

1 **Tracking CO₂ plume footprints in reservoir-caprock system**
2 **by fibre optic strain sensing**

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13 **Underground fluid storage and utilization, e.g., geological storage of carbon**
14 **dioxide (CO₂) and natural gas, require effective tools to monitor the vertical**
15 **pressure, deformation, and saturation migrations, and verify the secure**
16 **containment of the reservoir-caprock system. Here we demonstrate how to utilize**
17 **the swelling strain signals attributed to the footprints of pressure build up and the**
18 **adsorption of supercritical CO₂ in natural clay-rich rocks to track the**
19 **displacement of supercritical CO₂/brine under typical conditions of CO₂ geological**
20 **storage. Our study effectively captured the breakthrough of CO₂ plume from the**
21 **analogue “reservoir” part to the “caprock” part using the distributed strain,**
22 **measured by a high-resolution single-mode Rayleigh-scattering based optical fibre**
23 **sensor. The magnitude of strain change induced by CO₂ adsorptions on the clay**
24 **minerals is found to be significantly greater than that caused by pore pressure**

25 **changes alone. The findings suggest that the measured strain changes can not only**
26 **reveal the in situ deformation state but also be a valid indicator for tracking the**
27 **movement of CO₂ plume as it enters the clay-rich critical region in reservoir-**
28 **caprock systems, and thus serves as an early warning to the potential occurrence**
29 **of large deformations caused by high pressure build up.**

30 Underground geological reservoir engineering, e.g., enhanced oil recovery by water or
31 gas injections, geothermal exploitations, shale gas fracking, and natural gas and CO₂
32 storages, often involve fluid injections and/or extractions. During these operations, the
33 in situ state of pressure in the porous space would be inevitably changed, leading to the
34 mechanical deformations in geological formations due to the poroelastic mechanism—
35 the coupling between the deformation of porous matrix and pore fluid flow under stress.
36 The deformations may be significant to cause induced seismicity¹ and observable uplift
37 or subsidence of earth ground². CO₂ sequestration into underground geological
38 reservoirs is now considered as one of the best near-term solutions to mitigate global
39 warming effects³⁻⁵. In a large-scale geological CO₂ storage project, another potential
40 problem is associated to the impact of pressure build up on the long-term integrity of a
41 CO₂ repository⁶ and the risk of CO₂ or brine leakages⁷. CO₂ could migrate into the
42 caprock if the pressure in the reservoir overwhelms the threshold pressure of caprock. In
43 addition, the reactivation of faults and breach of caprock could also create or enhance
44 vertical leakage pathways, contributing to an unexpected leakage of CO₂^{8,9}. For these
45 reasons, valid methods are required to closely monitor the development of pressure
46 build up, deformations and CO₂ plume migration in reservoir formations and caprocks
47 above the storage reservoir.

48 Besides poroelastic changes, one of the poromechanical responses, i.e., clay swelling
49 phenomena has been well studied from different aspects of geoscience, e.g., in shales,
50 coals, soils, etc. CO₂ can be energetically adsorbed on the surface and edge of clay

51 minerals or into the crystal interlayers, resulting in significant swelling¹⁰⁻¹². The swelling
52 can be also significant to change the stress state¹³. At a given pressure, the swelling is
53 more pronounced with carbon dioxide than with other fluids, like water and methane¹².
54 Therefore, it might be anticipated that, as CO₂ displaces brine through clay-rich strata
55 (e.g., caprocks), strain changes due to the preferential adsorption of CO₂ in the clay, in
56 addition to the poroelastic mechanism, could be observed. Taking advantage of this
57 characteristic, we propose to monitor the migration of CO₂/brine displacement front in
58 clay-rich region in a reservoir-caprock via monitoring the strain (Fig. 1 a).

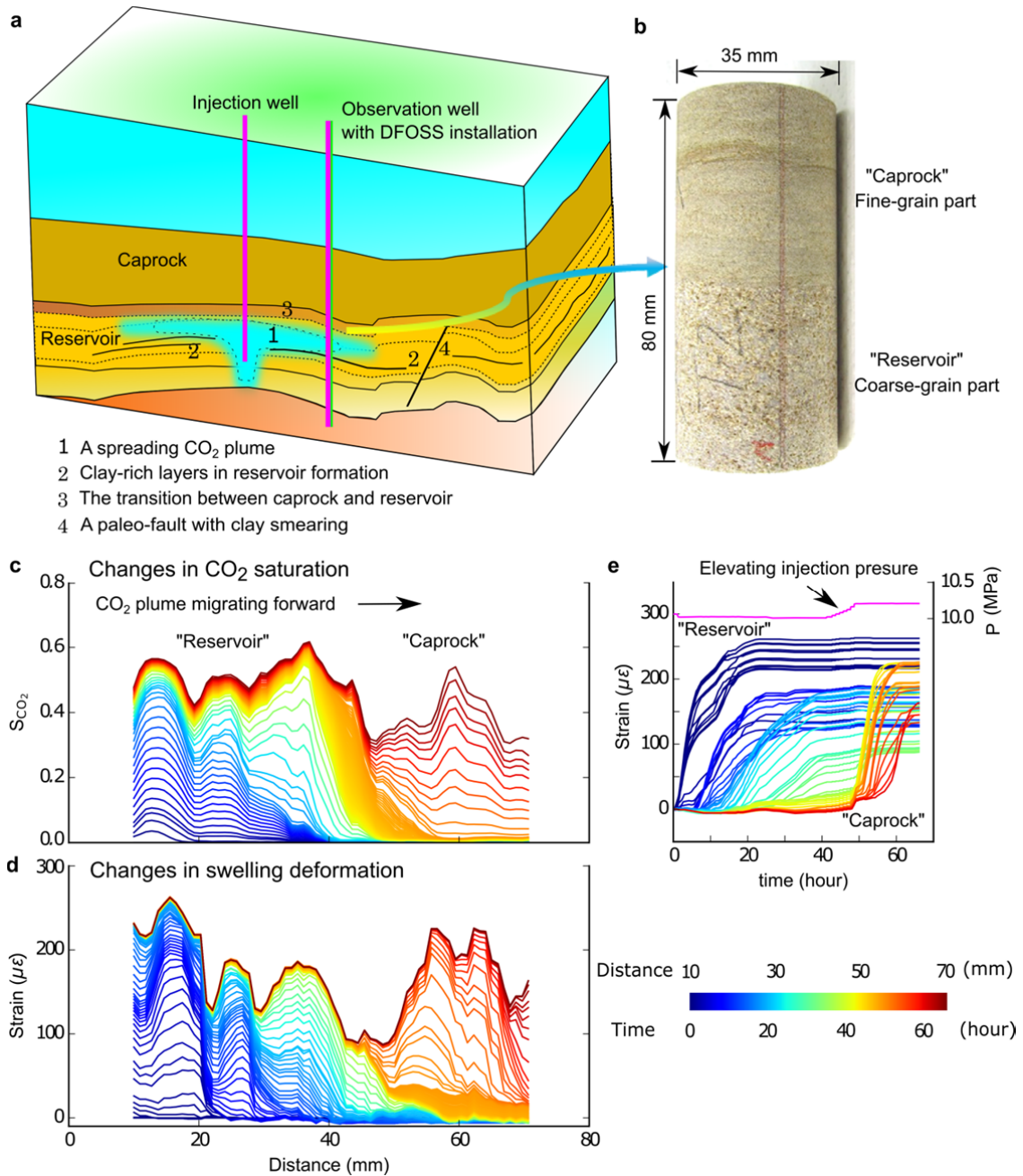
59 With the recent development of distributed fibre optical strain sensing (DFOSS)
60 technique¹⁴, the response at each spatial location due to rock deformation may be
61 tracked in real-time. Therefore, the tool can potentially give an improved understanding
62 of the roles of pressure migration and CO₂ plume migration along the vertical direction
63 while incorporating the concept of in-reservoir and above-zone pressure monitoring¹⁵
64 (e.g., at Decatur CO₂ storage site¹⁶), and also give important constraints in
65 geomechanical modelling^{7,17}.

66 This study aims to measure rock swelling strain caused by CO₂ displacing brine through
67 an analogue reservoir-caprock system. A clay-rich Tako sandstone (from Gunma,
68 Japan) developed with both coarse-grain high-permeability (“reservoir”) and fine-grain
69 low-permeability (“caprock”) regions is used for the experiment (Fig. 1 b). The strain is
70 continuously measured using a high-resolution DFOSS tool (see Methods) during CO₂
71 drainage and brine imbibition in the sample under a typical reservoir condition (10 MPa
72 and 40 °C). In addition, the X-ray CT is used to image the spatial fluid saturation in the
73 rock.

74 **Experimental tests**

75 The experiment was started from a dry state of the rock sample after two days of
76 vacuum-drying. Then the sample was pressurized step-by-step by increasing the
77 confining pressure (0~15 MPa). After this the confining pressure was reduced to 5 MPa
78 and then the brine (potassium iodide solution, 11.5 wt%) was injected to the vacuumed
79 sample until the sample was fully saturated. Continuous brine injection was conducted
80 to measure the permeability. Then we elevated the confining pressure (to 15 MPa) and
81 pore pressure (to 10 MPa) to investigate the poroelastic effect. After that the CO₂
82 drainage and brine imbibition were conducted. During the CO₂ drainage, the injection
83 pressure at the inlet and outlet were initially set to 10.1 and 10 MPa, respectively. Under
84 the low differential pressure, the CO₂ fluid flow front gradually migrated with the
85 displacement at the coarse-grain part for approximately 40 hours and then it was
86 blocked by one low-permeability capillary barrier in the fine-grain part. After we
87 elevated the inlet pressure up to 10.3 MPa step-by-step (8 steps, approximately 1
88 hour/step), the CO₂ flow was able to percolate into the fine-grain part. The overall
89 drainage persisted approximately 68 hours. After the drainage, a forced brine injection
90 from the outlet with a pressure of approximately 10.8 MPa was conducted. The brine
91 injection lasted approximately 20 hours. During these operations, the X-ray CT imaging
92 was only conducted at the day time intermittently to obtain the information of spatial
93 saturation of CO₂ in the rock sample. The distributed strain was continuously measured
94 using the DFOSS system.

95

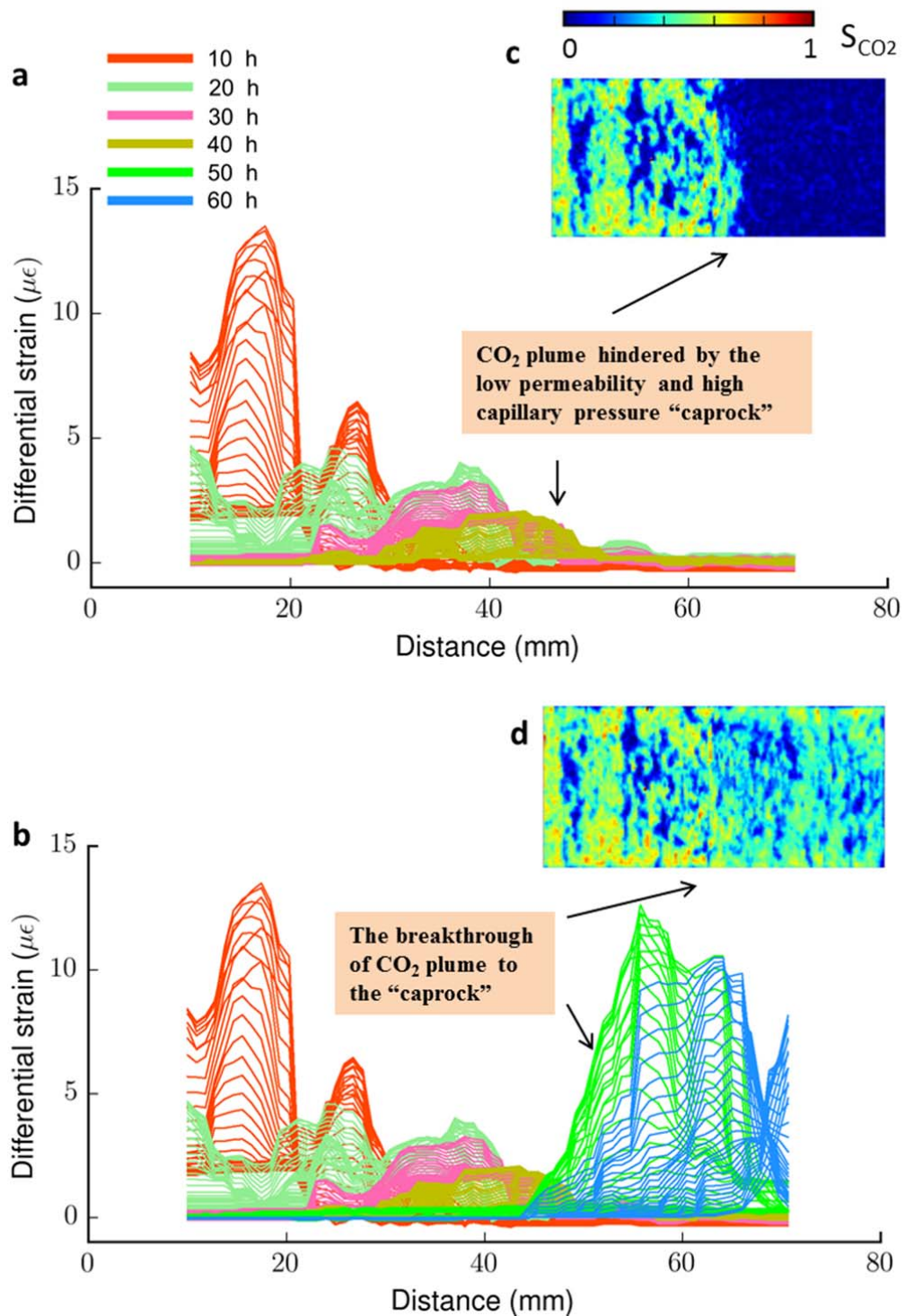


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98 Fig.1 **a**, Schematic geological model of CO₂ storage site, with emphasis on the clay-rich
 99 components in the caprock-reservoir transition, tight clay-rich layers within reservoir
 100 formations, and paleo-fault with clay smearing. An observation well with the DFOSS
 101 installation is used to illustrate the distributed strain sensing across the overall system. **b**,

102 The Tako sandstone sample—an analogue to the reservoir-caprock system or clay-rich
103 layers. **c**, The experimentally obtained profiles of averaged CO₂ saturation (S_{CO_2})
104 (computed from X-ray CT imaging) along the rock sample. Note that the displayed
105 saturation data are linearly interpolated and coloured with elapse time for a better view.
106 The only measured data is shown in the Supplementary Fig.7. **d**, The profiles of
107 circumferential strain (experimentally measured using the DFOSS) on the surface of the
108 rock sample. **e**, The circumferential strain against time coloured with spatial distance to
109 the inlet end of the sample. Note that the pressure increase between 40 and 50 hours
110 cause only slight swelling strains ($< 10 \mu\epsilon$).



111

112 Fig.2 Differential strain changes (strain/hour) calculated by subtracting two sets of
 113 measured strains with a time interval of approximately 3.3 hours. Overall results are
 114 divided in to six stages (with a time interval of approximately 10 hours) that expressed

115 in different colours to emphasize the temporary changes. **a**, Changes before stage 5
116 (former 40 hours); **b**, Changes of all stages. Colours in **a** and **b** denote time. **c/d**, Images
117 of CO₂ saturation (S_{CO_2}) at 40/60 hours are attached. The changes clearly show the
118 structure-affected deformation behaviour and the breakthrough of CO₂ plume from the
119 high permeability (“reservoir”) part to the low permeability (“caprock”) part.
120

121 **CO₂ plume migration and distributed strain response**

122 The experimental results reveal that the deformation in the reservoir-caprock system can
123 be effectively monitored using the DFOSS tool. The strain changes well correspond to
124 the spreading of CO₂ plume and show significant differences between the reservoir and
125 caprock parts. During the CO₂ drainage (Fig. 1c-e), the injected CO₂ displaced the brine
126 out from the rock sample as indicated by the saturation changes. The CO₂ plume was
127 retarded by the low-permeability caprock part and then migrated through it after
128 elevating the injection pressure. Before the breakthrough of CO₂ plume to the caprock
129 part, the corresponding strain mainly responded at the reservoir region with the
130 gradually increased strain magnitude (0~260 $\mu\epsilon$). Unlike the relatively steep gradients
131 near the CO₂ saturation front, the changes of strain showed a more gradual trend and
132 occurred earlier than the changes in the CO₂ saturation due to the mechanical effect and
133 forward pressure migration. However, the large changes (~200 $\mu\epsilon$) were mainly within
134 the reservoir part where the CO₂ plume developed. Moreover, the strain changes show
135 spatial dependence on the rock structure. Several local low-porosity layers showed
136 strain changes with a reduced magnitude and also a lower CO₂ saturation. These layers
137 acted as the temporal in-passing capillary barriers for the forward CO₂/brine
138 displacement.

139 The breakthrough of CO₂ plume occurred after the injection pressure was step-elevated
140 from 10.1 MPa to 10.3 MPa. Subsequently, significantly large and rapid strain changes
141 in the caprock part were observed (Fig. 1d and e). The breakthrough pressure for CO₂
142 percolating through the rock was less than 300 kPa under the setting condition. The
143 breakthrough behaviour was well indicated by the strain signals. Note that the pressure
144 change (200 kPa) itself only produced a small strain (< 10 $\mu\epsilon$) (Fig. 1e). The large strain
145 changes in the caprock part mainly occurred following the migration of CO₂ plume with
146 CO₂/brine replacement. The dynamic deformations are better depicted using the

147 differential strain between steps, which show more discernible time-dependent
148 behaviour (Fig. 2). Overall, during the drainage, the changes in strain can well reveal
149 the migration of CO₂/brine displacement front in the reservoir and breakthrough to the
150 caprock in the rock sample. On the contrary, during the imbibition, the rock sample
151 showed a general trend of shrinking after an initial short period of swelling caused by
152 the elevated pore pressure (Supplementary Fig. 8).

153 **Adsorption-induced swelling as a CO₂ migration indicator**

154 Note that the values of the strain changes are significantly larger than that would be
155 expected due to pure pore pressure effects as estimated from the poroelastic mechanism.
156 Under the fully brine saturated state, the bulk modulus of the rock was estimated
157 approximately 7.6 ~ 8.8 GPa (the coarse-grain part) and 8.3 ~ 10.0 GPa (the fine-grain
158 part, see Supplementary Fig. 9). Consequently, under the given maximum 300 kPa of
159 change in the effective pressure, the upper limit of the poroelastic (circumferential)
160 deformation was expected to be smaller than 20 $\mu\epsilon$. The limit well constraints the
161 actually produced strain changes (< 10 $\mu\epsilon$) due to pressure change (200 kPa) alone
162 when elevating the pore pressure during the drainage. However, the measured strain
163 values (~260 $\mu\epsilon$) after CO₂ displacing brine are in fact much larger than this limit, and
164 even larger than the strain value produced by 2 MPa of pore pressure changes
165 (approximately 100 $\mu\epsilon$) when the sample is fully saturated with brine (Supplementary
166 Fig. 9b).

167 The large changes in strain are likely related to the abundant content of clay mineral
168 (kaolinite) in the rock. CO₂ is preferentially adsorbed on clay minerals with respect to
169 brine. An injection of CO₂ into a clay-rich rock initially full of brine causes a
170 differential swelling of the porous matrix. In contrast, during brine imbibition, the CO₂
171 desorption by brine induces a shrinkage. Several recent studies have shown that CO₂
172 can be adsorbed in clay-rich rocks accompanying significant swelling¹⁰. The CO₂

173 adsorption in kaolinite was supported by both physical chemistry studies¹⁸⁻²⁰ and direct
174 experimental studies¹². Unlike the very large swelling strain ($\sim 10^4 \mu\epsilon$) observed in coal
175 samples due to CO₂ adsorption, however, the CO₂ adsorption induced strain in a
176 kaolinite bearing rock (i.e. shales) can be as large as $\sim 10^3 \mu\epsilon$ ¹². Compared with other
177 fluids, e.g., methane and water, the adsorption ability of CO₂ in clay minerals is more
178 outstanding^{12,21}. Such enhanced swellings can also be found in several previous studies
179 where strain measured using conventional strain gauges when CO₂ displacing brine in
180 rocks²². However, it lacks an explicit explanation in these studies.

181 We numerically simulated the effects of adsorption induced strain using a sequentially
182 coupled one-dimensional two-phase fluid flow and poromechanical model. A
183 Langmuir-type adsorption relationship with saturation scaling was used to model
184 adsorption induced strain changes during CO₂ saturating³². In order to see the maximum
185 value of probable deformations due to the poroelastic mechanism alone, with a
186 comparison to the adsorption included model, we used a lower limit of measured bulk
187 modulus (7.6 GPa). The modelling results show that the poroelastic alone can only
188 produce a maximum circumferential strain of approximately 16 $\mu\epsilon$ (Supplementary Fig.
189 10). In contrast, by adding the adsorption induced deformation mechanism, the
190 magnitude of measured strain ($\sim 260 \mu\epsilon$) can be well interpreted (Supplementary Fig.
191 11). Moreover, the modelling can generally reproduce the behaviour of the
192 breakthrough to caprock and structure-dependent deformations. The rock porosity
193 structure controls the spatial distribution of capillary entry pressure, which further
194 causes differences in the spatial CO₂ saturation. The latter controls the adsorption
195 induced deformations together with the role of pore pressure.

196 **Implications for reservoir monitoring using DFOSS**

197 Clay-rich rocks are common in a reservoir-caprock system for geological storage of
198 CO₂ and natural gas. These rocks may appear at the transition between the reservoir and

199 caprock, tight interbedding shale-contained layers within overall high-permeability
200 reservoir (e.g. the CO₂ storage sites at Sleipner, Norway²³ and Nagaoka, Japan) and
201 sealed paleo-fault zones with the clay smearing²⁴. In a large-scale CO₂ storage project,
202 during CO₂ injection and post-injection stages, the appearances and migrations of CO₂
203 and deformations at all these parts are critical and required to be closely monitored. In
204 addition, even some of the targeted reservoir formations for CO₂ storage have been
205 found to contain a rich amount of clay minerals. For example, the sampled rocks from
206 the Takinoue formation, which is the targeted CO₂ storage reservoir for a large-scale
207 CO₂ storage demonstration project at Tomakomai, Hokkaido, Japan, have been found
208 containing smectite and montmorillonite²⁵. Our studies indicate that the changes in
209 strain can be significantly enhanced (in several hundreds of $\mu\epsilon$) with the adsorption of
210 CO₂ in the clay-rich parts, under slight pore pressure changes, compared with the role of
211 poroelastic effect played alone. Moreover, to field applications, utilizing these signals,
212 one can directly monitor the migration of CO₂ plume—whether the CO₂ plume has
213 passed through the monitored regions (e.g., lower caprocks), and diagnose the potential
214 fluid leakage risk, to assist in further decision making, injection planning and reservoir
215 management. The study helps in how to distinguish between the pore pressure
216 propagation effects (pressure footprint) and CO₂ migration through caprock (saturation
217 footprint) from the deformation pattern and magnitude. The former shows gradual
218 changes with the magnitude increasing from small to large with the pressure build up;
219 whereas the latter could give a relatively large strain change in a short term once CO₂
220 has migrated to the clay-rich caprock.

221 Our study straightforwardly demonstrates that the spatial strain can be effectively
222 measured using the DFOSS with high accuracy and resolution. Overall, the utilization
223 of DFOSS tool can not only directly give the information in deformation but also the
224 migration of fluid plume in a reservoir-caprock system. The findings and application of
225 distributed fibre-optical strain sensor are thought to be advantageous in underground

226 reservoir monitoring and management. In practice, the technique can be integrated with
227 other tools, e.g., pressure¹⁶, seismic³³, electric, microseismic monitoring⁹, etc., for
228 further more solidly tracking CO₂ plume and diagnosing potential reservoir
229 geomechanical and fluid flow problems in a broad area. Here we focus on the strain
230 responses to CO₂ injections for the purpose of CO₂ storage in underground reservoirs.
231 However, the same technique can be also valid for other geological problems involving
232 geomechanical monitoring, e.g., fluid extractions or injections in oil and gas reservoirs²⁶
233 and geothermal exploitations, mine exploitations, crustal deformations relevant to
234 earthquakes²⁷, etc.

235

236

237 **End of main text**

238

239 **METHODS**

240 **Rock sample**

241 The rock sample used in this study is cored along a direction perpendicular to the
242 bedding plane from a low-permeability (~ 0.02 mD) heterogeneous Tako sandstone
243 (from Gunma, Japan). The cylinder sample, with a diameter of 35 mm and length of 80
244 mm, visually contains two regions, a coarse-grain and a fine-grain region (Fig. 1 b). The
245 two parts have different petrophysical characteristics, including permeability, porosity
246 (Supplementary Fig. 1) and pore size (Supplementary Fig. 2). The coarse-grain part has
247 a larger pore size, porosity and permeability than the fine-grain part. Both of the two
248 parts have high content of the clay mineral—kaolinite (Supplementary Fig. 3).
249 According to the results of X-ray powder diffraction (XRD) analysis, the mineral
250 compositions of the two parts (coarse and fine) are as follows: quartz (52.2% and
251 38.0%), kaolinite (36.0% and 52.8%), Muscovite (2.8% and 8.3%), K-feldspar (8.0%
252 and 0.9%) (Supplementary Fig. 4). The sample could be seen as an analogue for two
253 typical components in a reservoir-caprock system, such as the transition part between
254 caprock and reservoir, sand mud alternation, and the tight interbedding layers within a
255 reservoir (Fig. 1 a). The rocks in these components are usually clay-rich and have a low
256 permeability compared to common reservoir sandstones.

257 **Experimental settings**

258 A quartz-made single-mode holey optical fibre was spirally bonded on the surface of the
259 cylindrical sample in order to increase the measurement density (Supplementary Fig. 5).
260 The holey fibre, with empty holes distributed in the cladding outside fibre's glass core,
261 has a low bending loss for light signals, making it is suitable to measure the
262 circumferential deformation of cylindrical sample under high stress conditions. To be
263 able to apply a high injection pressure and confining pressure, the sample was jacketed

264 using the epoxy paste to insulate the confining oil and pore fluids and then put into a
265 core holder (Supplementary Fig. 6), which is made of an X-ray transparent and high
266 strength material (PEEK). Two high accuracy syringe pumps were connected to the two
267 side of rock sample to control the pressure and injection. In order to keep the CO₂ in a
268 supercritical condition, the pore pressure was set above 10 MPa and the temperature of
269 the whole system was kept at 40 °C using the carbon cloth heaters or water circulation
270 heaters. The fibre was passed through the being pressurized core holder by a
271 feedthrough component, which prohibits the leakage of fluids. The entire core holder
272 was put on the bed of a medical X-ray CT scanner to make imaging available. The fibre
273 segment was connected to prolonged segments and then connected to an integrated
274 measurement equipment (Neubrex 7020 type). The strain sensing tool is developed on
275 the basis of a coherent optical time-domain reflectometer (COTDR) method utilizing
276 Rayleigh backscattering signals from the inherent random defects in an intact fibre to
277 detect deformations^{28,29}. It can give a high measurement accuracy (0.5 µε) and spatial
278 resolution (2 cm), allowing to monitor very small strains in a long-range distributed
279 mode. The accuracy of the DFOSS technique has been demonstrated in several prior
280 tests with a comparison with the conventional strain gauges^{30,31} and successfully applied
281 in field wellbores with water injection tests and measurements³².

282 **Measurement principle of COTDR using optical fibre**

283 The COTDR (coherent optical time domain reflectometry) technique calculates strain or
284 temperature using the shift in the frequency of the power spectral of the Rayleigh
285 backscatter traces from a single-mode fibre²⁸. In the method, the light source is
286 precisely frequency-controlled. The frequency shift is calculated using the cross-
287 correlation between two spectral of COTDR measurements at two time points. At a
288 spatial location of a fibre, if there is no change in strain and temperature during the time
289 interval, the two power-spectral should be coincident in the frequency domain. Instead,

290 a phase difference between them exists if there are changes in strain or temperature due
 291 to the changes in local refractive index. The strain or temperature is calculated from the
 292 shift in the frequency of the power spectral.

293 **Model of adsorption induced swelling strain**

294 The adsorption induced swelling strain ε in confine pores can be described by a refined
 295 poromechanical model³³,

$$296 \quad \varepsilon = \varepsilon_0 + \int_{P_{b0}}^{P_b} \frac{dP_b}{K} \{ \alpha(1 - \chi)^{-1} - 1 \} \quad (1)$$

297 where ε_0 is the initial strain, K is the bulk modulus, α is the Biot's coefficient, P_b ($\mathbf{0}$
 298 denotes the initial state) is the new defined effective pore pressure, and χ is the
 299 confinement degree. The confinement degree χ scales the pore fluid pressure P_f and P_b
 300 as

$$301 \quad dP_f = \frac{dP_b}{1 - \chi} \quad (2)$$

302 The equation (1) predicts a swelling strain similar to the Langmuir type sorption model.
 303 Analytically, it is equivalent to an empirical Langmuir type deformation model³⁴,

$$304 \quad \varepsilon_{\text{sorp}} = \frac{\varepsilon_{\text{max}} P}{P + P_\varepsilon} \quad (3)$$

305 where $\varepsilon_{\text{sorp}}$ is the adsorption induced volumetric strain at pressure P , and ε_{max} and P_ε
 306 are similar to the Langmuir-type constants for scaling the strain.

307 **The poroelastic model for the two-phase fluid flow**

308 A sequentially coupled one-dimensional two-phase fluid flow and poromechanical
 309 model was used for the numerical simulation. The model was developed on the basis of
 310 an open source reservoir simulator (MRST)³⁵.

311 The two-phase fluid flow equation is as following,

$$312 \quad \frac{\phi \rho_\alpha S_\alpha}{\partial t} + \nabla \cdot (\rho_\alpha v_\alpha) = \rho_\alpha Q_\alpha \quad (4)$$

313 where ϕ is the porosity, α (w or nw) denotes the wetting and nonwetting fluid, ρ and S
314 are the density and saturation of the fluids, and Q is the source. The flow flux v is
315 expressed using the extended Darcy's law,

$$316 \quad v_\alpha = -\frac{K k_{r\alpha}}{\mu_\alpha} (\nabla p_\alpha - \rho_\alpha g \nabla h) \quad (5)$$

317 where K is the permeability, $k_{r\alpha}$ is the relative permeability, μ_α is the viscosity of a
318 fluid, p is the pressure, and $\rho_\alpha g \nabla h$ accounts for the gravity force.

319 In the modelling, the permeability at each location is estimated using the Leverett J-
320 function as following³⁶,

$$321 \quad K_i = \phi_i \frac{1}{\bar{P}_c(S_w)^2} [\sigma \cos(\theta) J(S_{w,i})]^2 \quad (6)$$

322 where i denotes the spatial location, σ and θ are the interfacial tension and contact
323 angle between CO₂ and brine, and \bar{P}_c is the global capillary pressure at each saturation
324 (S_w) estimated from mercury injection method. The Leverett J-function J is expressed as

$$325 \quad J(S_w) = \frac{P_c}{\sigma \cos \theta} \sqrt{\frac{k}{\phi}} \quad (7)$$

326 The local capillary pressure is also accounted by scaling the global capillary pressure
327 using local porosity ratio as following,

$$328 \quad P_c(S_w, i) = \bar{P}_c(S_w, i) \left(\frac{\phi_0}{\phi}\right)^n \quad (8)$$

329 where ϕ_0 is the mean porosity and n is a tuning exponent.

330 The mechanical responses are modelled using following equations,

$$331 \quad \nabla \cdot \sigma_{ij} + f = 0 \quad (9)$$

$$332 \quad \varepsilon_{ij} = \frac{1}{2} (\nabla u_i + \nabla u_j) \quad (10)$$

333
$$\varepsilon_{ij} = \frac{1}{2G} \sigma_{ij} - \left(\frac{1}{6G} - \frac{1}{9K} \right) \sigma_{kk} \delta_{ij} + \frac{b}{3K} p \delta_{ij} + \frac{\varepsilon_{\text{sorp}}}{3} \delta_{ij} \quad (11)$$

334 where σ_{ij} is the stress tensor, f is the body force, ε_{ij} is the strain tensor, u_i is the
 335 displacement, G and K are the shear and bulk modulus, b is the Biot's coefficient, p is
 336 the pore pressure, δ_{ij} is the Kronecker delta, and $\varepsilon_{\text{sorp}}$ is the adsorption-induced strain.

337 **Calculation of CO₂ saturation from X-ray CT imaging**

338 A medical X-ray CT scanner (Aquilion ONE TSX 301A, Toshiba Medical Systems
 339 Corp.) was utilized to image the rock sample. The reconstructed image volume has total
 340 160 slices along the axis direction (80 mm long). Each slice has total 512×512 pixels.
 341 The length of each pixel is approximately $71 \mu\text{m}$.

342 The following equation is used to determine the CO₂ saturation, S_{CO_2} , of each voxel,

343
$$S_{\text{CO}_2} = c \left(CT_{\text{obs}} - CT_{\text{brine}}^{\text{sat}} \right) = \frac{CT_{\text{obs}} - CT_{\text{brine}}^{\text{sat}}}{CT_{\text{CO}_2}^{\text{sat}} - CT_{\text{brine}}^{\text{sat}}} \quad (12)$$

344 where $CT_{\text{brine}}^{\text{sat}}$ and $CT_{\text{CO}_2}^{\text{sat}}$ are the voxel CT values of the brine-saturated and CO₂
 345 saturated states, respectively, CT_{obs} is the CT value of the state which saturation is
 346 being calculated, c is the coefficient that relates S_{CO_2} to the changes between CT
 347 values.

348 **Mineral analysis**

349 The mineral analysis was conducted using the Rigaku Smartlab Intelligent X-ray
 350 diffraction (XRD) system.

351 **Acknowledgements**

352 This work is part of an R&D project “the Development of Safety Management
 353 Technology for Large-Scale CO₂ Geological Storage, commissioned to the Geological
 354 Carbon Dioxide Storage Technology Research Association by the Ministry of Economy,
 355 Trade and Industry (METI) of Japan”. We thank K. Nakano for the help in XRD
 356 mineral analysis.

357 **Author contributions**

358 Z.X. and Y.Z. conceived the project, Y.Z. designed the experiments, H.P., Y.Z. and
 359 T.K. performed the experiments, Y.Z., J.S. and X.L. contributed to data analysis and

360 theoretical interpretations, Y.L contributed to theoretical interpretations and molecular
 361 dynamics simulations, Y.Z wrote the first draft of the paper and all co-authors improved
 362 the final version.

363 **Data availability**

364 The authors declare that all necessary data supporting the findings of this study are
 365 available within the article and its Supplementary Information files. Any further data
 366 (for example, X-ray CT images and strains) are available from the corresponding author
 367 upon request.

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