#### 1 Acoustic Emission Monitoring of Hydraulic Fracturing Using Carbon Dioxide in a

2 Small-scale Field Experiment

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#### 13 Abstract

- Carbon capture and storage (CCS) is a promising method for mitigating the greenhouse 14 15 effect. If we could utilize carbon dioxide (CO<sub>2</sub>) for recovery of geothermal energy and 16 shale gas, CCS will be much more eagerly developed and adopted by compensating its cost. We clarified that hydraulic fracturing (HF) using CO<sub>2</sub> tended to induce three-17 dimensionally sinuous cracks with many secondary branches, which appear to be 18 19 desirable pathways for energy recovery. However, in a laboratory experiment, it was difficult to evaluate crack extension size and effects of macroscopic pre-existing cracks. 20 21 Thus, we conducted a HF experiment using CO<sub>2</sub> in a hole 10 m long that was drilled in a hot rock mass, thus satisfying the temperature necessary to form supercritical (SC) 22 CO<sub>2</sub>. A bi-wing crack 3.33 m in length was induced along the hole; this crack was 23 significantly larger than the approximately 0.5 m long crack we made with water at 24 another site. The fact that acoustic emission (AE) hypocenters distributed in almost 25 perpendicular to an initial HF crack direction without pressure suggested that CO<sub>2</sub> could 26 easily intrude and enhance AE occurrence along a pre-existing crack. Focal mechanisms 27 using P wave first-motion polarities elucidated that many compression-dominant AE 28 events were recorded; these were never observed in similar HF experiment we 29 conducted using water. The compression-dominant events were probably induced by 30 crack closure due to the degassing of injected CO<sub>2</sub>. Even if CO<sub>2</sub> injection induces many 31 AE events, the compressive-dominant events out of them unlikely trigger natural 32
- 33 earthquakes because they never create new cracks.

#### 34 **1 Introduction**

Carbon capture and storage (CCS) in underground reservoir is a promising and 35 feasible method for mitigating the greenhouse effect by decreasing the amount of 36 carbon dioxide ( $CO_2$ ) emitted into the air. If we could utilize  $CO_2$  for energy 37 production, CCS will be much more eagerly developed and adopted by compensating its 38 cost. The technology utilized is called carbon capture, utilization, and storage (CCUS), 39 and for examples of applications of CCUS to underground resources, Xie et al.<sup>1</sup> shows 40 the recovery of enhanced geothermal systems (CO<sub>2</sub>-EGS), enhanced shale gas (CO<sub>2</sub>-41 ESG), enhanced natural gas (CO<sub>2</sub>-EGR), enhanced coalbed methane (CO<sub>2</sub>-ECBM), 42 enhanced oil (CO<sub>2</sub>-EOR) and others. Among these, for geothermal energy extraction in 43 EGS, Brawn<sup>2</sup> and Pruess<sup>3</sup> stated that  $CO_2$  is superior to water as a working fluid. 44 Regarding exploitation of shale gas in ESG, Kalantari-Dahaghi, <sup>4</sup> Liu et al., <sup>5</sup> Middleton 45 et al.<sup>6</sup> and Godec et al.<sup>7</sup> suggested that it is feasible through numerical simulations, 46 because CO<sub>2</sub> has a higher affinity for shale than methane (CH<sub>4</sub>) and CH<sub>4</sub> is desorbed 47 with the absorption of CO<sub>2</sub>.<sup>8</sup> For these projects, CO<sub>2</sub> is injected into rocks usually at a 48 depth of more than 1000 m and sometimes more than 3000 m, and the temperature and 49 pressure at these depths cause the CO<sub>2</sub> to reach its supercritical (SC) state. SC-CO<sub>2</sub> has 50 a much lower viscosity (0.01 mPa $\cdot$ s to 0.1 mPa $\cdot$ s) than that of water (1 mPa $\cdot$ s). We 51 clarified that hydraulic fracturing (HF) using SC-CO<sub>2</sub> tends to induce three-52 dimensionally sinuous cracks with many secondary branches, which appear to be 53 desirable pathways for such projects.<sup>9, 10</sup> 54 However, in a laboratory experiment, it is difficult to evaluate the crack extending 55 size and effects of pre-existing cracks because the cracks extend to the surfaces of the 56 specimen immediately after breakdown (BD) and a specimen usually does not contain a 57 macroscopic pre-existing crack. Thus, we conducted a HF experiment using CO<sub>2</sub> in a 58 hole 10 m long drilled from the tunnel floor of a hot rock mass, which had a pre-59 existing crack that satisfied the temperature necessary for SC-CO<sub>2</sub> to form. After we 60 conducted this experiment in 2014, we reported the experimental methods and 61 preliminary results on a relation between temporal change of AE source distribution and 62 a pre-existing crack in the symposium of EUROCK 2017.<sup>11</sup> In the symposium of 63 EUROCK 2018, we analyzed the focal mechanisms of these AE events and showed that 64 many of them show compression-dominant mechanism with shear-dominant 65 mechanism.<sup>12</sup> In this paper, we newly examine a size of HF cracks induced by the SC-66 CO<sub>2</sub> injection using images obtained by a borehole camera. In addition, we newly 67 examine relations between locations and focal mechanisms of the AE events, to clarify a 68 69 difference of effect on the focal mechanism between a new crack and a pre-existing

- rocrack. Through the new examinations, we concurrently discuss and elucidate
- remarkable features of HF using CO<sub>2</sub> in comparison to conventional HF using water,
- and effects of a pre-existing crack on them.
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# 74 2 Site and Experimental Setup

### 75 2.1 Site and Method of CO<sub>2</sub> Injection

76 As shown in Figure 1, the experimental site was situated in a small railroad tunnel 77 approximately 50 m below the surface of a mountainous area in central Japan. The rock mass around the site consisted of granitic rock formed from the late Miocene to the 78 79 Pliocene. Although the rock temperature was 165 °C when this tunnel was excavated in 80 1940, it is now 35 °C due to a cooling effect from air circulating through the tunnel for an extended time. A HF hole (86 mm in diameter) was drilled vertically downward from 81 the tunnel floor, and four acoustic emission (AE) monitoring holes (66 mm in diameter 82 each) were drilled parallel to and at 1 m from the HF hole. To inject CO<sub>2</sub>, we drilled a 83 pilot hole 36 mm in diameter at the center of the bottom of the HF hole. We selected the 84 85 pressurizing section from intact rock at a depth of 7.24 m to 7.40 m, where no visible crack was found, by checking the recovered core of the pilot hole. We sealed the upper 86 section of the pilot hole with two O-rings that were attached to a packer unit and poured 87 cement paste above the O-rings, as shown in the inset of Figure 1b. CO<sub>2</sub> was injected 88 into the pressurizing section under the O-rings. 89

Figure 2 shows the injection system used in the experiment. To inject  $CO_2$ , we used two syringe pumps and alternated between them to achieve continuous injecting during the experiment. We fed  $CO_2$  from a bomb into the syringe-pump cylinders, each of which had a capacity of 500 mL. To fill the cylinders as full as possible, we circulated coolant around them to cool the  $CO_2$  therein and to keep it in the liquid state. In the experiment, the liquid  $CO_2$  was discharged from the cylinder at a constant flow rate of

- system experiment, the right CO<sub>2</sub> was discharged from the cynneer at a constant now rate of
- 50 mL/min by controlling the displacement of the syringe. The discharged CO<sub>2</sub> was
- <sup>97</sup> heated up to approximately 50°C by a heater and was injected into the sealed section of
- the rock at a temperature of  $35^{\circ}$ C. As shown in the phase diagram of Figure 3, CO<sub>2</sub>
- reached a SC state at this temperature under the pressure higher than 7.38 MPa.
- 100 The  $CO_2$  was injected into a rock mass saturated with water because the groundwater 101 level was approximately 1 m below the tunnel floor.
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#### 103 **2.2 Measurement of AE, Pressure, and Temperature**

For AE monitoring, we installed four waterproof lead–zirconate–titanate (PZT) sensors with a resonance frequency of 70 kHz (AE703SW-GAMP-0542; Fuji Ceramics

Corp., Japan) in each of the four AE holes (AE1-4 in Figure 1b). We attached each sensor 106 to an aluminum rod with a pre-amplifier and inserted a thick rubber sheet between the 107 sensor and the rod to block any vibrations transmitted through the rod. After orienting 108 each sensor to face the HF hole, we pressed them onto the walls of the AE holes by 109 110 applying 1.5 MPa of oil pressure in a small hydraulic jack set behind each sensor. In each 111 AE hole, we installed four sensors over a 2 m long span at intervals of 0.7 m, 0.6 m, and 0.7 m to center the 2 m section at the center depth of the pressurizing section of the HF 112 113 hole.

The recording of an AE event was triggered when one of the signals from the 16 AE 114 115 sensors (in total) set in the four holes exceeded 1 V. After the AE signals detected at the 116 sensors were amplified by 40 dB in a pre-amplifier and 30 dB in a main-amplifier, they 117 were processed with a band-pass filter between 20 kHz and 200 kHz and recorded on a hard disk through an analog-to-digital (A/D) converter (PXI-5105; National Instruments 118 Corp., USA). The AE signal of each sensor for each event was digitized into 2048 samples 119 with a 1 µs sampling time. We stopped recording for 10 ms, just after recording an event 120 121 for 2.048 ms, to prevent the hard disk from recording too much noise caused by "ringing," which was the vibration following a large AE event. 122

We measured the injection pressure with a transducer (PW-50MPA; Tokyo Sokki Kenkyujo Co., Ltd., Japan) that was set on the injection pipe on the floor just outside of the HF hole. The temperature was measured with a T-type thermocouple glued onto the injection pipe just above the cement paste sealing the pressurized section in the HF hole. The pressure and temperature data were recorded by another A/D converter (PXI-6251; National Instruments Corp., USA) with a sampling time of 0.1 s.

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#### 130 2.3 AE Data Processing for Hypocenter Location

For hypocenter location, we adopted the least square iterative method using P wave 131 arrival time. We measured the P wave velocities between the HF hole and the 16 132 sensors just before and after the HF by using an emitter (AE703SWR-0840; Fuji 133 134 Ceramics Corp., Japan) attached to the upper part of the packer unit. After confirming 135 that the difference in P wave velocities before and after the HF was small, the average velocity of 5.4 km/s (standard deviation of 0.4 km/s) was used for the hypocenter 136 locations under the assumption that the rock mass was isotropic and homogeneous. 137 In the following analysis, we used well-located hypocenters whose accuracy is 138 expected to be within 50 mm by satisfying the following two conditions:<sup>13</sup> (1) The P 139

140 wave arrival times could be read at six or more sensors set in three or more different AE

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141 holes to enclose the hypocenter three-dimensionally; (2) the standard deviation and the

- maximum of residuals of arrival times were within 10  $\mu$ s and 20  $\mu$ s, respectively.
- 143 Satisfying these conditions, we located the hypocenters of 1249 AE events during the
- 144 CO<sub>2</sub> injection.
- 145

# 146 **3 Results**

#### 147 **3.1 Temporal Changes in Fluid Pressure, Temperature, AE Rate, and Flow Rate**

Figure 4 shows the temporal changes in fluid pressure, temperature, AE rate and flow rate with elapsed time, *t*. We took t = 0 as the time when the pressure reached breakdown (BD) pressure, which was defined as the peak pressure immediately before the large sudden drop. Here, the AE rate was counted for only well-located events (see Section 2.3).

When we opened the valve of the injection system, the bomb pressure of the  $CO_2$ (approximately 1 MPa) acted on the pressurized section. We injected  $CO_2$  at a constant flow rate of 50 mL/min, with a sudden stop for 30 s (from -15 min 13 s to -14 min 43 s), due to a problem with switching the two syringe pumps. The injected pressure increased with the elapsed time, and BD was induced at 15.62 MPa. Immediately after BD, the injection was stopped. A few minutes after BD, we found that bubbles of  $CO_2$  were gushing out from the water that filled the AE1 and AE3 holes.

Initially, the pressure increased gradually with time, and then it increased steeply at 160 161 approximately t = -3 min (indicated by the thick arrow in Figure 4a). This steep increase started at a pressure close to the critical pressure of 7.38 MPa. At that time, the 162 temperature was approximately 35 °C, which was higher than the critical temperature of 163 31.1 °C. Thus, the steep increase was most likely caused by the decrease in CO<sub>2</sub> 164 165 compressibility, due to the phase change from the gaseous state to the SC state (see Figure 3). The temperature decreased to 32.6 °C at BD, probably due to the adiabatic 166 expansion of the CO<sub>2</sub>. This likely occurred because of the pressure decrease due to the 167 leakage of SC-CO<sub>2</sub> into the induced cracks, which accompanied the phase change from 168 169 the SC state to the gaseous state.

There were three terms missing from the AE data, as shown by the thick solid line segments in Figure 4b. The first missing term from 1 to 26 s was due to a problem in the acquisition of AE data with an excess of induced AE events. The second and third terms, from 4 min 38 s to 6 min 12 s and from 20 min 31 s to 21 min 44 s, respectively, were due to replacing a file filled with AE data with a new file in the monitoring unit. The decrease in the located AE event rate of the second and third terms was caused by the missing data. 177

#### 178 **3.2 AE Source Locations and Crack Observations**

Figure 5 shows the locations of the AE hypocenters projected onto the horizontal 179 plane, XY, and the two vertical planes, YZ, and ZX. Figure 6 shows the AE hypocenter 180 181 distributions on the horizontal plane, XY, for the period from 0 (BD) to 1 min 15 s in 182 comparison with those from 1 min 15 s to 30 min 0 s. Figure 6 indicates AE hypocenters distributed along A-direction from the HF hole to the AE1 and AE3 holes 183 184 for the initial 1 min 15 s from BD. After that, they started to distribute along B-direction almost perpendicular to A-direction from the portion close to AE3 hole. From the 185 vertical distributions on the YZ and ZX planes in Figure 5, the AE hypocenters distribute 186 187 on the two nearly vertical planes having the strikes of A- and B-directions.

After we analyzed and discussed the AE source distributions, <sup>11,12</sup> we have newly made close observations of a core overcored around the pressurizing section and images obtained on the wall of the HF hole by a borehole camera, to clarify the cracks induced by the HF.

192 Figure 7 shows a photo of a core of the 36 mm diameter pilot hole drilled to make the pressurizing section before HF and a hollow core recovered by the 86 mm diameter 193 overcoring around the pressurizing section after HF. We could see a crack almost 194 195 parallel to the drilling direction, as indicated with a red line on the surface of the core recovered by overcoring. The crack direction was confirmed to face the AE1 hole from 196 197 the HF hole. Also, on the opposite side of the core, there was a similar crack facing the AE3 hole along the drilling direction. These cracks indicated that the HF induced bi-198 wing vertical cracks in A-direction. Because we confirmed that there was no pre-199 existing crack in the pressurizing section by inspecting the pilot hole core before HF 200 and the high BD pressure (15.62 MPa) was recorded, it was most likely that the cracks 201 202 were newly induced in the intact rock at BD, and they extended for 1 min 15 s from BD along the vertical plane having the strike of A-direction. The direction corresponded to 203 204 the observation that the injected CO<sub>2</sub> gushed out from the AE1 and AE3 holes.

Figure 8 shows cracks observed on the wall of the HF hole by a borehole camera 205 after HF. From this, we can confirm that two parallel cracks were induced in A-direction 206 in the interval of the pressurizing section, and they extended 3.33 m in length from a depth 207 of 6.15 m to 9.48 m, which resulted in an ellipsoidal crack. The vertical length of the 208 crack extension almost corresponds to the AE hypocenter distribution in Z-direction, as 209 shown in Figure 5. On the other hand, the AE hypocenters horizontally distributed along 210 B-direction were almost perpendicular to A-direction from the position of approximately 211 212 0.6 m away from the HF hole, although no pressure acted in the pressurizing section for

the term from 1 min 15 s to 30 min 0 s, as shown in Figure 4a. From this, we could infer that a vertical crack existed in B-direction before HF, and the AE events were induced with the intrusion of gaseous state  $CO_2$  into it.

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# 217 **3.3 Focal Mechanism from P Wave First-Motion Polarities of AE**

We examined the focal mechanisms by using P wave first-motion polarities of the 218 AE. The polarity response of the sensors was checked by dropping a steel ball onto the 219 220 surface of each AE sensor. We confirmed for each sensor that an upward trace corresponded to a compressive wave, and a downward trace corresponded to a dilatant 221 wave. Figure 9 shows the percentage R(%) of a number of the compression polarities to 222 a total number of the polarities we could read for each AE event along with the elapsed 223 time<sup>12</sup>. This analysis was conducted for AE whose polarities could be read at six or 224 more sensors out of the well-located AE events shown in Figures 5 and 6. The number 225 226 of AE that satisfied the condition was 161 out of the 1249 well-located hypocenters.

The ratio R approaches 100% for a tensile or explosion source, 50% for a pure shear 227 source, and 0% for a compression or implosion source when good coverage of AE 228 sensors around a source is achieved. In the present study, we labelled AE with 229  $100 \ge R \ge 80$  as tensile-dominated (type T), those with 80 > R > 20 as shear-dominant 230 (type S), and those with  $20 \ge R \ge 0$  as compression-dominant (type C). In Figure 9, 231 there are five events labelled as type T within 30 s after BD, although we can only see 232 three symbols in the figure due to overlapping. However, no event labelled as type T 233 234 was recorded after this time. Immediately after type T events were recorded, many type S events started to be recorded with type C events. They were recorded even after 235 injection was stopped and the injection pressure completely declined as shown in Figure 236 237 4a.

In addition to the temporal change of the ratio  $R^{12}$ , we newly show Figures 10 and 238 239 11 to examine relation between location of a source and its ratio R. Figure 10 plots the X, Y, and Z coordinates of the AE hypocenters along with the elapsed times for 30 min 240 after BD, which corresponds to those in Figure. 9. Figure 11 plots their distributions on 241 the horizontal plane, XY, for the period from 0 (BD) to 1 min 15 s in comparison with 242 those from 1 min 15 s to 30 min 0 s. The plotted AE events correspond to those shown 243 in Figures 9 and 10; in other words, there are only 161 events satisfying the condition, 244 out of the 1249 well-located AE hypocenters plotted in Figures 5 and 6. Figures 10 and 245 11 elucidate that four out of the five type T events occurred near the pressurizing 246 section of the HF hole immediately after BD. Following that, immediately after BD in 247

- Figure 10, the hypocenters of the type S and the C events cluster in the range of -0.5 m
- to 0.1 m in both the X and Y coordinates, corresponding to the distribution along A-
- 250 plane in Figure 11a. The type S and type C events are not only located along A-plane
- but also B-plane after 1 min 15 s, as shown in Figure 11b.
- 252

### **4 Discussion**

#### 4.1 Crack Extension with Injection of SC-CO<sub>2</sub>

We injected SC-CO<sub>2</sub> into the pressurizing section after confirming there were no pre-255 existing cracks in it by inspecting the recovered core. However, a 3.33 m long bi-wing 256 crack that extended vertically along the boring axis was observed on images of the HF 257 hole wall obtained by a borehole camera after HF. The horizontal direction of the crack 258 observed on the outer surface of the hollow core recovered by overcoring after HF was 259 consistent with the direction of the AE1 and AE3 holes where the injected CO2 gushed 260 261 out. The crack was most likely induced at BD, from the following reasons; (1) the BD pressure (15.62 MPa) was high, (2) the bi-wing crack extended in the vertical, and (3) 262 horizontal directions of the crack extension correspond to the AE hypocenter 263 distributions just after BD. 264

In a similar HF field experiment, water was injected into the same size pressurizing 265 section of granite (36 mm in diameter and 160 mm long) in Mizunami Underground 266 Research Laboratory (MIU). New cracks extended 0.4 m vertically and 0.3 m 267 horizontally under a flow rate of 10 mL/min, and they extended to 0.7 m and 0.5 m, 268 respectively, after the flow rate was increased to 30 mL/min.<sup>14</sup> In the present 269 experiment, SC-CO<sub>2</sub> was injected at a flow rate of 50 mL/min. Even considering that 270 the flow rate was greater than that of water, the crack extension of 3.33 m vertical and 2 271 m horizontal was still notably larger than that of water. The difference could be 272 attributed to the effects of expanding gaseous pressure with the phase change of the 273 CO<sub>2</sub> from SC to gas and the lower viscosity of gaseous state CO<sub>2</sub> than that of liquid 274 water. If so, SC-CO<sub>2</sub> can likely make a larger crack than water even under the same 275 276 flow rate, when both CO<sub>2</sub> and water are measured in a volume of the liquid state in the 277 cylinder of the syringe pump.

After 1 min 15 s from BD, AE hypocenters started to distribute along B-direction almost perpendicular to A-direction from the position of a 0.6 m distance from the HF hole. Because no pressure acted in the pressurizing section of the HF hole in the term, a pre-existing crack most likely existed in the position along B-direction. If so, this indicated that CO<sub>2</sub> could easily intrude and enhance AE occurrence along a pre-existing crack without pressure.

#### 284 **4.2 Fracturing Mechanism of Type T Events**

The type T events occurred near the pressurizing section of the HF hole immediately after BD, and this was consistent with the elastic theory explaining that tensile fracture initiates HF. <sup>15-18</sup> This finding was noteworthy because many researchers have reported that shear events are dominant rather than tensile at BD,<sup>19-22</sup> although some researchers have recently reported existence of tensile dominant events.<sup>23-26</sup> Sasaki et al.<sup>27</sup> performed a laboratory HF experiment in a block of impermeable acrylic resin and reported that all AE events (10 in total) were tensile without exception.

A granitic rock generally has two sets of preferred orientation of microcracks<sup>28, 29</sup>. 292 Most of microcracks preferentially oriented along the "rift plane", and a secondary 293 294 orientation is known as the "grain plane." The "hardway plane" is defined by the plane perpendicular to both the rift plane and the grain plane. The three planes were named by 295 quarrymen, and they split a granitic rock along the rift plane to sprit it easily. Also, in 296 HF experiments, there is a tendency that crack easily extends along the rift plane $^{30}$ . 297 Recently, using this nature of a granitic rock, Yamamoto et al.<sup>31</sup> conducted HF 298 experiments using samples of two types of granite under uniaxial loading, namely those 299 300 with a rift plane perpendicular to the expected direction of macroscopic fracture propagation and those with a rift plane parallel to it (i.e., along the loading axis). They 301 found tensile events to be dominant when the fracture propagated perpendicular to the 302 rift plane, whereas shear events were dominant when the fracture propagated parallel to 303 the rift plane. These laboratory experiments suggest that tensile events are dominant 304 when a fracture propagates in intact rock. In addition, Rodriguez et al.<sup>32</sup> showed that 305 tensile events are concentrated near the edges of the propagating fracture before BD. 306 Also in our similar field experiment using water in the MIU,<sup>14</sup> type T events were 307 recorded immediately after BD; furthermore, most of the type T events were distributed 308 on the borders of regions where AE events had already occurred. These results suggest 309 310 that new crack propagation in intact rock is induced by tensile fracturing.

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#### 312 4.3 Fracturing Mechanism of Type S Events

After type T events were recorded, many type S events were recorded with type C events. As mentioned above, many researchers have reported that most of the AE recorded with HF are shear events. Also, in volcanic earthquake swarms, significant parts of seismic events show a shear mechanism, although many events are characterized by magma intrusions or eruptions. To explain this observation, Hill<sup>33</sup> proposed the conceptual model that magma intrudes into the weak planes lying along the direction of the maximum compressive stress as well as among many weak planes prevailing in a volcanic region. The magma intrusion, forming a dike, would accompany some tensile fracturing, whereas shear fracture would form conjugate faults, thereby connecting the tips of dikes.

323 Even in a laboratory three-bending test, shear events were observed to dominate. Kao et al.<sup>34</sup> conducted a three-point-bend fracture test on a granite specimen measuring 217 324  $mm \times 73 mm \times 32 mm$  (span  $\times$  height  $\times$  thickness) with a 4 mm notch; the AE events 325 326 were located, and their focal mechanisms were analyzed. They found that all the AE events were shear dominant due to tortuosity reflecting the local deviation of the crack 327 path by grain-scale heterogeneity, although the macroscopic fractures were tensile. This 328 329 experiment suggests that macroscopic tensile fracture is a natural consequence of shear events associated with tortuosity, and that the local mechanism, i.e. the mechanism of an 330 AE event, does not necessarily reveal the nature of the macro failure mode. Microscopic 331 observations in our laboratory HF experiments using SC-CO<sub>2</sub> with very low viscosity 332 revealed that HF cracks propagated mainly along the grain boundaries of the constituent 333 334 minerals, producing many small cracks inclined in the direction of the maximum compressive stress,  $\sigma_1$ , which was the propagating direction of the main crack. Because 335 shear stress acted on a plane inclined in the direction of  $\sigma_1$ , a shear fracture could easily 336 occur on the plane.<sup>10</sup> 337

In the field experiment presented here, when cracks extended from intact rock 338 around the pressurizing section of the HF hole into inhomogeneous rock that contained 339 various weak discontinuities including pre-existing cracks, many type S events were 340 most likely induced with new crack extension or slippage on crack planes inclined to 341 the direction of the macroscopic crack propagation. If a concept, such as this, regarding 342 crack propagation and the origin of AE was accepted, we could understand the reason 343 why many researchers have reported that shear events are dominant with HF in an 344 actual field operation because rock masses in the field have various geological weak 345 discontinuities. In addition to the mechanism, it is likely that pore pressure increased 346 with HF and its increase also enhanced shear events. 347

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### 349 4.4 Fracturing Mechanism of Type C Events

As shown in Figures 9 through 11, many type C events were recorded with type S events. However, we never observed type C events in our similar HF experiment using water in the MIU.<sup>14</sup> Thus, type C events are probably a distinctive feature associated with CO<sub>2</sub> injection.

On the other hand, compressive events are often observed in seismicity in volcanic 354 regions. Shimizu et al.<sup>35</sup> and Foulger et al.<sup>36</sup> suggested that these events were induced 355 by crack closure due to degassing from magma. In addition, Ross et al.<sup>23</sup> expected that 356 compressive events should be induced with the removal of large volumes of steam 357 from a geothermal reservoir, and Martínez-Garzón et al.<sup>37</sup> actually observed 358 compressive events at The Geysers geothermal field, California, USA. Bohnhoff and 359 Zoback<sup>38</sup> observed non-shear-mechanism events with the leakage of injected CO<sub>2</sub> at a 360 depth of 900 m underground. Because the depth was close to that of the hydrostatic 361 pressure at which CO<sub>2</sub> changes from SC to the gaseous state, they reasoned that mass 362 advection of CO<sub>2</sub> caused the non-shear-mechanism events. In a laboratory experiment 363 for hydraulics, Warjito et al.<sup>39</sup> showed that a bubble breaking into smaller bubbles 364 induced acoustic waves, and Kolaini<sup>40</sup> pointed out that the acoustic radiation released 365 by bubble breakage changed with the concentration of salt. Thus, the type C events 366 observed in our present experiment were likely induced by crack closure due to the 367 degassing of injected CO<sub>2</sub>, mass advection, or the bursting of bubbles. Although CO<sub>2</sub> 368 injection could induce many type C and S events, at least the type C events out of them 369 unlikely trigger natural earthquakes because the type C events never create new cracks. 370

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### 372 **5 Conclusions**

We conducted a HF experiment using SC-CO<sub>2</sub> in a hole drilled into a granitic hot rock at a depth of 7.24 m to 7.40 m below the tunnel floor. During the experiment, we monitored the AE, pressure, and temperature of injected CO<sub>2</sub>. After the experiment, we observed a crack induced by HF on the surface of the core recovered by overcoring as well as on images of the HF hole wall obtained by a borehole camera. From the hypocenter location and the focal mechanism of the monitored AE and the crack observations, we concluded the followings:

1. We found a bi-wing vertical crack 3.33 m long along an HF hole on images of the HF 380 hole wall obtained after HF by a borehole camera. The horizontal direction of the 381 crack observed on the surface of the core (recovered by overcoring after HF) was 382 consistent with the directions from the HF hole to the AE1 and AE3 holes where the 383 injected CO<sub>2</sub> gushed out. Because the BD pressure of 15.62 MPa was sufficiently 384 high and the crack extension in the vertical and horizontal directions corresponded to 385 the AE hypocenter distributions just after BD, the crack was most likely induced at 386 BD. 387

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2. In our similar HF field experiment, we injected water into the same size pressurizing
section of granite (36mm in diameter and 160 mm in length) in the MIU, and new
cracks extended 0.4 m to 0.7 m vertically and 0.3 m to 0.5 m horizontally. Even
considering that the injection flow rate of SC-CO<sub>2</sub> in this experiment, 50 mL/min,
was nearly two times greater than that of water in the MIU, 30 mL/min, the crack
extension of 3.33 m vertically and 2 m horizontally was still quite greater than that of
water. This difference could be attributed to the effect of expanding gaseous pressure

with the phase change of  $CO_2$  from SC to a gas, as well as lower viscosity of the CO<sub>2</sub> gas than that of liquid water.

397 3. After 1 min 15 s from BD, AE hypocenters started to distribute along the direction
almost perpendicular to that of the initial crack induced by HF from the position of
0.6 m from the HF hole. Because no pressure acted in the pressurizing section of the
HF hole in the term, a pre-existing crack most likely existed in a position along that
direction. If so, this indicates that CO<sub>2</sub> can easily intrude and enhance AE occurrence
along a pre-existing crack without pressure.

- 4. From the examination of the focal mechanisms by using P wave first-motion
  polarities of the AE events, we found that type T (tensile-dominated) events occurred
  near the pressurizing section of the HF hole immediately after BD. Findings from
  laboratory experiments by many researchers, as well as our similar recently
  conducted field experiment, suggest that new crack propagation in intact rock is most
  likely induced by tensile fracturing. The fact that type T events were induced
  immediately after BD is consistent with the results from previous experiments and
- the conventional elastic theory, which state that tensile fracture initiates HF.
- 411 5. After type T events were recorded, many type S (shear-dominant) events were
- 412 recorded with type C (compression-dominant) events. When cracks extended from
- intact rock around the pressurizing section of the HF hole into inhomogeneous rock
- 414 having various weak discontinuities including pre-existing cracks, many type S
- events were likely induced with new crack extension or slippage on planes of pre-
- 416 existing cracks. In addition to the mechanism, it is likely that pore pressure increased
- 417 with HF and its increase also enhanced shear events.
- 6. Many type C events were recorded with type S events in this experiment, although
- 419 type C events were never observed in our similar HF experiment using water in the
- 420 MIU. Thus, type C events are probably a distinct feature associated with CO<sub>2</sub>
- 421 injection. Compressive events have been observed in volcanic regions and a
- 422 geothermal reservoir, and they were attributed to crack closure due to degassing from
- 423 magma in volcanic regions and the removal of steam in a geothermal reservoir. Thus,

- 424
- the type C events observed in our present experiment were most likely induced by
- 425 crack closure due to the degassing of injected  $CO_2$ . Even if  $CO_2$  injection induced
- 426 many AE events, compressive events (like the type C event) out of them unlikely
- 427 trigger natural earthquakes because they never created new cracks.
- 428

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435

# 436 **Declaration of interests**

The authors declare that they have no known competing financial interests or
personal relationships that could have appeared to influence the work reported in this
paper.

440

# 441 **References**

1. Xie H, Li X, Fang Z, Wang Y, Li Q, Shi L, Bai B, Wei N, Hou Z. Carbon geological
utilization and storage in China: current status and perspectives. Acta Geotechnica.
2014; 9: 7–27.

- 445 2. Brown DW. A hot dry rock geothermal energy concept utilizing supercritical CO<sub>2</sub>
- instead of water, paper presented at 25th Workshop on Geothermal Reservoir
  Engineering. 2000; Stanford Univ., Stanford, Calif.
- 448 3. Pruess K. Enhanced geothermal systems (EGS) using CO<sub>2</sub> as working fluid—A
- 449 novel approach for generating renewable energy with simultaneous sequestration of
  450 carbon, Geothermics. 2006; 35: 351–367.
- 451 4. Kalantari-Dahaghi A. Numerical simulation and modeling of enhanced gas recovery
- 452 and  $CO_2$  sequestration in shale gas reservoirs: A feasibility study, paper presented at
- International Conference on CO<sub>2</sub> Capture, Storage, and Utilization. 2010; Soc. of Pet.
  Eng., New Orleans, La.
- 454 Elig., New Offenis, La.
- 5. Liu F, Ellett K, Xiao Y, Rupp JA. Assessing the feasibility of CO<sub>2</sub> storage in the New
- Albany Shale (Devonian- Mississippian) with potential enhanced gas recovery using
   reservoir simulation, Int J Greenh. Gas Control. 2013; 17: 111–126.
- 458 6. Middleton RS, Carey JW, Currier R.P, Hyman JD, Kang Q, Karra S, Jiménez-
- 459 Martínez J, Porter ML, Viswanathan HS. Shale gas and non-aqueous fracturing

fluids: Opportunities and challenges for supercritical CO<sub>2</sub>, Appl Energy. 2015; 147:
500–509.

462 7. Godec M, Koperna G, Petrusak R, Oudinot A. Potential for enhanced gas recovery

and CO<sub>2</sub> storage in the Marcellus shale in the Eastern United States, Int J Coal Geol.
2013; 118: 95–104.

- 8. Nuttall BC, Drahovzal JA, Eble CF, Bustin RM. CO<sub>2</sub> Sequestration in Gas Shales of
  Kentucky, poster no. 106 at the 5th Annual Conference on Carbon Capture and
  Sequestration. 2006; Alexandria, VA, USA.
- 9. Ishida T, Aoyagi K, Niwa T, Chen Y, Murata S, Chen Q, Y. Nakayama Y. Acoustic
  emission monitoring of hydraulic fracturing laboratory experiment with supercritical
  and liquid CO<sub>2</sub>. Gephys Res Lett 2012; 39: L16309.

10. Ishida T, Chen Y, Bennour Z, Yamashita H, Inui S, Nagaya Y, Naoi M, Chen Q,

472 Nakayama Y, Nagano Y. Features of CO<sub>2</sub> fracturing deduced from acoustic

473 emission and microscopy in laboratory experiments, J Geophys Res Solid Earth.

474 2016; 121(11): 8080–8098. doi:10.1002/2016JB013365.

- I1. Ishida T, Desaki S, Yamashita H, Inui S, Naoi M, Fujii H, Katayama T. Injection of
  supercritical carbon dioxide into granitic rock and its acoustic emission monitoring,
  Procedia Engineering. 2017; 191: 476-482. (Proc. of Eurock 2017, Paper No. 106,
  Ottawa Grash Barahlia 2017.) doi: 10.1016/j.marana.2017.05.206
- 478 Ostrava, Czech Republic, 2017.) doi: 10.1016/j.proeng.2017.05.206
- 12. Ishida T, Desaki S, Kishimoto Y, Naoi M, Fujii H. Implosive acoustic emissions
  induced by injection of supercritical carbon dioxide into a hot granitic rock mass,
- 481 Geomechanics and Geodynamics of Rock Masses. 2018; 1369–1374, Taylor &
- 482 Francis Group, London. (Proc. of the International European Rock Mechanics
- 483 Symposium, EUROCK 2018, Saint Petersburg, Russia, 22-26 May 2018), ISBN
   484 978-1-138-61645-5)
- 13. Ishida T, Sasaki S. Numerical simulation to examine accuracy of AE source location
  and its applications to in-situ rock monitoring, J Acoust Emission. 2011; 29: 260–
  272.
- 14. Ishida T, Fujito W, Yamashita H, Naoi M, Fuji H, Suzuki K, Matsui H. Crack
  expansion and fracturing mode of hydraulic refracturing from acoustic emission
  monitoring in a small-scale field experiment, Rock Mech Rock Eng. 2019; 52(2):
  543-553. doi: 10.1007/s00603-018-1697-5
- 492 15. Hubbert MK, Willis DG. Mechanics of hydraulic fracturing, Trans Am Inst Min
  493 Metall Petrol Engrs. 1957; 210: 153-168.
- 494 16. Zoback MD, Rummel F, Jung R, Raleigh CB. Laboratory hydraulic fracturing
  495 experiments in intact and pre-fractured rock, Int J Rock Mech Min Sci. & Geomech

- 496 Abstr. 1977; 14: 49-58.
- 497 17. Haimson BC. The hydrofracturing stress measuring method and recent field results,
  498 Int J Rock Mech Min Sci & Geomech Abstr. 1978; 15: 167-178.
- 499 18. Schmitt DR, Zoback MD. Infiltration effects in the tensile rupture of thin walled
- cylinders of glass and granite: Implications for the hydraulic fracturing breakdown
  equation, Int J Rock Mech Min Sci & Geomech Abstr. 1993; 30: 289-303.
- 502 19. Talebi S, Cornet FH. Analysis of the microseismicity induced by a fluid injection in
- a granitic rock mass, Geophys Res Lett. 1987; 14: 227–230.
- 20. Cornet FH. Fracture processes induced by forced fluid percolation, Volcanic
  Seismology, IAVCEI Proceedings in Volcanology, vol 3, edited by Gasparini P,
  Scarpa R, Aki K, Springer Verlag, 1992; 407–431.
- 507 21. Horálek, J., Jechumtálová Z, Dorbath L, Šílený J. Source mechanisms of micro-
- <sup>508</sup> earthquakes induced in a fluid injection experiment at the HDR site Soultz-sous-
- 509 Forêts (Alsace) in 2003 and their temporal and spatial variations, Geophys J Int.
- 510 2010; 181: 1547-1565.
- 511 22. Maxwell SC, Cipolla C. What does microseismicity tell us about hydraulic
- fracturing? SPE Annual Technical Conference and Exhibition, Denver, Colorado,USA, 2011; SPE 146932.
- 23. Ross A, Foulger GR, Julian BR. Non-double-couple mechanisms at The Geysers
  geothermal area, California, Geophys Res Lett. 1996; 23: 877–880.
- 516 24. Vavryčuk V. Non-double-couple earthquakes of 1997 January in West Bohemia,
- 517 Czech Republic: evidence of tensile faulting, Geophys J Int. 2002; 149: 364-373.
- 518 25. Šílený J, Hill DP, Eisner L, Cornet FH. Non-double-couple mechanisms of
- microearthquakes induced by hydraulic fracturing, J Geophys Res. 2009; 114:
  B08307. doi:10.1029/2008JB005987.
- 521 26. Julian BR, Foulger GR, Monastero FC, Bjornstad S. Imaging hydraulic fracturing in
  522 a geothermal reservoir, Geophys Res Lett. 2010; 37: L07305.
  523 doi:10.1020/2000CL.040022
- 523 doi:10.1029/2009GL040933.
- 524 27. Sasaki S, Matsunaga I, Kobayashi H, Ishida T. Development of a fracture evaluation
   525 technique for HDR geothermal energy extraction --- Mechanism of hydraulic
- 526 fracturing inferred from acoustic emission ---. Central Research Institute of Electric
- 527 Power Industry, Abiko Research Laboratory Report, 1988; U88034 (in Japanese
- 528 with English abstract and figure captions)
- 529 28. Dale TN. The commercial granites of New England(I), U.S. Geological Survey
  530 Bull. 1923; 738: 1-97.
- 531 29. Sano S, Kudo Y, Mizuta Y. Experimental determination of elastic constants of

- Oshima granite, Barre granite, and Chelmsford granite, J Geophys Res. 1992;
  97(B3): 3367-3379.
- 30. Ishida T. Acoustic emission monitoring of hydraulic fracturing in laboratory and
  field, Construction and Building Materials. 2001; 15: 283–295. doi:
- 536 10.1016/S0950-0618(00)00077-5.
- 537 31. Yamamoto K, Naoi M, Chen Y, Nishihara K, Yano S, Kawakata H, Akai T,
- 538 Kurosawa I, Ishida T. Moment tensor analysis of acoustic emissions induced by
- laboratory-based hydraulic fracturing in granite, Geophys J Int. 2019; 216(3): 1507–
- 540 1516. doi: 10.1093/gji/ggy493.
- 32. Rodriguez IV, Stanchits S, Burghardt J. Data-driven, in situ, relative sensor
  calibration based on waveform fitting moment tensor inversion. Rock Mech Rock
  Eng. 2017; 50: 891–911. doi: 10.1007/s00603-016-1144-4
- 33. Hill DP. A model for earthquake swarms, J Geophys Res. 1977; 82: 1347-1352
- 34. Kao CS, Carvalho FCS, Labuz JF. Micromechanisms of fracture from acoustic
  emission, Int J Rock Mech Min Sci. 2011; 48: 666–673.
- 547 35. Shimizu H, Ueki S, Koyama J. A tensile-shear crack model for the mechanism of
  548 volcanic earthquakes, Tectonophysics. 1987; 144, 287-300.
- 549 36. Foulger GR, Long RE, Einarsson P, Bjornsson A. Implosive earthquakes at the
  550 active accretionary plate boundary in northern Ice land, Nature. 1989; 337: 640-642,
  551 doi:10.1038/337640a0.
- 37. Martínez-Garzón P, Kwiatek G, Bohnhoff M, Dresen G. Volumetric components in
   the earthquake source related to fluid injection and stress state, Gephys Res Lett.
- 554 2017; 44: 800-809. doi:10.1002/2016GL071963.
- 38. Bonhoff M, Zoback MD. Oscillation of fluid-filled cracks triggered by degassing of
   CO<sub>2</sub> due to leakage along wellbores, J Geophys Res. 2010; 115: B11305,
- 557 doi:10.1029/2010JB000848.
- 39. Warjito, Mochizuki O, Ishikawa H. Breakup of a single babble and its sound,
  Nagare. 2002; 21: 165-172 (in Japanese with English abstract)
- 40. Kolaini AR. Effects of salt on babble acoustic radiation in water, Journal of the
- 561 Acoustical Society of America. 1999; 105(4): 2181-2186.
- 562

# 563 **Figure Captions**

Figure 1 Site and experimental setup. (a) Layout of test site. <sup>11, 12</sup> (b) Arrangement of
acoustic emission (AE) sensors surrounding a pressurized section under the test site.
The inset shows a dimensioned diagram of packer used to seal the pressurized
section.

568 Figure 2 Injection system for SC-CO<sub>2</sub>.<sup>11, 12</sup>

569 Figure 3 Phase diagram of  $CO_2$ .<sup>11, 12</sup>

570 Figure 4 Temporal changes of fluid pressure, temperature, AE rate, and flow rate.

- 571 Thick line segments on the fine line in Figure 4b indicate terms missing AE data.<sup>11, 12</sup>
- 572 Figure 5 Projections of AE hypocenters onto horizontal plane, *XY*, and two vertical
- 573 planes, *YZ*, and *ZX*, where *X* axis is parallel to the direction from the hole AE1 to
- AE2 and that from AE4 to AE3, *Y* axis is perpendicular to *X* axis and *Z* axis is the
- 575 vertical. The origin is placed at the center of HF hole on the horizontal plane, *XY*.
- 576 The bars and the open rectangles on the *YZ* and *ZX* planes correspond to HF and AE
- holes and AE sensors, respectively. The broken lines on them indicate pressurizing
  section from 7.24 to 7.40 m. Note that +X direction is from right to left.<sup>11, 12</sup>
- 579 Figure 6 AE hypocenter distributions on horizontal XY plane, for period from 0 to 1

580 min 15 s in comparison with those from 1 min 15 s to 30 min 0 s.<sup>11, 12</sup>

581 Figure 7 Photo of a core of 36 mm diameter pilot hole drilled to make pressurize

- section before HF and a hollow core recovered by 86 mm diameter overcoringaround pressurizing section after HF.
- Figure 8 Cracks observed on wall of HF hole with a borehole camera after HF. CO<sub>2</sub>
  injected in section from 7.24 to 7.40 m most likely induced cracks traced with the
  black lines. Yellow thick lines indicate open cracks of aperture within 0.5 mm,
  whereas yellow broken lines indicate closed adherent cracks.
- 588 Figure 9 Compressive ratio *R* (%) of P wave first-motion polarity of AE. In the

present study, we labelled AE with  $100 \ge R \ge 80$ , 80 > R > 20 and  $20 \ge R \ge 0$  as

tensile-dominant (type T: red stars), shear-dominant (type S: blue circles) and

<sup>591</sup> compression-dominant (type C: green circles), respectively.<sup>12</sup>

592 Figure 10 X, Y, and Z coordinates of AE hypocenters along with elapsed times for 30

- 593 min after BD, corresponding to those of Figure 9. Red stars, open blue circles and
- closed green circles indicate the same AE types as those in Figure 9. A band of
- broken lines on the Z coordinate indicates the span of pressurizing section in the pilothole.
- 597 Figure 11 Distributions of AE hypocenters projected on horizontal plane, XY. (a)
- 598 From 0 s to 1 min 15 s. (b) From 1 min 15 s to 30 min 0 s. Figures 11a and 11b plot
- only 161 AE in total out of the 1249 well-located AE hypocenters plotted in Figures
- 600 6 and 7. The 161 AE correspond to those shown in Figures 9 and 10.



























