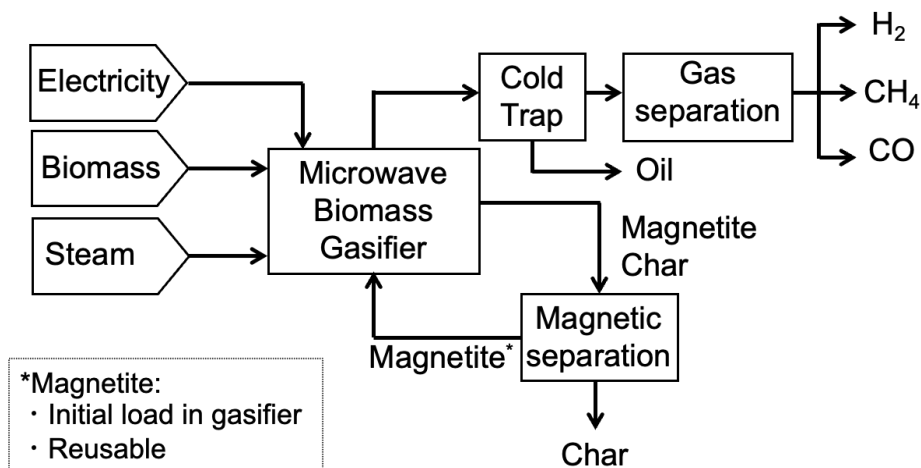


Fig. 4.1 Schematic of microwave biomass conversion system

4.2.2 Techno-economic assessment procedure

4.2.2.1 Input energy per hour (E_{in})

A magnetron generates the input energy of the microwave biomass conversion system. The magnetron (output power: 100 kW, oscillation efficiency: 80 %) is used, resulting in a system input energy (E_{in} : MJ/h) of 360 MJ/hour.

4.2.2.2 Amount of biomass (M_{bio})

According to a previous experimental study, the microwave input energy required 110 MJ/kg-biomass to convert biomass into products [120]. However, approximately 90 % of the input energy was heat dissipation from the thermal insulator. By changing the thickness of the insulation and diameter of the quartz glass tube feed-thorough port, the heat dissipation is expected to be reduced by about 90 % (described in 3.3.8). In the present study, the energy requirement for biomass conversion (E_{conv} : MJ/kg-biomass) was set to 20 MJ/kg-biomass. The amount of biomass is calculated as follows:

$$M_{bio} = \frac{E_{in}}{E_{conv}} \text{ (kg-biomass/h)} \quad (4.1)$$

4.2.2.3 Product amount (M_x)

The present study treats hydrogen, methane, carbon monoxide, char, and oil as products. Based on a previous experimental study, Table 4.1 summarizes the yield of products (Y_x : kg/kg-biomass, $x = H_2, CH_4, CO, \text{ oil, and char}$) based on the experimental data of the previous study [120]. Each product amount per hour (M_x : kg/h, $x = H_2, CH_4, CO, \text{ Oil, and Char}$) is calculated as follows:

$$M_x = M_{bio} \times Y_x \text{ (kg/h)} \quad (4.2)$$

4.2.2.4 Annual income (I_{annual})

The income per hour (I_x : USD/h) is calculated as follows:

$$I_x = M_x \times k_x \text{ (USD/h)} \quad (4.3)$$

where, k_x (USD/kg, $x = \text{H}_2, \text{CH}_4, \text{CO}, \text{Oil}, \text{and Char}$) is the sales price of each product. Annual income is obtained from products. The value of each product is summarized in Table 1. Annual income (I_{annual} : USD/year) is obtained from the following equations:

$$I_{\text{annual}} = I_{\text{total}} \times T_{\text{op}} \text{ (kg/h)} \quad (4.4)$$

$$I_{\text{total}} = \sum I_x \quad (4.5)$$

where, I_{total} (USD/h) and T_{op} (h) are the total income from products per hour and the operation time, respectively. The operation time is assumed to be the duration of sunshine in Kyushu (2,100 h) [127].

Table 4.1 Parameters for products and income.

Product	Yield of product : Y_x (kg/kg-biomass)	Product amount per hour : M_x (kg/h)	Sales price: k_x (USD/kg)	Income: I_x (USD/h)	Annual income: I_{annual} (USD/year)
H ₂	0.031	0.42	14.7 ^{*1}	2.66	7,314
CH ₄	0.011	0.16	2.00 ^{*2}	0.32	899
CO	0.148	1.98	0.21 ^{*3}	0.41	1,123
Char	0.11	1.41	1.03 ^{*4}	1.46	4,012
Oil	0.30	4.24	0.92 ^{*5}	3.90	10,715
Total					24,063

*1, 2, 3, 4: The sales price of H₂, CH₄, CO, char, and oil are referred to [18], [128], [129], [129], and [130] respectively.

4.2.2.5 Initial cost (C_{initial})

Initial costs (C_{initial} : USD) include a microwave heating unit (output power: 100 kW), a reaction vessel, a gas flow system, a power unit, a cold trap, a steam feeding system, a magnetite, and a magnetite/char separation (Table 4.2). In the present study, a quantity discount of 20 % is applied to the initial cost.

4.2.2.6 Operation cost (C_{op})

Operation costs (C_{op} : USD/year) include maintenance, insurance, tax, consumable items, other expenses, labor costs for the operator and field manager, biomass procurement, and magnetron replacement (Table 4.2). The operation costs of maintenance, insurance, tax, consumable items, and other expenses are calculated as a ratio to the initial cost [131]. The magnetron cost is 400 USD/kW, and the lifetime is 3 years. Moreover, the quantity discount of 20 % is applied. Thus, the cost of the magnetron replacement is 10,667 USD/unit when the magnetron with 100 kW output power is used.

4.2.2.7 Annual Revenue (R_{annual})

Annual revenue (R_{annual}) is defined as the difference between the annual income and the operation cost, calculated as follows:

$$R_{\text{annual}} = I_{\text{annual}} - C_{\text{op}} \text{ (USD/year)} \quad (4.6)$$

4.2.2.8 Number of units (N)

The above income and cost are calculated per one microwave biomass conversion system unit. The upper limit for the number of systems is the amount of biomass four operators can process yearly (10,000 t/year/two operators).

Table 4.2 Parameters for initial and operation costs.

Initial cost	USD/unit	Note
Microwave heating unit ^{*1}	100,000	100 kW, oscillation efficiency 80 % (including power unit)
Reaction vessel	6,667	The size of the reaction vessel is 1m ³ *2
Gas flow system	3,333	
Power unit	3,333	Except for the microwave heating unit
Cold trap	3,333	
Steam feeding system	3,333	
Magnetite	1,129	66.7 USD/kg ^{*3}
Magnetite/char separation	3,333	
Gaseous products separation	6,667	
Total initial cost	131,128	
Quantity discounted initial cost	104,902	Quantity discount rate 20 %
Operation cost	USD/unit/ year	
Maintenance	2,512	4.0 % relative to initial cost
Insurance and tax	942	1.5 % relative to initial cost
Consumable	2,826	4.5 % relative to initial cost
Other expenses	1,256	2.0 % relative to initial cost
Labor for operator (two)	106,664	26,666 USD per operator
Field manager	53,333	
Biomass procurement	1,260	33.3 USD/t-biomass ^{*4}
Magnetron replacement ^{*5}	10,667	A lifetime of the magnetron 3 year
Total operation cost	179,460	

- *1: The microwave heating unit cost was calculated using 1,000 USD/kW [132].
- *2: The reaction vessel of the biomass gasification was 6,667 USD/m³ [133].
- *3: The magnetite sales price was estimated at 66.7 USD/kg [134].
- *4: The biomass procurement cost was 33.3 USD/t-biomass [135,136].
- *5: The magnetron cost was calculated using 400 USD/kW [137].

4.2.3 Calculation of Cash flow, IRR, and NPV

Cashflow is calculated as follows:

$$CF_t = R_{\text{annual-}t} \times (1 - C_{\text{tax}}), (1 \leq t \leq 30 \text{ year}, CF_0 = -C_{\text{initial}}) \quad (4.7)$$

C_{tax} is income tax (30 %). The lifetime of the microwave biomass conversion system is assumed to be 30 years. The discount rate is assumed to be 3 %. The internal rate of return (IRR) is obtained from the following equations.

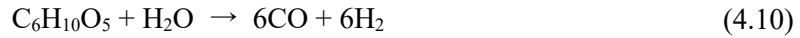
$$\sum_{t=0}^n \frac{CF_t}{(1 + IRR)^t} = 0 \quad (4.8)$$

The net present value (NPV) is the cumulative value (revenues – expenses).

$$NPV = \sum_{t=1}^n \frac{CF_t}{(1+IRR)^t} - C_{\text{initial}} \quad (4.9)$$

4.2.4 Sensitivity analysis

The sensitivity analysis for the IRR and NPV of the microwave biomass conversion system evaluates the influence of parameters such as the energy requirement for biomass conversion (E_{conv} : MJ/kg-biomass) and the cost of the magnetron (C_{MW} : USD/unit). The E_{conv} ranged from 15 to 20 MJ/kg-biomass. Here, the chemical formula of biomass is assumed to be $C_6H_{10}O_5$. The steam gasification of biomass reaction for syngas production is described as follows:



The required energy for the steam gasification of biomass reaction is 5 MJ/kg-biomass. The heat dissipation (described in 2.2.2) occurred at 10 MJ/kg-biomass. Therefore, the minimum value of the energy requirement for biomass conversion was set to 15 MJ/kg-biomass. The C_{MW} ranged from 66,667 to 100,000 USD/unit. Moreover, the Japanese government aims to reduce the k_{H_2} to 2.23 USD/kg- H_2 by 2030 [138]. The present study performed the sensitivity analysis for the E_{conv} and the C_{MW} , considering the hydrogen sales price 2030.

4.2.5 Power conditioning ability (A_{power}) and Utilization ratio of forest residue (U_{residue})

The power conditioning ability of the microwave biomass conversion system is defined as follows:

$$A_{\text{power}} = \frac{E_{\text{mw}}}{E_{\text{pump}}} \times 100 (\%) \quad (4.11)$$

E_{mw} (MWh) and E_{pump} (MWh) are the input energy for the microwave oscillation and the energy capacity of the pumped storage. E_{pump} in Kyushu used 15,840 MWh [139]. The present study simulates the microwave biomass conversion system installed at 15 sites in Kyushu [140]. Moreover, the microwave biomass conversion system functions as the power conditioning from 7:00 a.m. to 5:00 p.m. (10 h). Therefore, E_{MW} is obtained from the following equation, considering the oscillation efficiency of the magnetron (80 %).

$$E_{\text{MW}} = 0.1 (\text{MW}) \times \frac{100}{80} \times N \times 15 (\text{site}) \times 10 (\text{h}) \quad (4.12)$$

The utilization ratio of forest residues of the microwave biomass conversion system is defined as follows:

$$U_{\text{residue}} = \frac{M_{\text{treat}}}{M_{\text{forest}}} \times 100 (\%) \quad (4.13)$$

M_{treat} (t/year) and M_{forest} (t/year) are the annual treatment amount of the microwave biomass conversion system and the annual generated amount of forest residues, respectively. M_{forest} in Kyushu is estimated as follows:

$$M_{\text{forest}} = M_{\text{Japan}} \times \frac{V_{\text{Kyushu}}}{V_{\text{Japan}}} \quad (14)$$

Where M_{Japan} (970 million t/year) [113], V_{Kyushu} (m^3), and V_{Japan} (m^3) are the annual generated volume of forest residues in Japan and the annual volume of forest residues in Kyushu and Japan, respectively [141]. As a result, M_{forest} was 1,418,111 t/year.

4.3. Results and Discussions

4.3.1 Cash flow, IRR, and NPV

To evaluate the economic feasibility of the microwave biomass conversion system, the IRR, the NPV, and the payback period were calculated [142–146]. Fig. 4.2 shows that the cash flow and cumulative cash flow change over the operation period. The initial cost was the first year's cash flow ($t = 0$). The cash flow decreased according to the discount rate (3 %) after the second year ($t \geq 1$). The initial cost was not recovered within the system's lifetime. The IRR and the NPV were calculated using Eq. (4.8) and (4.9), respectively. As a result, the IRR and the NPV were 1.43 % and -6.81 million USD, respectively. Here, the microwave biomass conversion system is assumed economically feasible when (i) the IRR is higher than the discount rate (3 %), (ii) the NPV is a positive value, and (iii) the initial cost can be recovered within the system's lifetime. The IRR was less than the discount rate (3 %), and the NPV was negative. Moreover, the initial cost was not recovered within the system's lifetime. Therefore, this system was not an economically feasible investment. The following section describes which system parameters can be improved to become economically viable.

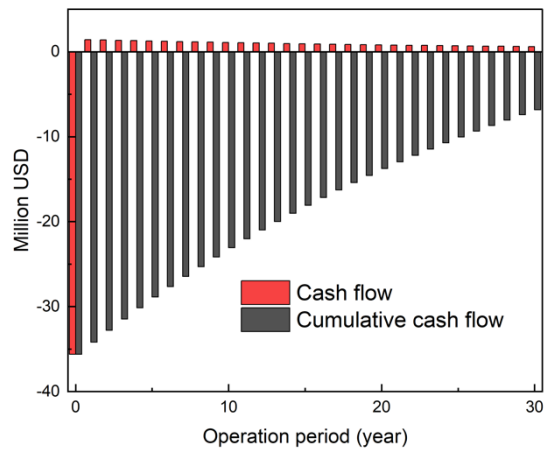


Fig. 4.2 Cash flow and cumulative cash flow change over the operation period (E_{conv} : 20 MJ/kg-biomass, C_{MW} : 100,000 USD/unit, k_{H_2} :14.7 USD/kg).

4.3.2 Sensitivity analysis and payback period

Fig. 4.3 shows the sensitivity analysis for the IRR and the NPV when the E_{conv} and the C_{MW} are 20 MJ/kg-biomass and 100,000 USD/unit, respectively. The range of variables of 0 % means that the E_{conv} and C_{MW} are 20 MJ/kg-biomass and 100,000 USD/unit, respectively. The E_{conv} ranged from -25 % (15 MJ/kg-biomass) to 0 % (20 MJ/kg-biomass). The C_{MW} ranged from -33.3 % (66,667 USD/unit) to 0 % (100,000 USD/unit). Section 4.3.1 showed that the microwave biomass conversion system was not an economically feasible investment when the E_{conv} and the C_{mw} were 20 MJ/kg-biomass and 100,000 USD/unit, respectively. Therefore, the present study demonstrates the IRR and the NPV when the E_{conv} and the C_{mw} are reduced. When the E_{conv} was reduced by 10 % (18 MJ/kg-biomass), the IRR was higher than the discount rate (3 %), and the NPV became positive. Moreover, the initial cost was recovered within the system's lifetime (Fig. 4.4 (a)). Therefore, the E_{conv} must be reduced by 10 % to become economically feasible when the C_{MW} is fixed at 100,000 USD/unit. When the C_{MW} was reduced by 13 % (86,667 USD/unit), the IRR was higher than the discount rate, and the NPV became positive. In addition, the initial cost was recovered within the system's lifetime (Fig. 4.4 (b)). Therefore, the C_{MW} must be reduced by 13 % to become economically feasible when the E_{conv} is fixed at 20 MJ/kg-biomass.

Fig. 4.5 shows the IRR and the NPV when the E_{conv} and the C_{MW} are varied in combination. When the E_{conv} and the C_{MW} were reduced by 5 % (19 MJ/kg-biomass) and 7 % (93,333 USD/unit), respectively, the IRR was higher than the discount rate, and the NPV became positive. When the E_{conv} and the C_{MW} were reduced by 25 % (15 MJ/kg-biomass) and 33 % (66,667 USD/unit), the IRR and the NPV were 10.6 % and 31.7 million USD. These results indicate that reducing the E_{conv} and the C_{MW} effectively makes microwave biomass conversion systems more economical. To reduce the E_{conv} , the methods for more energy-efficient decomposition of biomass need to be developed. In the case of microwave biomass conversion, selecting the microwave susceptor and developing a microwave cavity to decompose the biomass efficiently are considered necessary. 5.2 Future works discuss the method to reduce the E_{conv} . To reduce the C_{MW} , improvements in the manufacturing technology of microwave heating systems are considered necessary. In addition, the use of Japanese government subsidies is effective. On the other hand, to introduce subsidies, the evaluation of social acceptability is necessary. Section 4.3.4 evaluates the social acceptability of the microwave biomass conversion system.

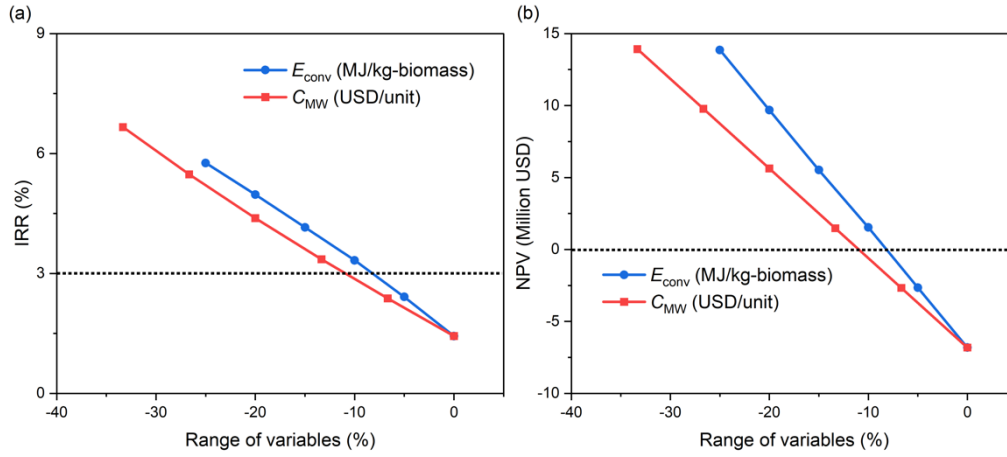


Fig. 4.3 Sensitivity analysis for (a) the IRR and (b) the NPV ($15 \leq E_{conv} \leq 20$ MJ/kg-biomass, $66,667 \leq C_{MW} \leq 100,000$ USD/unit, k_{H_2} is fixed at 14.7 USD/kg)

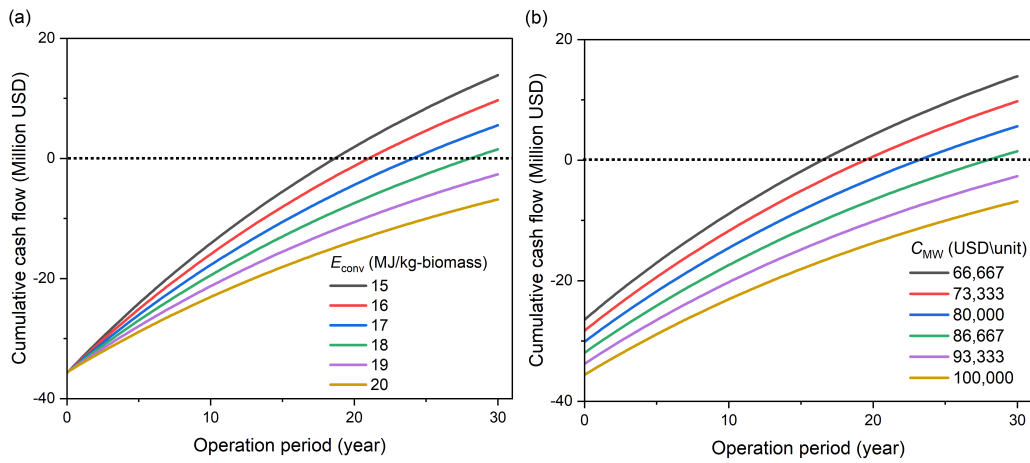


Fig. 4.4 Change in cumulative cash flow relative to change in (a) E_{conv} and (b) C_{MW} (k_{H_2} is fixed at 14.7 USD/kg)

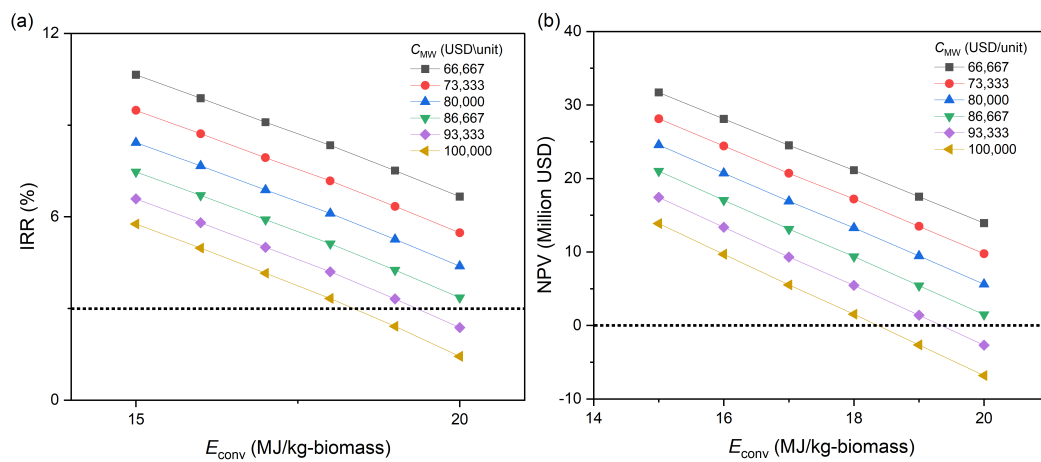


Fig. 4.5 Changes in (a) IRR and (b) NPV relative to changes in C_{MW} and E_{conv} (k_{H_2} is fixed at 14.7 USD/kg).

4.3.3 H₂ price in 2030 model

The Japanese government aims to reduce the hydrogen sales price to 2.33 USD/kg-H₂ by 2030, so the price will likely decline. Fig. 4.6 shows the change in cumulative cash flow when k_{H_2} is fixed at 2.23 USD/kg. The initial cost was not recovered within the system's lifetime of every E_{conv} and C_{MW} . Moreover, the initial cost was not recovered within the system's lifetime when the E_{conv} and the C_{MW} were 15 MJ/kg-biomass and 66,667 USD/unit, respectively (shown in Fig. 4.7 (a)). To be economically feasible, the quantity discount is applied to reduce the initial cost of the microwave biomass conversion system. Fig. 4.7 (b) shows that the cash flow and cumulative cash flow change over the operation period when the quantity discount to initial cost and magnetron replacement is applied to 50 %. As a result, the initial cost was recovered within the system's lifetime by applying the quantity discount of 50 %. However, the quantity discount rate is subject to negotiation, and a 50 % discount cannot be guaranteed with certainty. Therefore, other methods to reduce initial costs must be considered. The application of subsidies is also effective in reducing the initial cost. In 2024, The Japanese government provided several billion USD in support for renewable energy projects [147]. The initial cost of the microwave biomass conversion system, which consisted of 264 units, was about 30 million USD. Thus, Japanese government subsidies could cover the initial cost of the microwave biomass conversion system. On the other hand, to introduce subsidies, it is necessary to demonstrate not only economic viability but also social acceptability. The following section evaluates the social acceptability of the microwave biomass conversion system.

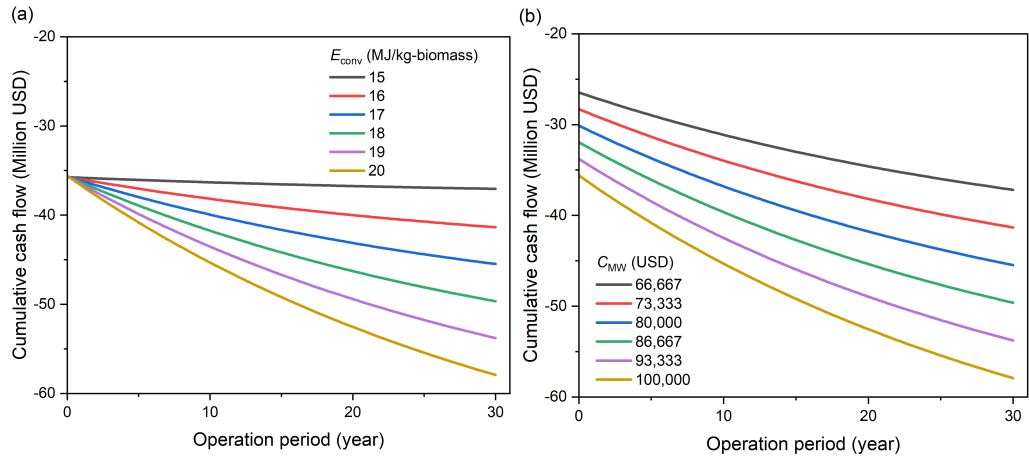


Fig. 4.6 Change in cumulative cash flow when k_{H_2} is fixed at 2.23 USD/kg relative to change in (a) E_{conv} and (b) C_{MW} .

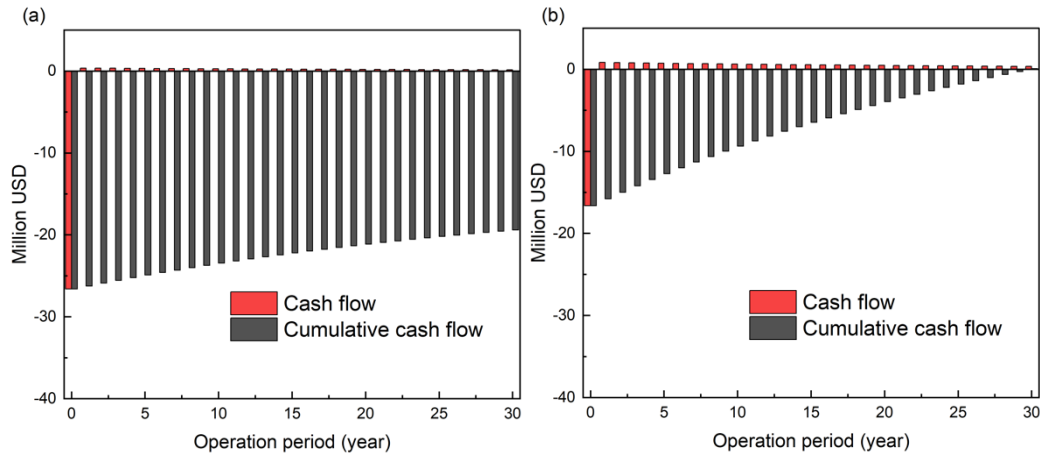


Fig. 4.7 Cash flow and cumulative cash flow change over the operation period, E_{conv} : 15 MJ/kg-biomass, C_{mw} : 66,667 USD/unit, k_{H_2} : 2.23 USD/kg, the quantity discount to initial cost and magnetron replacement: (a) 20 %, (b) 50 %.

4.3.4 Power Conditioning Ability and Utilization ratio of forest residues in Kyushu

The power conditioning ability in Kyushu (A_{power}) was calculated using Eq. (4.11). The input energy for the microwave oscillation (E_{MW}) depends on the number of microwave heating units (N), which in turn depends on the energy requirement for biomass conversion (E_{conv}). Fig. 4.8 shows that the N and the A_{power} change over the E_{conv} . The N and the A_{power} increased with increasing in the E_{conv} . The maximum A_{power} was 31 % when the E_{conv} was 20 MJ/kg-biomass, while the minimum A_{power} was 24 % when the E_{conv} was 15 MJ/kg-biomass. This result indicates that reducing the E_{conv} is economically favorable while decreasing the power conditioning ability. The power conditioning ability of the microwave biomass conversion system was lower than that of the pumped storage power plant. On the other hand, the equipment cost of the microwave biomass conversion system (104 USD/kWh) was lower than that of the pumped storage (233 USD/kWh) [148]. The equipment cost of the NAS storage battery is 467 USD/kWh [149]. The equipment cost of the microwave biomass conversion system was lower than that of the NAS storage battery. Therefore, the microwave biomass conversion system can be introduced more reasonably than the pumped storage and the NAS storage battery. Moreover, the convenience of energy after storage with the microwave biomass conversion system was higher than that with the pumped storage power plant. In the case of the pumped storage power plant, energy is stored as electricity and emptied daily to secure the energy storage for the next day. In the case of the microwave biomass conversion system, energy is stored as hydrogen, carbon monoxide, methane, etc. If spare tanks are prepared, the stored energy does not need to be used daily, and the convenience is considered higher than that of pumped storage. The utilization ratio of forest residues (U_{residue}) in Kyushu was calculated using Eq. (4.13). The annual treatment amount of the microwave biomass conversion system (M_{treat}) was 149,665 t/year (= 9,978 t/year/site $\times$ 15 sites). As a result, the U_{residue} was 10.6 %. Therefore, the microwave biomass conversion system improves the utilization rate of forest residues. The existing biomass power plant utilizes approximately 10,000 tons of biomass per year, the same as the microwave biomass conversion system [150]. Therefore, the microwave biomass conversion system is expected to improve the utilization rate of forest residues to the same extent as the introduction of biomass power generation. On the other hand, the output of biomass power generation is reduced when there is excess grid power. Considering the excess power will increase, the microwave biomass

conversion system is more beneficial than biomass power generation because it can effectively use excess electricity and forest residues simultaneously.

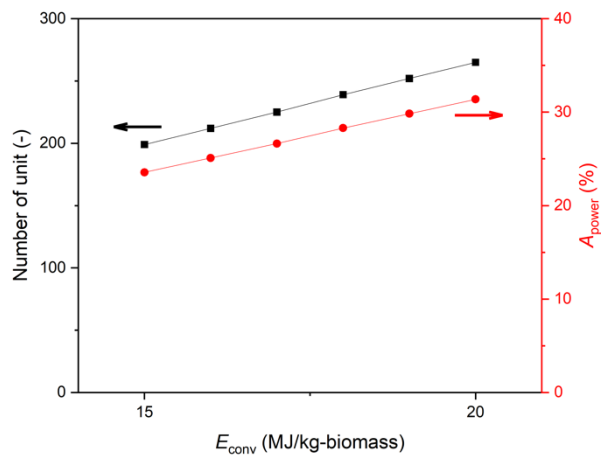


Fig. 4.8 Number of microwave heating units and A_{power} change over the E_{conv} .

4.4 Conclusions

The present study performed the techno-economic assessment of the microwave biomass conversion system to demonstrate economic and social acceptability. Based on the experimental data of the previous study, the internal rate of return (IRR) and the net present value (NPV) were 1.43 % and -6.81 million USD, respectively, when the required energy for biomass conversion (E_{conv}) was 20 MJ/kg-biomass. Moreover, the initial cost was not recovered within the system's lifetime. These results indicated that the microwave biomass conversion system was not economically feasible. To make this system economically feasible, the E_{conv} and the cost of the microwave heating unit (C_{MW}) must be reduced by 10 % and 13 %, respectively. In the future, research and development are needed to reduce the E_{conv} and the C_{MW} . Regarding the hydrogen sales price in 2030, the microwave biomass conversion system was not economically feasible when the E_{conv} and the C_{MW} were minimum values. The quantity discount of 50 % must be applied to the initial cost to become economically feasible. To evaluate the social acceptability of the microwave biomass conversion system in Kyushu, the power conditioning ability (A_{power}) and the utilization rate of forest residues (U_{residue}) were calculated. As a result, the maximum A_{power} was 31 % when the E_{conv} was 20 MJ/kg-biomass, while the minimum A_{power} was 24 % when the E_{conv} was 15 MJ/kg-biomass. The U_{residue} was 10.6 %, comparable to existing biomass power generation. These results suggest that the microwave biomass conversion system functions as a power conditioning and contributes to improving the utilization of forest residues. The Japanese government may provide subsidies for the microwave biomass conversion system because this system can simultaneously solve the problems of excess solar power and forest residues.

The present study assumes a 90 % reduction in heat dissipation based on lab-scale experiments. Although a method of reducing heat dissipation by 90 % has been proposed, it has not yet been implemented. Furthermore, experimental data on heat dissipation in the scaled-up system has not been obtained. Therefore, obtaining experimental data from the scaled-up system will enable a more precise evaluation for social implementation and facilitate a suitable economic assessment. Regarding syngas production, the theoretical value of the E_{conv} is 5 MJ/kg-biomass when the reaction in Eq. (4.10) is assumed. Even if the heat dissipation is reduced, the E_{conv} is 10 MJ/kg-biomass. Thus, the E_{conv} can be further reduced, shifting the system in a more economically viable direction. In the future, developing a microwave heating reaction field to reduce the E_{conv} is expected to demonstrate the effectiveness of the microwave biomass conversion system.

5. Conclusions and future works

5.1 Conclusion

The present study focused on microwave heating (MWH), which is anticipated to be more efficient than conventional heating (CH) for syngas production from biomass through thermochemical conversion. Additionally, the economic feasibility and social acceptability of the microwave biomass conversion system are assessed.

In Chapter 2, the effect of the magnetite addition was evaluated. It was mixed with cellulose for microwave heating at 800 °C. The results revealed that syngas yield increased with the addition of magnetite, having a maximum yield of 19.1 mmol/g-cellulose with a magnetite–cellulose weight ratio of 1. This yield was higher than conventional heating at 800 °C and equivalent to conventional heating at 1000 °C. Moreover, the heat transfer equation calculation indicated that microwave heating induces internal heating when the sample section is volumetrically heated. These results suggest that the magnetite addition induces internal heating, which increases syngas yield and improves energy efficiency. However, the syngas yield of microwave heating decreased by 40 % with an increasing cycle number owing to the reduction of magnetite to iron. This result suggests that the sample section is not volumetrically heated because iron reflects microwaves. Thus, steam oxidation treatment was performed to regenerate magnetite. With the treatment, the syngas yield of the regenerated magnetite was like that in the 1st cycle. Therefore, steam oxidation is effective for the repeated use of magnetite.

Based on the findings in Chapter 2, Chapter 3 examines the potential of microwave heating for syngas production from cellulose in a steam atmosphere using magnetite as a susceptor. The effects of steam feeding and microwave heating on product yields were studied and compared to conventional heating. The results demonstrated that steam feeding improved H₂ and CO₂ yields in MWH and CH due to the water-gas shift reaction. However, with steam feeding, steam gasification of carbon was more prevalent in CH than in MWH, as indicated by a more significant reduction in carbon content in the char from CH. Conversely, MWH minimized secondary char formation, likely alleviating operational challenges such as gas line clogging or blockage. These findings imply that MWH with steam feeding presents a viable and efficient method for H₂ production while addressing issues related to secondary char, indicating that the possibility of

continuous operation is also expected. The SEM images of MWH without steam feeding suggest that the pig iron melts. The melting point of pig iron is 1147 °C. In Chapter 2, the gas analysis shows that the gas yield of MWH 800 °C and CH1000 °C were similar. Moreover, the heat transfer calculation suggests that the cylindrical center temperature reaches 990 °C. These results indicate that the internal heating forms inside the sample section.

Expanding on the experimental findings from Chapters 2 and 3, this chapter evaluates the techno-economic feasibility and social acceptability of the microwave biomass conversion system. Based on the experimental data of the previous study, the internal rate of return (IRR) and the net present value (NPV) were 1.43 % and − 6.81 million USD, respectively, when the required energy for biomass conversion (E_{conv}) was 20 MJ/kg-biomass. Moreover, the initial cost was not recovered within the system's lifetime. These results indicated that the microwave biomass conversion system was not economically feasible. To make this system economically feasible, the E_{conv} and the cost of the microwave heating unit (C_{MW}) must be reduced by 10 % and 13 %, respectively. In the future, research and development are needed to reduce the E_{conv} and the C_{MW} . Regarding the hydrogen sales price in 2030, the microwave biomass conversion system was not economically feasible when the E_{conv} and the C_{MW} were minimum values. The quantity discount of 50 % must be applied to the initial cost to become economically feasible. To evaluate the social acceptability of the microwave biomass conversion system in Kyushu, the power conditioning ability (A_{power}) and the utilization rate of forest residues (U_{residue}) were calculated. As a result, the maximum A_{power} was 31% when the E_{conv} was 20 MJ/kg-biomass, while the minimum A_{power} was 24 % when the E_{conv} was 15 MJ/kg-biomass. The U_{residue} was 10.6 %, comparable to existing biomass power generation. These results suggest that the microwave biomass conversion system functions as a power conditioning and contributes to improving the utilization of forest residues. The Japanese government may provide subsidies for the microwave biomass conversion system because this system can simultaneously solve the problems of excess solar power and forest residues.

Overall, microwave heating presents a promising approach to biomass conversion, offering advantages in efficiency and sustainability. The present study indicates that magnetite addition induces internal heating, leading to the improvement of syngas production. Moreover, the combination of microwave heating and steam-feeding reduces secondary char formation, indicating the possibility of continuous operation. The present study successfully provides

insights into microwave biomass conversion using a susceptor.

5.2 Future works

Future work needs to analyze oil and measure the temperature inside samples. Determining the oil products can indicate the usefulness of the magnetite microwave biomass conversion system. Chapter 4 indicates that the required energy for biomass conversion needs to be reduced in terms of economic feasibility. The required energy considering the reduction of heat dissipation is 20 MJ/kg-biomass in the present study. On the other hand, the required energy of biomass steam gasification to syngas is 5 MJ/kg-biomass. Therefore, the required energy for biomass conversion can still be reduced by suppressing heat dissipation and improving the microwave heating technique. To reduce the heat dissipation, appropriate heat insulation needs to be installed. Moreover, the control of hot spots is effective. The present study suggests that low thermal conductivity improves the internal heating effect. Thus, the effect of internal heating should be optimized by selecting a susceptor regarding thermal conductivity in the future. The present study discussed the temperature distribution of the mm scale. To discuss this more properly, it is necessary to clarify the temperature distribution on the scale from μm to nm during microwave heating. Moreover, the temperature distribution during microwave heating undergoes complex changes due to temporal variations. The establishment of a real-time and trans-scale temperature measurement method is required. Kishimoto et al. investigated the selective heating of ions in a zeolite (Cs⁺-FAU) [151]. They conducted the in situ high-energy x-ray total scattering (HEXTS) measurement in the microwave heating of zeolite. HEXTS obtained the X-ray peak pattern $S(Q) = S(Q)^0 \exp\{-M_D(T)\}$, ($Q = 4\pi/\lambda \sin\theta$, $M_D(T)$: Debye-Waller factor, λ : X-ray wavelength, θ : angle of diffraction). The pair Distribution Function ($G(r)$) can be obtained by the Fourier transform of $S(Q)$. Fig. 5.1 shows the experimental setup for in situ high-energy x-ray total scattering (HEXTS) measurement in microwave heating of zeolite (Cs⁺-FAU) and the results of the pair Distribution Function. The characteristic shifts under microwave heating were observed in the peaks attributed to the pair distribution function (PDF) around Cs⁺. That is, the peak at 3.1 Å attributed to the Cs-O pair was shifted to longer, while the T-O or Cs-T pair peak at 3.8 Å and the T-O, T-T, or Cs-O pair peak at 4.3 Å was shifted to shorter compared to the $G(r)$ of conventional heating as the temperature rose under microwave heating. They demonstrate that

atomic-level selective heating occurs through microwave heating by real-time and nm scale measurement. Establishing a real-time and trans-scale temperature measurement method will deepen the academic knowledge of microwave biomass conversion technology, and the control indicators for hot spots will be determined. It is expected that controlling hot spots enables putting in as much energy as needed for the reaction. Microwave biomass conversion system can be realized by reducing the required energy for biomass conversion. Finally, establishing a syngas production process independent of fossil resources is also anticipated.

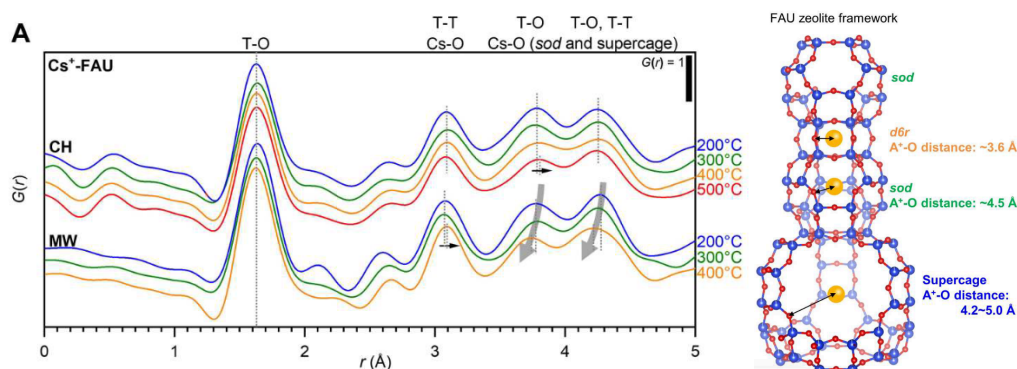
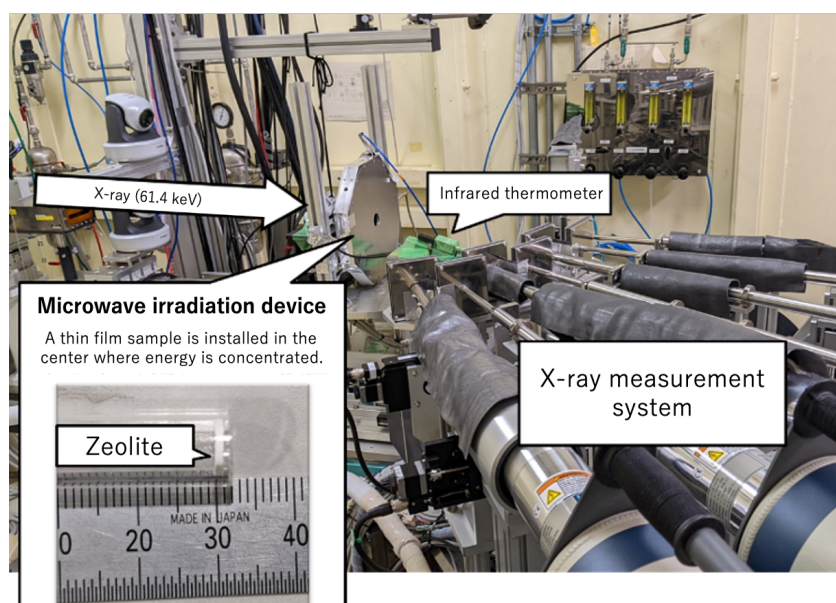


Fig. 5.1 Experimental setup for in situ high-energy x-ray total scattering (HEXTS) measurement in microwave heating of zeolite (Cs⁺-FAU) [151].

Appendix

Python code for calculation of heat transfer equation

```
import numpy as np

import matplotlib.pyplot as plt

from scipy.integrate import solve_ivp


# constant

R = 0.018 # radius [m]

L = 0.039 # length [m]

Q_total = 500 # input energy [W]

k = 4.02 # thermal conductivity [W/(m*K)]

rho = 1306 # mass density [kg/m^3]

c_p = 1124 # specific heat [J/(kg*K)]


# Stefan-Boltzmann constant, emissivity

epsilon = 1 # emissivity

sigma = 5.67e-8 # Stefan-Boltzmann constant [W/(m^2 K^4)]

T_env = 20 # environmental temperature [°C]


# volumetric heat

V = np.pi * R**2 * L # volume of sample [m^3]

Q = Q_total / V # volumetric heat amount [W/m^3]
```

```

# boudary condition

T_center_initial = 800 # initial temperature [°C]

T_surface_fixed = 800 # surface temperature [°C]


# mesh size

dr = 0.006

radius_values = np.arange(0, R + dr, dr) # 0.006m

num_points = len(radius_values) # calculation points


# definition of equations considering time evolution

def heat_eq(t, T):

    dTdt = np.zeros_like(T)

    for i in range(1, num_points - 1):

        r_i = radius_values[i]

        d2Tdr2 = (T[i+1] - 2*T[i] + T[i-1]) / dr**2

        dTdr = (T[i+1] - T[i-1]) / (2 * dr)


        # internal radiant heat transfer

        T_i_K = T[i+1] + 273.15 # temperature of adjacent elements [K]

        T_ip1_K = T[i-1] + 273.15 # temperature of adjacent elements [K]

        q_rad_internal = epsilon * sigma * (T_ip1_K**4 - T_i_K**4) / (2 * dr) # internal
radiant heat


        dTdt[i] = (k / (rho * c_p)) * (d2Tdr2 + (1/r_i) * dTdr) + Q / (rho * c_p) -

```

$q_{\text{rad_internal}} / (\rho * c_p)$

temperature change at center

$T_{\text{center_K}} = T[0] + 273.15$

$T_{1_K} = T[1] + 273.15$ # temperature of adjacent elements [K]

$q_{\text{rad_internal_center}} = \epsilon * \sigma * (T_{\text{center_K}}^{**4} - T_{1_K}^{**4}) / (dr)$ # internal
radiant heat

$dTdt[0] = 2 * (k / (\rho * c_p)) * (T[1] - T[0]) / dr^{**2} + Q / (\rho * c_p) - 2 * q_{\text{rad_internal_center}} / (\rho * c_p)$

heat dissipation from surface

$T_{\text{surface_K}} = T[3] + 273.15$

$T_{\text{env_K}} = T_{\text{env}} + 273.15$

$q_{\text{rad_surface}} = \epsilon * \sigma * (T_{\text{surface_K}}^{**4} - T_{\text{env_K}}^{**4})$

$heat_loss = q_{\text{rad_surface}} / (\rho * c_p * dr)$

$dTdt[3] -= heat_loss$ # heat dissipation from surface

surface temperature fixed

$dTdt[3] = 0$ # change in surface temperature to zero

$T[3] = T_{\text{surface_fixed}}$ # surface temperature 800 C

return dTdt

calculation time

```

t_span = (0, 100) # range of calculation time [s]

t_eval = np.arange(0, 101, 10) # 10 s step

T_init = np.linspace(T_center_initial, T_surface_fixed, num_points)

sol = solve_ivp(heat_eq, t_span, T_init, t_eval=t_eval)

# plot

plt.figure()

for i in range(len(sol.t)):

    plt.plot(radius_values, sol.y[:num_points, i], marker='o', label=f"t={sol.t[i]:.1f}s")

plt.xlabel("Radius [m]")

plt.ylabel("Temperature [°C]")

plt.legend(loc='upper right')

plt.grid()

plt.show()

```

References

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List of Publications

- **Hiroyuki Tamiya**, Sadatsugu Takayama, Keisuke Mukai, Juro Yagi, “Effects of steam feeding on microwave heating of cellulose with magnetite used as susceptor”, *International Journal of Hydrogen Energy*, Vol. 71, pp.1–7, June 2024
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List of Presentations

International (oral)

- **Hiroyuki Tamiya**, Keisuke Mukai, Sadatsugu Takayama, Juro Yagi, “Effect of Fe_3O_4 grain size on Microwave Pyrolysis of Cellulose”, 57th Annual Microwave Power Symposium (IMPI 57), Denver, Colorado, USA, Jun 29th, 2023,
Adoption of Student Travel Scholar Ship
- **Hiroyuki Tamia**, Yang Hui, Khadija Usher, Soumya Basu, Rei Akasegawa, Qu Chen, Jordi Cravioto, Hideaki Ohagaki, “Synergies and Trade-offs between SDGs and 3R policies of Kyoto City”, KYOTO-AJOU-ZHEJIANG Joint Symposium on Energy Science, Kyoto University, Kyoto, Japan, Dec 9th, 2022
- **Hiroyuki Tamiya**, Keisuke Mukai, Sadatsugu Takayama, Juro Yagi, “Influence of temperature on microwave heating of cellulose with magnetite under steam atmosphere”, The 5th Global Congress on Microwave Energy Applications, Kyusyu University, Fukuoka, Japan

Japanese (oral)

- 田宮裕之、向井啓祐、高山定次、八木重郎「マイクロ波加熱による木質バイオマスガス化における水蒸気供給の効果」、『第 18 回若手研究者による研究発表会』、2023 年 3 月 1 日
- Hiroyuki Tamiya, Keisuke Mukai, Sadatsugu Takayama, Juro Yagi, “Effect of steam feeding on microwave pyrolysis of cellulose”, The 17th Symposium of Japan Society of Electromagnetic Wave Energy Applications (Sympo2023), Kokura, Fukuoka, Japan, Oct 26th, 2023, Oral, Best Presentation Award

Japanese (poster)

- 田宮裕之、八木重郎、向井啓祐、高山定次、小西哲之、「二酸化チタン共存下セルロースのガス化におけるマイクロ波効果」、『公益社団法人日本セラミックス協会 2022 年年会』、オンライン発表、2022 年 3 月 10 日-12 日
- 田宮裕之、向井啓祐、八木重郎、高山定次、小西哲之「マイクロ波による木質バイオマスの熱分解及び水素生成」、『第 14 回 核融合エネルギー連合講演会』、オンライン発表、2022 年 7 月 7 日-8 日

List of Award

- Hiroyuki Tamiya, Keisuke Mukai, Sadatsugu Takayama, Juro Yagi, “Effect of steam feeding on microwave pyrolysis of cellulose”, The 17th Symposium of Japan Society of Electromagnetic Wave Energy Applications (Sympo2023), Kokura, Fukuoka, Japan, Oct 26th, 2023, Oral, Best Presentation Award

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