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Blackening of fault gouge by comminution and pyrolysis of carbonaceous materials during earthquake slip

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ABSTRACT

Black fault gouges sometimes develop, mainly in sedimentary rocks, but the cause of the color transformation is not well understood. Here we demonstrated the blackening of synthetic mixtures of montmorillonite and bituminous coal and of montmorillonite and magnetite in milling, heating, and friction experiments. Mixed samples with a higher volume fraction of coal or magnetite before the experiments showed lower L* values (lightness index; lower values indicate darker blacks), because coal and magnetite are intrinsically black. The milling and heating experiments showed that the L* values of mixed samples of montmorillonite and coal drastically decreased with longer milling times and higher temperatures. The L* values of mixed samples of montmorillonite and magnetite also decreased with longer milling times, but no notable change was observed in the samples after the heating experiments. Because comminution by milling induces granulation of the constituent materials, blackening of the experimental samples was primarily caused by dispersal through the sample of fine black particles such as coal and magnetite, but it could be strengthened by adsorption onto host particles of organic gases produced by pyrolysis of carbonaceous material at high temperature. The friction experiment with mixed samples of montmorillonite and coal produced the remarkably low L* values. Friction induces both comminution and heating of samples, so the blackening could be greater than after either milling or heating alone. Therefore, relatively black fault gouges, compared with the surrounding host rocks, might have experienced comminution and heating, probably related to earthquake slip. Thus, black coloration could be one of the important information on fieldwork.

1. Introduction

Fault gouges often differ in color from the surrounding host rock, and when the host rock is of sedimentary origin, relatively black fault gouges are often observed (e.g., Balsamo et al., 2014; Buatier et al., 2012; Manatschal, 1999; Tsutsumi et al., 2004; Yao et al., 2013). The main damage zone of the Chelungpu fault, which slipped during the 1999 Taiwan Chi-Chi earthquake, is very black (Hirono et al., 2006, 2007a). High magnetic susceptibility in this zone has been attributed to the new formation of magnetic minerals such as magnetite and maghemite at high temperatures induced by friction (Chou et al., 2012; Hirono et al., 2006; Mishima et al., 2009). Tanikawa et al. (2008) experimentally reproduced the blackening of fault gouge by subjecting a host-rock sample to high-velocity (on the order of 1 m s^{-1}) friction, and they suggested that the color change resulted from the new formation of black magnetic minerals. However, the small amount of magnetic minerals (<1 wt.%) in the black gouge of the Chelungpu fault zone (Hirono et al., 2014) would be unlikely to affect the color of the bulk rock.

Because determination of the peak temperature reached during slip is crucial for identifying the faulting mechanism of an earthquake, various temperature proxies have been proposed, for example, magnetite formation at high temperature (≥400 °C) (Chou et al., 2012; Mishima et al., 2009); anomalies in the concentrations of fluid-mobile trace elements (Sr, Cs, Rb, and Li) and in the Sr isotope ratios, indicating the presence of high-temperature (≥350 °C) fluid (Ishikawa et al., 2008); dehydroxylation of clay minerals (Hirono et al., 2008); thermal decomposition of carbonate minerals (Han et al., 2007; Hirono et al., 2007b); and thermal maturation of carbonaceous material, as indicated by vitrinite reflectance (Maekawa et al., 2014; O'Hara et al., 2006; Sakaguchi et al., 2007) and by infrared and Raman spectroscopy (Hirono et al., 2015). However, these proxies require sophisticated laboratory analyses; an easy method for preliminary detection of a record of high temperature in the field is needed.

In this study, we used a high-velocity friction apparatus, a thermogravimetric (TG)–differential scanning calorimeter (DSC), and a milling machine to produce blackening in synthetic fault samples, and we used color digital spectrophotometry and pyrolysis–gas chromatography–

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mass spectrometry (py-GC/MS) to evaluate color changes and the organic chemical properties of the samples. We then considered how comminution and pyrolysis of carbonaceous materials contributed to the blackening.

2. Methods

2.1. Samples

As analogues of fault gouge components, we used Na-rich montmorillonite (Crook County, Wyoming, USA; SWy-2 supplied from The Clay Minerals Society) and bituminous coal (Ashibetsu, Hokkaido, Japan). We also used magnetite (Diamantina, Minas Gerais, Brazil), because the color change in the Chelungpu fault was ascribed to black magnetic minerals (Tanikawa et al., 2008). Their reported grain densities are 2.8, 1.3, and 5.2 g cm^{-3}, respectively (Holmboe et al., 2012; Meyers, 1982; Schön, 1996). We investigated the process and mechanism of the color transition caused by the effects of high-velocity friction, milling, and heating on montmorillonite samples and on mixtures of montmorillonite and coal in three series of experiments. We also examined color changes in mixtures of montmorillonite and magnetite in milling and heating experiments. All samples were dried at 50 °C for several hours just before each experiment.

Before the experiments, we characterized the Ashibetsu bituminous coal thermochemically by using a TG-DSC apparatus (STA 449 C Jupiter balance, Netzsch) to heat the coal to 800 °C at a rate of 10 °C min^{-1} under an Ar atmosphere; the resultant TG profile indicated that a weight loss was started at approximately 150 °C and a large amount of weight loss occurred above approximately 400 °C (Fig. 1).

We analyzed the bituminous coal on the molecular level by py-GC/MS, using a pyrolyzer (Py-3030D, Frontier Lab; furnace temperature 700 °C, interface temperature 330 °C) directly coupled to a gas chromatograph–mass spectrometer (GCMS-QP2010 Ultra, Shimadzu) equipped with a DB-5MS column (length 30 m, inner diameter 0.25 mm, film thickness 0.25 μm; Agilent J&W). Approximately 1 mg of the coal sample was heated from 100 to 330 °C at a rate of 50 °C min^{-1} under vacuum, and the gas-phase pyrolysates that were collected in a cryotrap were analyzed by GC/MS. In the GC/MS analysis, helium was used as the carrier gas at a flow rate of 1 mL min^{-1}, and the split ratio was 50. The injection temperature was 300 °C, and the column temperature program was as follows: hold at 40 °C for 2 min, heat to 310 °C at 15 °C min^{-1}, and hold at 310 °C for 10 min. The mass spectrometer was operated in the electron ionization mode (70 eV, scan range m/z 35–600, scan interval 0.2 s), and the ion source temperature was kept at 230 °C. The pyrolysates were identified by comparing their mass spectra with the reported spectra of standards in the library of the National Institute of Standards and Technology. The resultant pyrogram of the Ashibetsu bituminous coal showed abundant naphthalene and normal chain aliphatic hydrocarbons (Fig. 2).

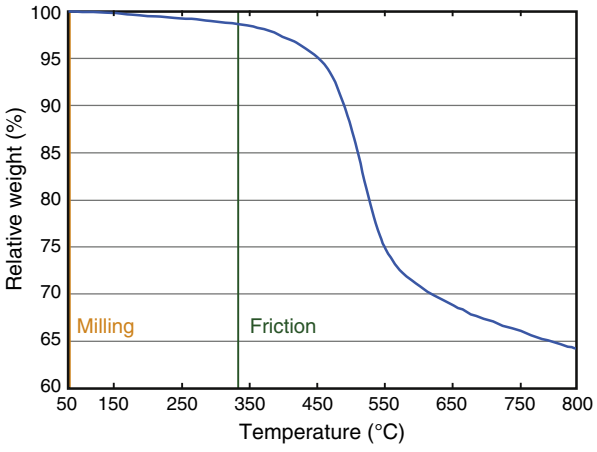

Fig. 1. Thermogravimetric curve of the Ashibetsu bituminous coal. Maximum temperatures reached during the friction and milling experiments were approximately 330 and 40 °C, respectively.

2.2. Experiments

2.2.1. Mixing procedure

Before the friction, milling, and heating experiments, we examined the color differences due to different initial concentrations of coal and magnetite particles. Mixed samples of 99.99 wt.% montmorillonite + 0.01 wt.% coal, 99.9 wt.% montmorillonite + 0.1 wt.% coal, 99 wt.% montmorillonite + 1 wt.% coal, 97.3 wt.% montmorillonite + 2.7 wt.% coal, 90 wt.% montmorillonite + 10 wt.% coal, 99.99 wt.% montmorillonite + 0.01 wt.% magnetite, 99.9 wt.% montmorillonite + 0.1 wt.% magnetite, 99 wt.% montmorillonite + 1 wt.% magnetite, 96.1 wt.% montmorillonite + 3.9 wt.% magnetite, 90 wt.% montmorillonite + 10 wt.% magnetite, and 69.2 wt.% montmorillonite + 30.8 wt.% magnetite were prepared (Table 1). The samples were manually mixed in an agate mortar.

2.2.2. Friction experiment

For the friction experiments we used a high-velocity rotary shear apparatus to examine the effects of mechanical damage and frictional heat on synthetic fault samples (Fig. 3a). In this series of experiments, we placed approximately 500 mg of dry powdered sample, consisting of (1) montmorillonite (100 wt.%), (2) montmorillonite (99.9 wt.%) + coal (0.1 wt.%), or (3) montmorillonite (99 wt.%) + coal (1 wt.%), between the ends of two gabbro cylinders (25-mm diameter) and then sealed the cylinders within a polytetrafluoroethylene (Teflon) sleeve to prevent leaks (Fig. 3b). Because in the Chelungpu fault example the concentration of carbonaceous material in the black gouge and the surrounding rocks was reported to be approximately 0.27 ± 0.09 wt.% (Ikehara et al., 2007), we adopted coal concentrations of 0.1 and 1 wt.% for our friction experiments. The experiments were carried out at a velocity of 1 m s^{-1} with 10 m of displacement under 1 MPa of normal stress under a dry Ar atmosphere; the resultant friction data are shown in Fig. A.1. The temperature distribution within the sample during the experiments was estimated by using an axisymmetric 2-D model based on the finite-element method, in which the highest temperature was approximately 330 °C (Fig. A.2). After the friction experiments, damaged powder from the outer 6.0–12.5 mm of the sample was collected for analysis.

2.2.3. Milling experiment

For the comminution experiments, a laboratory planetary mill (Pulverisette 6, Fritsch) was used to grind samples for 1, 2, 4, or 6 h at 600 rpm. Grinding was suspended for 5 min after each 20 min of milling to prevent the sample temperature from rising; thus, the temperature at the end of each experiment, measured by using a needle probe thermometer, was only slightly higher than room temperature (approximately 40 °C). In this series of experiments, we used (1) mixed samples of 99 wt.% montmorillonite + 1 wt.% coal, (2) 99 wt.% montmorillonite + 1 wt.% magnetite, (3) 96.1 wt.% montmorillonite + 3.9 wt.% magnetite, and (4) samples consisting only of montmorillonite (100 wt.%).

2.2.4. Heating experiment

For the heating experiments, we used the TG-DSC apparatus to heat (1) mixed samples of 99 wt.% montmorillonite + 1 wt.% coal, (2) 99 wt.% montmorillonite + 1 wt.% magnetite, (3) 96.1 wt.% montmorillonite + 3.9 wt.% magnetite, and (4) samples consisting only of montmorillonite (100 wt.%). Approximately 30 mg of sample was placed in a covered $Pt_{90}Rh_{10}$ crucible and heated from room temperature to a target temperature of 200, 400, or 600 °C at a rate of 20 °C min^{-1} under a flow of Ar gas (50 mL min^{-1}).