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## **Earthquakes produce carbon dioxide in crustal faults**

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Fourier transform infrared (FTIR) micro-analysis of pseudotachylytes (i.e. friction-induced melts produced by seismic slip) from the Nojima fault (Japan) reveals that earthquakes almost instantaneously expel 99 wt% of the wall rock CO<sub>2</sub> content. Carbon is exsolved because it is supersaturated in the friction melts. By extrapolation to a crustal-scale fault rupture, large events such as the M7.2 Kobe earthquake (1995) may yield a total production of 1.8 to  $3.4 \times 10^3$  tons CO<sub>2</sub> within a few seconds. This extraordinary release of CO<sub>2</sub> can cause a flash fluid pressure increase in the fault plane, and therefore enhance earthquake slip or trigger aftershocks; it may also explain the anomalous discharge of carbon monitored in nearby fault springs after large earthquakes. Because carbon saturation in silicate melts is pressure-dependent, FTIR can be used as a new tool to constrain the maximum depth of pseudotachylyte formation in exhumed faults.

**Keywords:** pseudotachylyte, infrared spectroscopy, fluids, CO<sub>2</sub> exsolution, earthquake, slip weakening, Nojima fault.

## Introduction

An earthquake occurs because the shear resistance of a slipping fault becomes weaker with increasing slip [1]. The fault rocks are crushed by friction and may even melt if the heat generated is large enough [2]. Knowing how much energy is released as frictional heat, or converted into fractures and elastic waves, is essential for assessing how tectonic stress relaxation generates near-fault acceleration and surface damage, and how much stress is left

for future rupture of the Earth's crust. However, the production of heat is poorly constrained in the energy budget of earthquakes because the magnitude of fault weakening and its mechanism are not well known. In most theoretical or experimental mechanisms proposed to explain slip weakening, the lubricating agent is a fluid medium (either aqueous, melt or slurry) trapped within the fault plane and put under pressure by thermal or dynamic effects [3-8]. The efficiency of these slip-weakening mechanisms depends largely on the quantity of fluid involved and the pressurization achieved, which are difficult to assess. Therefore, determining the magnitude of coseismic shear stress reduction and the energy balance of earthquakes is still a major issue in seismology.

To constrain the fluid budget of a slipping fault and improve our understanding of weakening processes, we have analyzed the water and carbon dioxide content of pseudotachylytes (i.e. melts produced by friction during earthquakes) using a Fourier transform infrared microspectrometer (micro-FTIR). FTIR is a conventional tool for the study of volatile species in magmatic melts [9], albeit never applied to friction melts so far. We show that pseudotachylytes may be used to infer the fluid concentration in fault rocks at the time of seismic slip, providing new insights on earthquake mechanics.

The studied example is the Nojima fault (Fig. 1) that was activated on a dextral, 10.5 km-long surface rupture [10] by the Kobe (Hyogo-ken Nanbu) earthquake ( $M_{\text{JMA}} = 7.2$ ,  $M_{\text{W}} = 6.9$ , focal depth = 16 km, January 17<sup>th</sup>, 1995), killing more than 6400 people. Immediately after the shake, the  $\text{HCO}_3^-$  concentration increased by 30 wt% in nearby springs [11]. This unexplained carbon discharge, together with other coseismic geochemical anomalies, decreased gradually to normal values in the following ten months [12-15]. One year after the earthquake, the Geological Survey of Japan drilled a borehole (Fig. 1) that reached the

upper boundary of the 83° dipping fault at 623.1 m depth within Cretaceous granodiorites [16]. The fault core consists in a 30 cm-thick cataclasite composed of quartz, albite and K-feldspar clasts derived from the granodiorite protolith, crushed together with secondary clays, zeolites and carbonates [16, 17]. The carbonates have a biogenic isotopic signature as well as present-day pore fluids in the fault, which suggests a superficial or sedimentary source for the alteration fluids in general and the carbon in particular [18-20]. Because of hydrothermal alteration, the cataclasite is enriched in SiO<sub>2</sub>, CaO and depleted in MgO, Fe<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>O, K<sub>2</sub>O relative to the granodiorite protolith. The cataclasite also contains 3.5 to 10 wt% equivalent CO<sub>2</sub> with an average at 7.6 wt%, and 1 to 5 wt% H<sub>2</sub>O with an average at 2.7 wt% (mean of 5 measurements taken from Figure 7 in [17]). This study focuses on two millimetre-thick pseudotachylytes (Fig. 2a) embedded in the cataclasite layers at 624.59 m within the Nojima fault core [17]. The texture of these pseudotachylytes has already been investigated in detail [21, 22] and is only summarized briefly. The dark layer of pseudotachylyte (PT1) contains fragments of cataclasite (isolated or aggregated quartz, albite and calcite clasts with molten rims) in a brown matrix. These fragments, and the fresh PT1 – cataclasite interface devoid of shear localization structures, indicate that the cataclasite is the precursor of PT1. A second, clearer pseudotachylyte (PT2), containing plastically deformed fragments of PT1 and fewer mineral remnants in a transparent matrix (Fig. 2a), re-melted the dark layer. The random occurrence of fluid-filled vesicles in the two matrixes, and the homogeneous aspect of these matrixes under the SEM [21] suggest that the two pseudotachylytes are made of undevitrified glass [22]. The composition of the vesicles (84 mol% H<sub>2</sub>O; 15.3 mol% CO<sub>2</sub>; 0.7 mol% NaCl) attest to the supersaturation of H<sub>2</sub>O and CO<sub>2</sub> in the melts [22]. In addition, the molten rims of clasts in the two

pseudotachylytes indicate that melting reached a minimum temperature of 1200 °C [22], which is consistent with melting temperatures estimated from other pseudotachylyte samples in the Nojima fault [23]. The two matrixes have a dacite composition (~68 wt% SiO<sub>2</sub>, 3 wt% Na<sub>2</sub>O + K<sub>2</sub>O) similar to the bulk composition of the cataclasite. Calcite fragments in the melts, and the fact that both matrixes contain up to twice the CaO content of the granodiorite protolith (4.06 wt%), imply that the precursory cataclasite was already carbonate-rich before melting. Carbonate precipitation continued between and after the melting events [24]. Pseudotachylytes were quenched at ambient temperatures of 150 – 350 °C and a depth range of 4 – 15 km [22, 25, 26]. These pseudotachylytes formed well after the cooling of the granodiorite basement (74 Ma [27]), and were then uplifted with the basement by a left-lateral thrust motion of the fault, mostly before the Eocene [26]. Exhumation is still occurring in the present even though the fault has shifted to a right-lateral thrust movement since the Plio-Pleistocene [28]. It is therefore likely that these exhumed pseudotachylytes represent slipping processes similar to those occurring today between the Kobe earthquake hypocenter and the surface. The multiple occurrence of pseudotachylytes within carbonate-bearing cataclasites on surface exposures of the Nojima fault [23], and on other faults along the Median Tectonic Line running through Japan, suggests that the lateral extent of melts is large, and/or that friction melting is a widespread phenomenon along those faults.

## **Analytical methods and results**

Infrared spectra were measured on a JASCO, FTIR 620plus, IRT 30 apparatus, equipped with an automatic XY mapping stage moving the sample at 50  $\mu\text{m}$  steps on X and Y under the microscope. Spectra were acquired by collecting 50 scans in the range 6000 – 700  $\text{cm}^{-1}$  with a 4  $\text{cm}^{-1}$  resolution and a 50  $\times$  50  $\mu\text{m}$  aperture. Every ten spectra during the mapping, the automatic stage moved to a position beyond the sample in order to take a background air spectrum. All the spectra were automatically corrected from background by the JASCO Spectra Manager software, and then corrected using the same linear baseline in the range 5000 – 1300  $\text{cm}^{-1}$ . The thickness of the wafer was measured using a laser scanning confocal microscope (KEYENCE VK-8500) at the centre and corners of each of the mapped areas, and was found to be  $63 \pm 3 \mu\text{m}$ . Absorbance peaks were converted into volatile concentrations following the conventional procedure for  $\text{SiO}_2$ -rich melts [9], assuming a glass density of 2.7  $\text{g cm}^{-3}$ . The total error on concentration estimates is 5 to 10 % [9]. Concentration maps were built using the JASCO Spectra Manager software.

Four areas were analyzed by FTIR in the sample (Fig. 2). Each 50  $\times$  50  $\mu\text{m}$  pixel corresponds to an infrared spectrum, and hence to a couple of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  concentrations (typical FTIR spectra provided in Fig. 3). The measured concentrations include the fraction of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  dissolved in the glass and the fraction trapped in fluid inclusions. Water is observed entirely as molecular  $\text{H}_2\text{O}$  (no dissolved OH). The two pseudotachylytes display a similar water content of about 2 wt%  $\text{H}_2\text{O}$  (Fig. 2b), except in cracks. There is, however, an extremely sharp contrast of molecular  $\text{CO}_2$  concentration between the dark older PT1 containing up to 5000 ppm  $\text{CO}_2$ , and the clear younger PT2 nearly devoid of  $\text{CO}_2$  (Fig. 2c). The presence of water and carbon dioxide dissolved as molecular species, not as ionic

species, is typical of dacite type, silica-rich melts [29]. In addition,  $\text{CO}_3^{2-}$  is detected as secondary carbonate minerals (mostly calcite) precipitated in microcracks or as mineral fragments in the glass (Fig. 2d). PT1 contains 1 wt%  $\text{CO}_3^{2-}$  or 0.7 wt% equivalent  $\text{CO}_2$  (average of all the spectra measured on PT1). Because PT2 is younger and less fractured than PT1, it contains only 0.3 wt%  $\text{CO}_3^{2-}$  or 0.2 wt% equivalent  $\text{CO}_2$  in average. FTIR data are also reported in an  $\text{H}_2\text{O}$ - $\text{CO}_2$  diagram before (Fig. 4a) and after filtering (Fig. 4b) in order to consider only pure matrix glass without carbonates. PT1 contains 1.5 – 2.8 wt%  $\text{H}_2\text{O}$  and 1000 – 5000 ppm  $\text{CO}_2$ , whereas PT2 contains 1 – 2.8 wt%  $\text{H}_2\text{O}$  and 0 – 500 ppm  $\text{CO}_2$  away from its contact with PT1.

## Discussion and conclusion

### 1. Budget of volatile exsolution by friction melting

Our results show that friction melting causes carbon devolatilization of the fault rocks. Because PT1 contains up to 0.7 wt% equivalent  $\text{CO}_2$  as crystallized carbonates (i.e. 1 wt%  $\text{CO}_3^{2-}$ , Fig. 2d), some carbonates may postdate the first melting event such that the cataclasite possibly contained less carbon before PT1 formation than it does now, i.e. only 2.8 – 6.9 – 9.3 wt% equivalent  $\text{CO}_2$  (minimum – average – maximum of Tanaka's et al. measurements [17], minus 0.7 wt%). In order to set a lower bound to our quantification of  $\text{CO}_2$  loss, we therefore assume the precursory cataclasite to have contained no more than 2.5 – 3.5 wt% equivalent  $\text{CO}_2$  before PT1. Even though, PT1 (1.5 – 2.8 wt%  $\text{H}_2\text{O}$ , 1000 – 5000 ppm  $\text{CO}_2$ ) contains as much  $\text{H}_2\text{O}$  as the parent cataclasite, but about ten times less  $\text{CO}_2$ . Likewise, PT2 (at 1 – 2.8 wt%  $\text{H}_2\text{O}$ , 0 – 500 ppm  $\text{CO}_2$  away from its contact with



PT1) contains as much  $\text{H}_2\text{O}$  as PT1 from which it melted, but ten times less  $\text{CO}_2$  (for a minimum estimate of the  $\text{CO}_2$  loss, we neglect the carbonates precipitated after PT1 and before PT2). Therefore, each fusion expels at least 90 wt% of the  $\text{CO}_2$  dissolved or trapped as carbonates in the protolith, because carbon is supersaturated in the molten phase. Only 1 wt% of the initial  $\text{CO}_2$  content is kept in the fault plane after the two melting events, while the remaining 99 % are exsolved as fluid  $\text{CO}_2$ .

Assuming a rock density of  $2.7 \text{ g.cm}^{-3}$ , the fusion of  $10^{-3} \text{ m}^3$  (i.e. 1 mm thickness  $\times$  1  $\text{m}^2$  of fault plane) of cataclasite (at 2.5 – 3.5 wt%  $\text{CO}_2$ ) into PT1 (at 1000 – 5000 ppm  $\text{CO}_2$ ) yields 56 to 95 g  $\text{CO}_2$ . Likewise, the re-fusion of  $10^{-3} \text{ m}^3$  of PT1 into PT2 (at 0 – 500 ppm  $\text{CO}_2$ ) releases 1.4 to 14 g  $\text{CO}_2$  in 1  $\text{m}^2$  of fault plane. Frictional melting is therefore an unexpected and significant source of  $\text{CO}_2$  in the Nojima fault.

## 2. Degassing behaviour of friction melts

FTIR data yield constraints on the ambient pressure during friction melting, because  $\text{H}_2\text{O}$  and  $\text{CO}_2$  saturation in silicate melts is pressure-dependent. Spectra measured in PT1 define a trend in the high  $\text{CO}_2$  field, whose slope indicates that the supersaturated melts were quenched while degassing an 80 mol%  $\text{H}_2\text{O}$  and 20 mol%  $\text{CO}_2$  fluid mixture (Fig. 4b), consistent with the measured composition of fluid inclusions [22]. The lower end of the degassing trend (2 wt%  $\text{H}_2\text{O}$ , 1000 ppm  $\text{CO}_2$ ) is the closer to the composition of the pure matrix glass without fluid inclusions. Considering the  $\sim 1200^\circ\text{C}$  temperature of melting, a maximum pressure of 300 MPa may then be estimated for the formation of the PT1 glass using the VolatileCalc software [30], or a 11 km depth assuming a  $2.7 \text{ g.cm}^{-3}$  rock density.

Spectra acquired in PT2 lie in the low CO<sub>2</sub> range (Fig. 4b). The low slope of PT2 degassing path suggests that the glass exsolved a much higher proportion of H<sub>2</sub>O relative to CO<sub>2</sub> than PT1 ( $\geq 97$  mol% H<sub>2</sub>O,  $\leq 3$  mol% CO<sub>2</sub>). If PT2 ever reached equilibrium saturation, it is again in the lower end of the degassing path at less than 2 wt% H<sub>2</sub>O and 200 ppm CO<sub>2</sub>, or a 130 MPa maximum pressure (5 km). The consistency of these depths with independent depth estimates [25, 26] implies that PT1 and PT2 nearly reached equilibrium volatile saturation. Our results suggest that the two melting events occurred at different depths, and therefore were separated by a large time span. FTIR mapping is a new method that provides depth constraints on the formation of individual layers of glassy pseudotachylyte in exhumed faults, without any assumption on the geotherm.

In addition, the solubility laws of volatiles in silicate melts may be used to explain the degassing pattern during friction melting. In doing so, we assume that H<sub>2</sub>O and CO<sub>2</sub> exsolution in pseudotachylytes is ruled by local liquid-gas equilibrium as a first approximation, keeping in mind that true equilibrium was probably not achieved within the short time lapse of heating and quenching. Under this assumption, CO<sub>2</sub> exsolves faster than H<sub>2</sub>O due to the low solubility of carbon compared to H<sub>2</sub>O. Consequently, a silicate melt degassing in an open system (i.e. the exsolved fluid mixture is removed from the melt) follows a steep path in the H<sub>2</sub>O-CO<sub>2</sub> space, because CO<sub>2</sub> is almost entirely lost before H<sub>2</sub>O supersaturation begins. On the other hand, a magma degassing in a closed system (in which the exsolved fluid stays in contact with the melt) follows a gently sloping path in the H<sub>2</sub>O-CO<sub>2</sub> space, because the partial pressure of CO<sub>2</sub> gradually increases in the exsolved phase and hampers further CO<sub>2</sub> degassing. Accordingly, FTIR data reveal that each pseudotachylyte degassing proceeds in two stages: 1) Loss of 90 wt% CO<sub>2</sub> without

significant H<sub>2</sub>O loss from the cataclasite to PT1, and from PT1 to PT2 (dashed arrows in Fig. 4b). This degassing path is consistent with an open system that prevails as long as the melt viscosity is low enough for CO<sub>2</sub>-rich bubbles to escape the pseudotachylytes. Because it is an open system, this stage is not recorded in H<sub>2</sub>O – CO<sub>2</sub> concentrations, yet is evidenced by comparing volatile concentrations before and after melting. 2) Loss of water-rich mixtures (80 mol% H<sub>2</sub>O, 20 mol% CO<sub>2</sub> for PT1;  $\geq 97$  mol% H<sub>2</sub>O,  $\leq 3$  mol% CO<sub>2</sub> for PT2) is consistent with melts degassing in closed system (solid lines in Fig. 4b). This second stage of closed system degassing occurs because the viscosity of the cooling melts becomes too large to let the bubbles escape, resulting in the entrapment of fluid inclusions. To summarize, pseudotachylytes in the Nojima fault behave as open systems at the beginning of flash friction melting, and then as closed systems during the last stage of melt cooling.

### 3. Implication for fault mechanics

The first essential implication of our findings is that CO<sub>2</sub> degassing may increase the fluid pressure (P) in the fault zone during and/or after an earthquake, depending on the rate of exsolution, the permeability of the fault core, the carbonate content of the precursory fault rocks and the depth considered. To estimate the fluid pressure increase solely caused by frictional exsolution, we consider the effect of CO<sub>2</sub> emission at constant temperature (~200 °C at 5 km and ~400 °C at 11 km), and neglect elastohydrodynamic or thermal pressurization [4, 6-8], keeping in mind that these effects may be considerable if combined with exsolution. Moreover, the ~15 volume % increase caused by the transformation of

cataclasite into pseudotachylyte is neglected, although it may contribute to increasing the fluid pressure and reducing the shear stress on the fault plane. For the sake of simplicity, we also consider the porosity of the fault zone to be entirely filled with pure CO<sub>2</sub>, even though other fluids such as H<sub>2</sub>O may be dominant before an earthquake. This assumption does not yield significant error in the calculation because the compressibility factors and viscosities of H<sub>2</sub>O and CO<sub>2</sub> are nearly equal in the supercritical state within the pressure and temperature ranges considered here. Accordingly, the fluid pressure increase ( $P_f/P_0$ ) is computed from the equation of state (EOS) of pure CO<sub>2</sub>:

$$P = \frac{mZRT}{M^{CO_2} \phi_w}, \quad (1)$$

which yields

$$\frac{P_f}{P_0} = \frac{\phi_0 Z_f m_f}{\phi_f Z_0 m_0} \quad (2)$$

Parameter values, units and definitions are given in Table 1. The subscripts 0 and *f* refer to quantities before and after exsolution, respectively. *R*, *T*,  $\phi$  and  $M^{CO_2}$  denote the gas constant, the temperature, the porosity of the fault zone and the molar mass of CO<sub>2</sub>, respectively. *Z*, the compressibility factor of pure CO<sub>2</sub>, is calculated using the Saxena and Fei EOS [31]:

$$Z = (1 - 0.5917) \left( \frac{T^c}{T} \right)^2 + 0.09122 \frac{T^c P}{T P^c} + \left[ 1.4164 \times 10^{-4} \left( \frac{T^c}{T} \right)^2 - 2.8349 \times 10^{-6} \ln \frac{T}{T^c} \right] \left( \frac{P}{P^c} \right)^2 \quad (3)$$

where  $P^c$  and  $T^c$  represent the pressure and temperature at the critical point of CO<sub>2</sub>.  $m_0$  and  $m_f$  are the masses of CO<sub>2</sub> present before and after friction melting in a volume of fault zone

221 equal to  $1 \times 1 \times w \text{ m}^3$ .  $m_0$  is calculated assuming hydrostatic pressure in equation (1).  $m_f$  is  
 222 equal to  $(m_0 + m_e)$ , where  $m_e$  is the mass of  $\text{CO}_2$  exsolved by a 1 mm thick  
 223 pseudotachylyte.  $w$  is the maximum distance of coseismic  $\text{CO}_2$  outflow in the direction  
 224 perpendicular to the fault plane, taken as the half width of the pseudotachylyte (0.5 mm)  
 225 plus twice the hydraulic diffusion length scale of  $\text{CO}_2$  ( $L_f$ ).  $L_f$  is given by  $L_f = \sqrt{D_f \psi}$  [8],  $\psi$   
 226 being the characteristic timescale of frictional exsolution, set at 5 sec (the time lapse for  
 227 glass melting and quenching does not exceed a few seconds [32]).  $D_f$  is the hydraulic  
 228 diffusivity after exsolution, given by  $D_f = k_f / \eta \beta_f$ , where  $k_f$  denotes the final permeability  
 229 of the fault zone,  $\beta_f$  its storage capacity, and  $\eta$  the P-T dependent viscosity of supercritical  
 230  $\text{CO}_2$ .  $\beta_f$  is computed from  $\beta_f = \beta^b - \beta^s + \phi_f(\beta_f^{\text{CO}_2} - \beta^s)$ ,  $\beta^s$  being the compressibility of  
 231 mineral grains,  $\beta^b$  the bulk rock compressibility, and  $\beta_f^{\text{CO}_2} = -\frac{P_f}{Z_f} \left( \frac{\partial Z/P}{\partial P} \right)_f^{T=\text{cte}}$  the  
 232 compressibility of supercritical  $\text{CO}_2$ .

233 To calculate the pressure-dependent permeability of the slipping fault ( $k_f$ ), we use an  
 234 empirical law (Fig. 5) derived from the available data at different effective pressures ( $P_{\text{rock}} -$   
 235  $P$ ):

$$236 \quad k = 3 \times 10^{-17} e^{-6.14 \times 10^{-8} (P_{\text{rock}} - P)} \quad (4)$$

237 Note that permeability in the Nojima fault is at least two orders of magnitude larger than  
 238 the permeability of some other crustal faults (at similar effective pressures) investigated so  
 239 far [8, 33-36]. In absence of specific laws of pressure dependence for  $\phi$  and  $\beta^b$  on the  
 240 Nojima fault core, we are left to adapt to our case the empirical relations determined on the  
 241 Median Tectonic Line [8]:

$$\phi = \phi e^{-2.97 \times 10^{-8} (P_{rock} - P)} \quad (5)$$

$$\beta^b = 2.5 \times 10^{-10} e^{-1.38 \times 10^{-13} (P_{rock} - P)} \quad (6)$$

using  $\phi = 0.03$ , which is the mean porosity measured on samples from the Nojima fault core at room pressure [37].

The fluid pressure increase ( $P_f/P_0$ ) due to frictional exsolution is depicted in Figure 6. At 11 km depth, a flash release of 56 – 95 g CO<sub>2</sub> per m<sup>2</sup> of fault plane raises the pore fluid pressure of 78 to 90 % above the hydrostatic pressure, or an 80 – 100 MPa increase, even without considering thermal pressurization. At 5 km, the release of 1.4 – 14 g CO<sub>2</sub> raises the fluid pressure by 5 – 15 % above the hydrostatic pressure. Note that the magnitude of fluid pressure increase strongly depends on the permeability of the fault core (Fig. 6), as do other mechanisms of pore pressurization. For example, at 11 km depth the fluid pressure increases by 100 – 110 % if the permeability is ten times smaller than the selected permeability law (Fig. 5), and is still of 50 – 60 % if the permeability is ten times larger. Frictional CO<sub>2</sub> exsolution can therefore be a significant pressurization mechanism in fault zones, even at relatively high permeability, and even at hydrostatic starting fluid pressure. Slip-weakening by frictional exsolution should be taken into account when considering earthquake processes at slip velocities large enough to produce friction melts. In addition, the low velocity of seismic waves from the Kobe earthquake aftershocks suggests the presence of high pore fluid pressures in the fault zone after the main shock [38]. Since the coseismic exsolution of CO<sub>2</sub> may sustain such fluid pressurization, it could also enhance later ruptures and trigger aftershock sequences in the Nojima fault, in a same way than mantle CO<sub>2</sub> degassing triggers aftershock trees in thrust faults from Northern Italy [39].

264

265 **4. Implication for fault degassing**

266 The second major consequence of our findings is that co-seismic carbon discharge in near-  
267 fault springs might result from friction melting. The relative abundance of pseudotachylytes  
268 in carbonate-rich cataclasites on different cores and outcrops of the Nojima fault suggest  
269 that coseismic CO<sub>2</sub> loss might be the rule rather than an exception. In addition, core sample  
270 studies conclude that magnitude 6 – 7 earthquakes are responsible for the formation of  
271 millimetre thick pseudotachylytes [22]. In the following discussion, we assume that an  
272 earthquake such as the M7.2 1995 Kobe earthquake produces pseudotachylytes at least as  
273 wide as those observed in cores [22] and outcrops [23] of the Nojima fault. Under this  
274 assumption, the friction melts caused by a single earthquake at 11 km and 5 km along the  
275 fault plane should be similar to the pseudotachylytes (PT1 and PT2) caused by two separate  
276 slip events in the sample. To calculate the minimum CO<sub>2</sub> production of a Kobe-like  
277 earthquake, we hence consider a very idealistic model in which the rupture produces PT1  
278 from 11 km to 8 km and PT2 from 8 km to 5 km. The production of CO<sub>2</sub> beneath 11 km  
279 and above 5 km is unknown and therefore not considered, even though it might be  
280 significant. Accordingly, a minimum range of 170 to 330 kg CO<sub>2</sub> is released per meter of  
281 fault trace, or a total emission of  $1.8 - 3.4 \times 10^3$  tons CO<sub>2</sub> for a 10.5 km surface rupture of the  
282 Nojima fault such as in the Kobe earthquake. Note that thermal decomposition of  
283 carbonates in cataclasites could also contribute to release carbon in the Nojima fault, and  
284 hence increase the proposed budget. However, a large part of this CO<sub>2</sub> likely re-precipitates  
285 as new carbonate minerals soon after the earthquake. Yet a fraction of CO<sub>2</sub> may be

evacuated through the ground waters that leach the fault. Indeed, an excess of ~20 tons equivalent CO<sub>2</sub> was emitted by the Awaji Island springs within the ten months of the Kobe earthquake [11, 14], i.e. about 1 wt% of the CO<sub>2</sub> possibly produced by friction melting. As the carbon isotopic signature in past and present ground waters from the Nojima fault is biogenic and not consistent with a deep degassing source [18, 19], the coseismic carbon discharge in springs might well have come from frictional exsolution. This possibility opens promising applications for soil and spring gas monitoring in active faults, such as in the San Andreas fault that is also degassing biogenic carbon [40]. In the absence of a deep contribution, post-seismic CO<sub>2</sub> discharge might be taken as an evidence of frictional exsolution. The amount of CO<sub>2</sub> released should be proportional to the amount of heated carbon-bearing fault rock, and to the heat generated by shear friction. In deep wells probing those faults, measuring the CO<sub>2</sub> flux could therefore provide an additional tool to constrain the frictional strength of the fault, and the physical processes that govern seismic slip.

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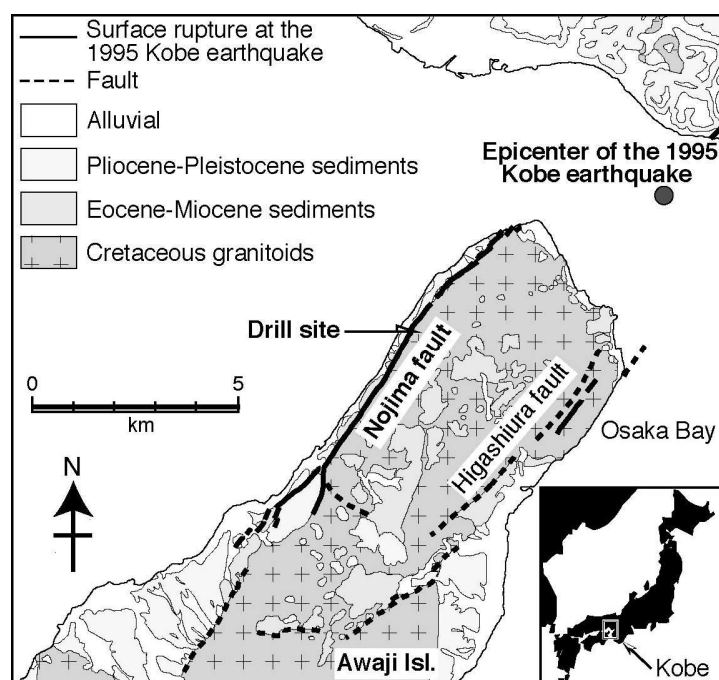
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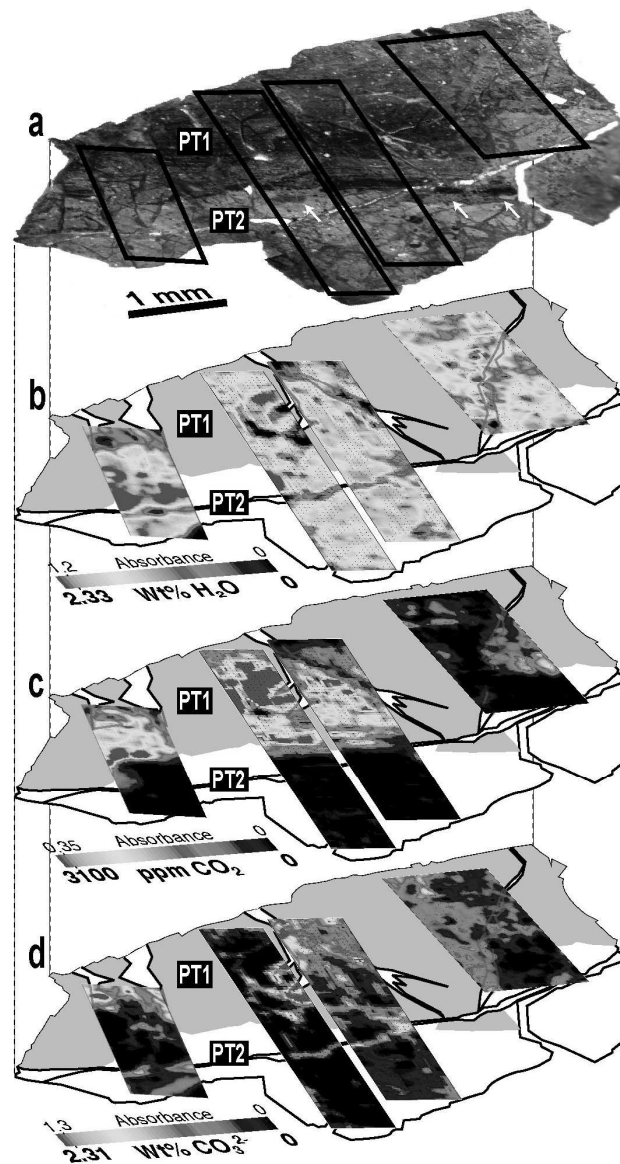
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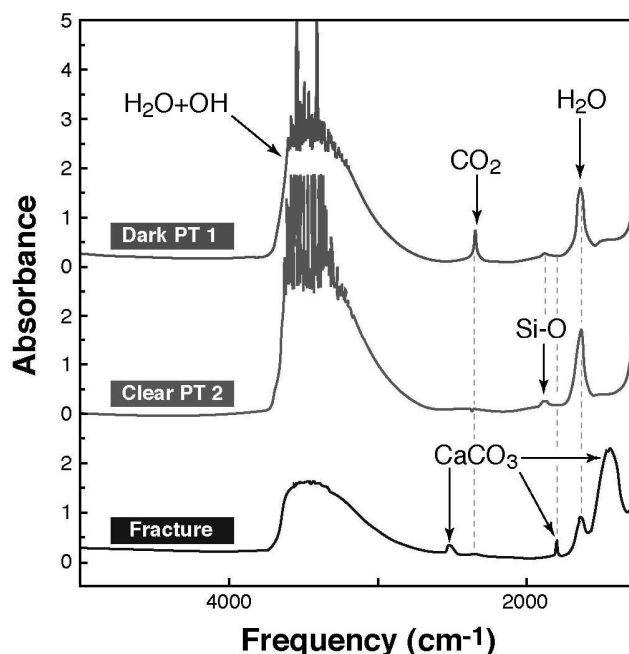
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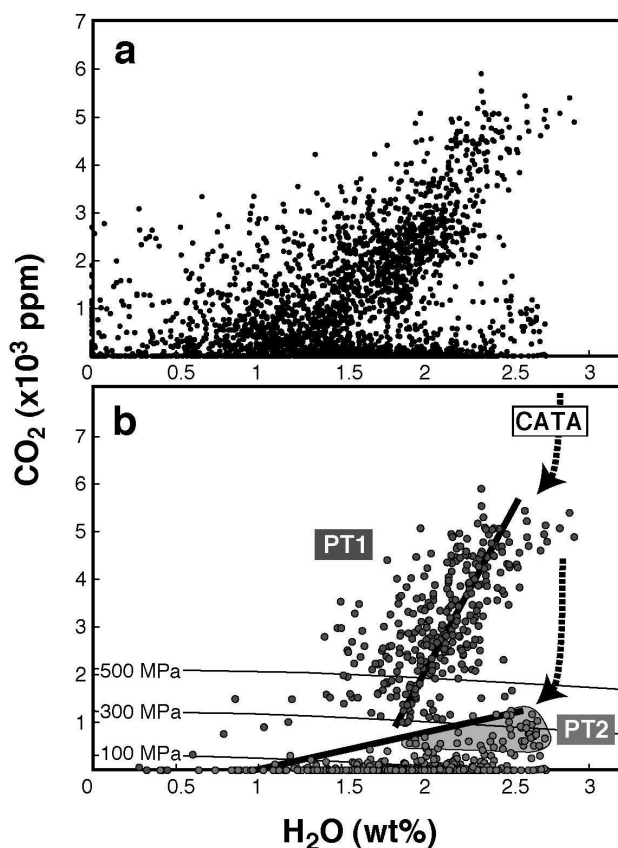
**Figure 1:** Simplified geological map of Awaji Island, modified from [16].



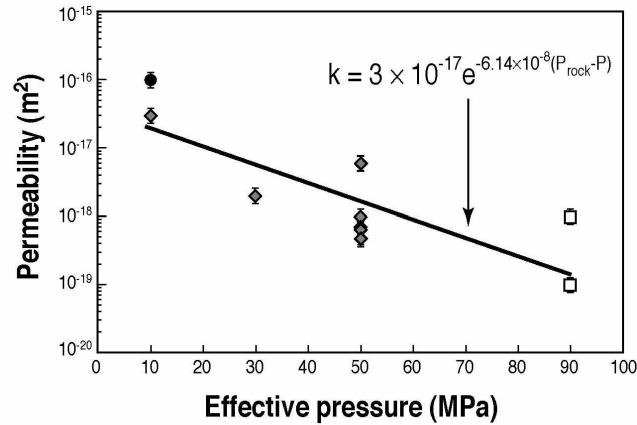
**Figure 2:** FTIR mapping in pseudotachylytes from the Nojima fault. a) Optical microphotograph showing the older dark pseudotachylyte (PT1) re-molten into a younger clear pseudotachylyte (PT2). PT2 contains plastically deformed fragments of PT1 (white arrows) and fewer mineral remnants than PT1. Carbonate veins crosscut the two generations of friction melts. b-d) Peak height (absorbance) mapping of molecular H<sub>2</sub>O (1630 cm<sup>-1</sup>), molecular CO<sub>2</sub> (2340 cm<sup>-1</sup>) and CO<sub>3</sub><sup>2-</sup> (1460 cm<sup>-1</sup>) converted into concentrations (wt%).



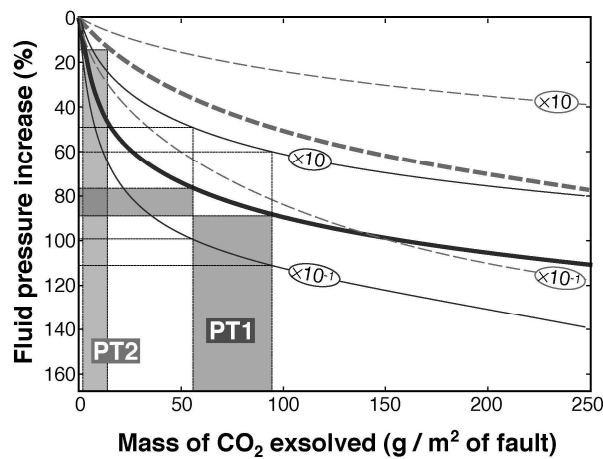
**Figure 3:** Typical FTIR spectra of pseudotachylytes from the sample cored in the Nojima fault. Upper (red) spectrum: dark older pseudotachylyte (PT1). Middle (green) spectrum: clear younger pseudotachylyte (PT2). Lower (blue) spectrum: fracture. There is virtually no dissolved hydroxyl in the glass (the absorption peak of bound OH is at  $4500\text{ cm}^{-1}$ ). The absorption peak of total OH (corresponding to the O-H stretching vibration at  $3450\text{ cm}^{-1}$  in hydroxyl and water molecules) is saturated (and useless) in the glasses due to the high water concentration, but not in the fractured areas. The absorption peak of molecular  $\text{CO}_2$  (recognized by its stretching vibration at  $2340\text{ cm}^{-1}$ ) is observed in the dark pseudotachylyte, but not in the clear one. Molecular  $\text{H}_2\text{O}$  (recognized by its bending vibration at  $1630\text{ cm}^{-1}$ ) displays a higher absorption peak and is therefore more abundant in the pure glass than in the fractures. The three absorption bands of calcite (at  $2510$ ,  $1790$  and  $1460\text{ cm}^{-1}$ ) are observed in fractured zones. A combination of Si-O vibrations (at  $1870\text{ cm}^{-1}$ ) is detected in pure glass.



**Figure 4:** H<sub>2</sub>O versus CO<sub>2</sub> concentrations measured by FTIR in the pseudotachylytes. a) Complete dataset from the 3420 spectra. b) Dataset after filtering: spectra showing carbonate peaks (2510, 1790 and 1460 cm<sup>-1</sup>) were removed in order to consider only pure glass without calcite. The isobars have been calculated using the VolatileCalc software [30]. Pseudotachylytes first degas as an open system (thick dashed arrows). Because of melt cooling, the pseudotachylytes eventually enter into a second step of closed-system degassing (solid lines). The degassing trend of PT1 (red dots) is consistent with an exsolved fluid phase at 80 mol% H<sub>2</sub>O, 20 mol% CO<sub>2</sub> (upper solid line drawn by linear regression). PT2 (green dots) degassed a more water-rich mixture ( $\geq 97$  mol% H<sub>2</sub>O;  $\leq 3$  mol% CO<sub>2</sub>, domain delimited by the lower solid line). The shaded area corresponds to the border of PT1, which is affected by incomplete re-melting and has a transitional CO<sub>2</sub> content between fresh PT1 and final PT2 (Fig. 2c).



**Figure 5:** Best fit log-linear equation for permeability in the Nojima fault core with increasing effective pressure. Black dots compiled from [41], grey diamonds from [42], and empty squares from [43].



**Figure 6:** Fluid pressure increase ( $P_f/P_0$ ) caused by  $\text{CO}_2$  exsolution at two different depths, assuming a hydrostatic initial fluid pressure, and without considering thermal pressurization. Plain (blue) curves: 11 km, 400 °C. Dashed (orange) curves: 5 km, 200 °C. Each color set includes three curves: One curve calculated using the permeability law defined by equation 4 (Fig. 5), one curve using a permeability ten times larger ( $\times 10$ ), and one curve using a permeability ten times smaller ( $\times 10^{-1}$ ). The red and green areas correspond to the  $\text{CO}_2$  production caused by PT1 (56 – 95  $\text{g/m}^2$ ) and PT2 (1.4 – 14  $\text{g/m}^2$ ), respectively, on 1  $\text{m}^2$  of fault plane. Calculation detailed in text.

**Table caption**

**Table 1:** Abbreviations and parameter values used in the calculation of the fluid pressure.

$i$	The subscripts $i = 0$ and $i = f$ refer to quantities before and after $\text{CO}_2$ exsolution, respectively
$P_i$	Fluid pressure (Pa)
$P_{\text{rock}}$	Lithostatic load (Pa)
$T$	Temperature (K)
$R$	Gas constant = $8.314472 \text{ J.mol}^{-1}.\text{K}^{-1}$
$Z_i$	Compressibility factor of $\text{CO}_2$ given by the Saxena & Fei EOS [31]
$T^c$	Temperature of the critical point of $\text{CO}_2 = 304.14 \text{ K}$ [44]
$P^c$	Pressure of the critical point of $\text{CO}_2 = 73.8 \times 10^5 \text{ Pa}$ [44]
$M^{\text{CO}_2}$	Molar mass of $\text{CO}_2 = 44 \text{ g.mol}^{-1}$
$m_i$	Mass of fluid $\text{CO}_2$ in contained in a fault zone of thickness $w$ , i.e. in the porosity of $1 \times 1 \times w \text{ m}^3$ of fault rock (g)
$\phi_i$	Porosity of the fault zone
$w$	Maximum distance of $\text{CO}_2$ outflow in the direction perpendicular to the fault plane, within the time span of friction melting (m)
$L_f$	Hydraulic diffusion length scale of $\text{CO}_2$ (m)
$D_f$	Hydraulic diffusivity after exsolution ( $\text{m}^2.\text{s}^{-1}$ )
$\psi$	Characteristic timescale of frictional exsolution set at 5 sec (the time lapse for glass melting and quenching does not exceed a few seconds [32])
$k_i$	Permeability of the fault zone ( $\text{m}^2$ )
$\eta_i$	Viscosity (Pa.s) of $\text{CO}_2$ in the supercritical state (data from [44])
$\beta_i$	Storage capacity of the fault zone ( $\text{Pa}^{-1}$ )
$\beta_i^{\square}$	Compressibility of the bulk fault core ( $\text{Pa}^{-1}$ )
$\beta^s$	Compressibility of the mineral grains assumed to be constant at $1.2 \times 10^{-11} \text{ Pa}^{-1}$
$\beta_i^{\text{CO}_2}$	Compressibility of $\text{CO}_2$ ( $\text{Pa}^{-1}$ )