



Vitrinite reflectance and Raman spectra of carbonaceous material as indicators of frictional heating on faults: Constraints from friction experiments



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ABSTRACT

Vitrinite reflectance (R_o) and Raman spectra of carbonaceous material (RSCM) are both widely used as indicators of the maximum attained temperatures in sedimentary and metamorphic rocks. However, the potential of these methods to estimate temperature increases associated with fault slip has not been closely studied. To examine this issue, friction experiments were conducted on a mixture of powdered clay-rich fault material and carbonaceous material (CM) at slip rates of 0.15 mm/s and 1.3 m/s in nitrogen (N_2) gas with or without distilled water. After the experiments, we measured R_o and RSCM and compared to those in starting material. The results indicate that when fault material suffers rapid heating at $>500^\circ\text{C}$ in ~ 9 s at 1.3 m/s, R_o and the intensity ratio of D1 and D2 Raman bands of CM (I_{D2}/I_{D1}) markedly increase. Comminution with very small temperature rise in ~ 32 min at 0.15 mm/s is responsible for very limited changes in R_o and I_{D2}/I_{D1} . Our results demonstrate that R_o and RSCM could be useful for the detection of frictional heating on faults when the power density is ≥ 0.52 MW/m². However, the conventionally used R_o and RSCM geothermometers are inadequate for the estimation of peak temperature during seismic fault slip. The reaction kinetics incorporating the effects of rapid heating at high slip rates and studies of the original microtexture and composition of CM are required to establish a reliable thermometer for frictional heating on faults.

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1. Introduction

The detection of heat anomalies associated with frictional heating on faults yields valuable information on coseismic shear stress and frictional work during an earthquake (e.g., Kano et al., 2006; Fulton et al., 2013). The extreme example of frictional melting results in the formation of pseudotachylyte and its presence provides unequivocal evidence of past seismic slip (Cowan, 1999). However, pseudotachylyte is an uncommon rock type and it is desirable to have other indicators capable of detecting and preserving evidence for less extreme shear heating. Various indicators of frictional heating have been proposed including changes in vitrinite

reflectance, usually expressed as the quantity R_o (O'Hara, 2004; Sakaguchi et al., 2011), magnetite formation associated with thermal decomposition of siderite (Hirono et al., 2006; Han et al., 2007), changes in the concentration of fluid-mobile trace elements (Ishikawa et al., 2008), stretching of fluid inclusions in calcite (Ujiie et al., 2008), changes in clay mineralogy (Kameda et al., 2011), graphitization of carbonaceous material (CM) (Oohashi et al., 2011; Kuo et al., 2013), thermal decomposition of carbonate (Collettini et al., 2013), and biomarker thermal maturity (Savage et al., 2014). However, these indices are commonly associated with considerable uncertainties in estimates of the associated temperature rise.

Systematic changes in the molecular structure and geochemistry of CM including vitrinite have been well documented for increases in temperature on geological time scales associated with burial (e.g., Aihara, 1989; Beyssac et al., 2002). Thermal maturation of CM is an irreversible reaction and is dependent on both duration of heating and maximum temperature. Two widely

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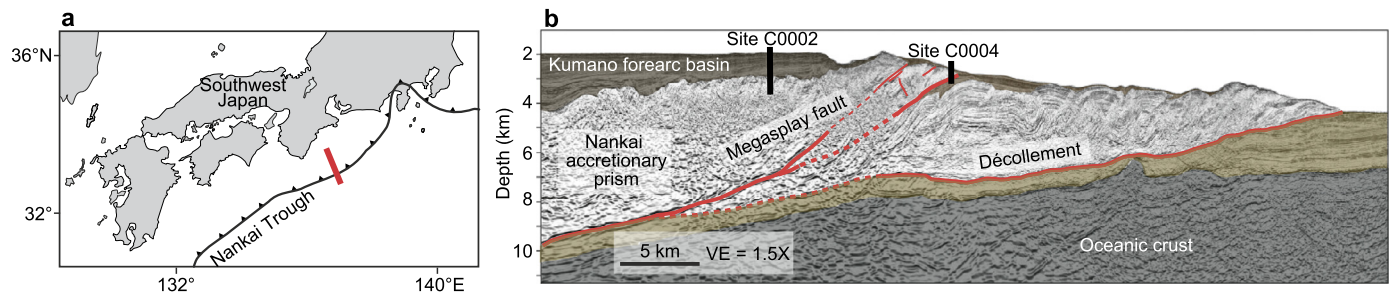


Fig. 1. The Nankai accretionary prism offshore southwest Japan. (a) Location map of the Nankai Trough. The red line indicates the location of the seismic reflection line in Fig. 1b. (b) Seismic reflection profile of the Nankai accretionary margin showing drilled sites C0002 and C0004 at the Kumano forearc basin and the shallow portions of a megasplay fault, respectively (modified from Moore et al., 2009). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

applied thermometers for CM are based on vitrinite reflectance (R_0) and the Raman spectra of CM grains and these methods have been used to estimate maximum temperature experienced by sedimentary and metamorphic rocks (e.g., Barker and Goldstein, 1990; Sweeney and Burnham, 1990; Beyssac et al., 2002; Aoya et al., 2010; Kouketsu et al., 2014). R_0 has also been applied for the detection of increased heating on faults (O'Hara, 2004; Sakaguchi et al., 2011). However, the estimation of the peak temperature during frictional heating on faults from R_0 is questionable because it is uncertain whether the existing kinetic model can also be applied for a short-lived thermal event such as frictional heating (Hirono et al., 2009; Fulton and Harris, 2012; Kitamura et al., 2012). In contrast to R_0 , no attempts have been made to assess the utility of the Raman spectra of CM (RSCM) for detecting frictional heating.

Previous friction experiments on CM were performed under air conditions or a mixture of 90 wt% quartz powder and 10 wt% CM (O'Hara et al., 2006; Kitamura et al., 2012). However, friction experiments at high slip rates are likely to be associated with a large rise in temperature, and CM in samples that are exposed to air is likely to become oxidized resulting in the formation of CO_2 gas and not to be a good analogue for the behavior of faults. In addition, the previously employed mixture of quartz powder and CM is unlikely to be representative of natural fault rocks. Our experiments were performed on a mixture of 95 wt% clay-rich fault material and 5 wt% CM at slip rates of 0.15 mm/s and 1.3 m/s in nitrogen (N_2) gas with or without distilled water. We consider these conditions to be more appropriate to investigate the effects of rapid shear heating on the maturation of CM in faults. After the experiments, we examined the resultant microstructures, the temperature evolution during shearing and compared grain size, R_0 , and RSCM before and after shearing.

2. Methods

2.1. Friction experiments

The fault sample for the friction experiments was the same as that used by Ujiie and Tsutsumi (2010), which is obtained from microbrecciated hemipelagic mudstone immediately above the 10-mm-thick fault gouge in the shallow portion (271 m below sea floor) of the megasplay fault at site C0004 in the Nankai accretionary prism, southwest Japan (Fig. 1). The mineralogy of the fault sample determined from bulk powder X-ray diffraction indicates that the sample consists of clay minerals, quartz, plagioclase, and calcite in the proportions 59.8, 19.6, 17.9 and 2.7%, respectively. The clays are a mixture of smectite, illite, and chlorite. The CM was collected from the Kumano forearc basin sediments at site C0002 (Fig. 1b). The CM is dominated by very low-grade brown coal.

Friction experiments were conducted using the rotary shear, low- to high-velocity frictional apparatus at Kyoto University. The clay-rich fault sample was powdered using a pestle and sieved to a grain size of ≤ 0.12 mm. The powdered gouge composed of a mixture of 95 wt% sieved clay-rich fault sample and 5 wt% CM was used for experiments. To avoid oxidization of CM associated with temperature rise due to frictional heat, the apparatus was equipped with an acrylic vessel that has an inlet for N_2 gas (Fig. 2a). The vessel was charged with N_2 before the experiments, and the gouge was sheared under an N_2 atmosphere. The weight of the gouge was 0.5 g. For one of the experiments, we added 0.55 ml of distilled water to simulate water-saturated conditions. Hereafter we use the terms wet and dry tests to indicate gouge experiments with or without distilled water, respectively. The gouge was placed between a pair of solid cylinders of granite with a diameter of 25 mm and was surrounded by a hollow-cylindrical Teflon sleeve to prevent gouge leaking during rotary shearing (Fig. 2b). The assembly of granite specimen–gouge–Teflon sleeve was set in the apparatus in which the upper cylinder remained stationary while the lower one was rotated. Since the slip rate increases from the center to the edge of the solid-cylindrical specimens, we used an equivalent slip rate (V_e), which is defined such that V_e multiplied by the sliding surface area (S) and the shear stress (τ) gives the rate of frictional work on S , assuming that τ is constant over the sliding surface (Shimamoto and Tsutsumi, 1994). In this paper, we refer to V_e as the slip rate. Dry and wet tests were conducted at a slip rate of 1.3 m/s and a normal stress (σ_n) of 2.0 MPa with displacement of ~ 11 m. In addition, to examine the effects of compaction with very small temperature rise on maturation of CM, one dry test was performed at a low slip rate of 0.15 mm/s and low σ_n of 1.0 MPa with displacement of ~ 0.3 m. Before shearing, the sample assembly was consolidated until the axial shortening became stable (~ 30 min) at the same σ_n as that during shearing. The initial thickness of the gouge layer is 0.8 mm. After shearing, the sample assembly was cut through the axis of the solid-cylinder to make a radial thin section that was perpendicular to the shear direction and gouge layer (Fig. 2b). We also made a radial thin section from the sample assembly after 30 min consolidation at σ_n of 1.0 MPa and refer to the gouge formed after this compaction as the starting material.

We calculated temperature changes during experiments using the finite-element method (Kuroda, 2001), which considers an axisymmetric two-dimensional system. We assume that all of the frictional work is converted into heat production in the gouge domain, τ is constant over the sliding surface, and temperature changes are caused by heat production due to friction and heat loss due to conduction (e.g., Ujiie and Tsutsumi, 2010). The heat production term for a given radial position is found by multiplying τ by the slip rate appropriate for a particular radial position. The calculation of the heat production used the thickness of the gouge after the experiments. The initial temperature in a section of

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