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Identification and tectonic implications of nano-particle quartz (<50 nm) by synchrotron X-ray diffraction in the Chelungpu fault gouge, Taiwan

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1. Introduction

ABSTRACT

To determine the ultrafine nano-scale grains in a fault gouge; the Chelungpu fault gouge was sampled and analyzed. It was separated into different particle size ranges and was analyzed by synchrotron X-ray diffraction and transmission electron microscopy. The minerals of gouge are predominantly composed of quartz, plagioclase, smectite, illite, chlorite, and kaolinite. The mineral association of <100 nm particles are quartz, smectite, and illite. However, only smectite and illite are included except quartz in the 1-to-25 nm fractions. We propose that quartz is the index mineral associated with co-seismic fracture and the possible limit grain size is ~25 nm on fracturing. The smectite and illite nano-particles may be associated with weathering process of gouge at shallow or surface conditions.

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Earthquake is the most unpredictable and ruinous hazard in the world. In recent years, there were many disastrous earthquake events, such as 1995 Kobe earthquake, 1999 Chi-Chi earthquake, 2004 Sumatra earthquake, 2008 Wenchuan earthquake, 2011 Christchurch earthquake, 2011 Tohoku earthquake, etc., which caused many casualties and property loss. For studying the earthquake mechanism, gouge is a key to investigate the physical and chemical process of fault slipping. In principle, characterization of slip zones may be relevant for assessing physical processes such as frictional heating, thermal pressurization, fracture energy, redox conditions, etc.

The total energy releases of fault zone during an earthquake are still unknown clearly (Heaton, 1990; Kanamori, 1994; Kanamori and Heaton, 2000). Fault slip-weakening models have been proposed that provide a basis for relating the energy budget to physical processes of earthquake. The energy loss of earthquake slipping exhibits in three forms, radiated energy, frictional heat, and fracture energy (Beeler et al., 2003; Kanamori and Heaton, 2000; Venkataraman and Kanamori, 2004). Fracture energy calculation has been reported from





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seismological and experimental rock deformation data (Abercrombie and Rice, 2005; Guatteri et al., 2001; Okubo and Dieterich, 1984; Rice et al., 2005; Wong, 1982). During a fault slipping, gouge particles will be milling into smaller fractions to the nanometer scale (Wilson et al., 2005). In the past, numerous studies reported analysis of particle size distribution within ultrafine gouge to calculate total grain surface area by optical and electron microscopies, then to estimate the fracture energy associated with gouge formation (Chester et al., 2005; Ma et al., 2006; Wilson et al., 2005). Chester et al. (2005) observed the finest particle size being 1.6 nm within the Punchbowl fault gouge as the lower cut-off for fracture energy estimation. Ma et al. (2006) observed the gouge in the fault zone of Taiwan Chelungpu-fault Drilling Project (TCDP) at a depth of about 1 km. They used grain sizes larger than 50 nm as lower cut-off to estimate fracture energy, because the transmission electron microscope (TEM) image of grain sizes less than 50 nm shows rounded shapes. Therefore, they considered that those grains < 50 nm might be formed by chemical precipitation rather than fracturing. However, the TEM image of Chester et al. (2005) also shows rounded shapes in their ultrafine grains for calculation; the image by Ma et al. (2006) shows very fine grains around several nm in diameter, which was not considered for fracture energy. In addition, the very fine grains have larger surface area than coarse grains and could provide high percentage of total fracture energy significantly.

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Thus, determining the lower cut-off grain sizes is important for fracture energy calculation.

In order to understand the possible limit on the fracturing during faulting within ultrafine gouge, analysis of the finest mineral composition and grain size distribution are requisites. Nevertheless, nanoparticles generally aggregate to each other quickly or cling to large grain surface because of their surface reactivity at the nanometer scale and their relatively large surface area. Hence, how to collect nanoparticles in natural materials with high efficiency and in large quantities for analyses is a challenge. Tsao et al. (2009a,b) developed an automated ultrafilteration device (AUD) for high efficient collection of nanoparticles, which had been proved to be more efficient than the conventional ultra-centrifugation and syringe filtration methods. This device provides us a solution to nano-particle collection from ultrafine gouge for further analysis.

Here we report the mineral identification and microscope observation of nano-particles in fault gouge sample from Chelungpu fault branch of the 1999 Chi-Chi earthquake surface rupture. First, we separate different particle sizes ranging from 50 µm to 1 nm by centrifugation and AUD. Second, we focus on the mineralogy and appearance by using synchrotron X-ray diffraction (XRD) and TEM with energy dispersive X-ray spectrometer (EDX). Lastly, we propose that quartz is the index mineral associated with gouge formation and the low cut-off grain size is ~25 nm. The smectite and illite nano-particles could be associated with weather process of gouge at outcrop.

2. Geological setting and sampling

Taiwan is located at the boundary between the Eurasian Plate and the Philippine Sea Plate. The Philippine Sea Plate is moving toward W54°N at a speed around 80 mm per year (Yu et al., 1997) and is impacting with the Eurasian continental margin (Fig. 1A). This substantial collision and uplifting between two subduction systems result in active faulting, enhance the development of numerous earthquakes and crustal deformation within Taiwan Island. Taiwan is generally subdivided into some geological regions, from west to east; they are the Coastal Plain, the Western Foothills, the Central Range, the Longitudinal Valley, and the Coastal Range (Ho, 1986). The most active region within Taiwan is along the belt between the Coastal Plain and the Western Foothills. There are several fault zones around this active seismic region, include Shuilikeng, Shuangtung, Chelungpu, and Changhua faults.

The Chelungpu fault zone was described as the Chinshui Formation over-thrusting the Toukashan Formation in the southern Taichung Basin (Chang, 1971). This fault is a thrust fault of over 85 km in length and is a main boundary between the Plio-Pleistocence fold-and-thrust belt to the east and late Quaternary basin to the west. The fault is reasoned to have formed at the beginning of the Mid-Pleistocene, 0.7–0.5 Ma (Chen et al., 2001).

The Chi-Chi earthquake (M_w 7.6) took place to the central Taiwan on 21 September 1999. The hypocenter is located near Chi-Chi town (120.81° E, 23.86° N, depth ~10 km, Kao and Chen, 2000; Ma et al., 1999). The surface rupture was along the Chelungpu fault zone at about 85 km in length with large surface deformation (Angelier et al., 2003; Lee et al., 2002; Ma et al., 1999). The surface deformation provides us a good opportunity to sample an outcrop fault gouge of recent earthquake.

Our gouge sample was caught from the Chelungpu fault branch of the 1999 Chi-Chi earthquake surface rupture that passed through Wu-Feng downtown in center Taiwan (Fig. 1A, B). The hanging wall raised a height about 2.5 m in the campus of Kuang-Fu Junior High School during the 1999 Chi-Chi earthquake. The sample investigated in this study was caught in 2005, from the outcrop of Chi-Chi fault gouge, which is located at the riverbed behind the campus. The ~2.5 m height fault rupture which is across the river was eroded by water 5 years after the Chi-Chi earthquake. The fault black gouge zone was found nearby the riverbed and is up to 15 cm wide and 60° in dip within the Chinshui Formation (siltstone) (Fig. 1C). The sample is black with fine grain (Fig. 1D).

3. Methods

3.1. Separation of fault particles in different particle sizes

The sample was shattered into powder and put in a room to air dry for 2 weeks. After, the gouge powder was passed through a 300-mesh sieve (aperture 50 μ m) to remove coarse fraction (>50 μ m). Then the <50 µm size fraction (about 100 mg) was removed organic matter by H_2O_2 under 70 °C. Next, the <50 μ m size fraction was divided into <2 µm size fracture by sedimentation according to Stokes' Law (Jackson, 2005; Tanner and Jackson, 1947; Williams et al., 1958). The <2 µm size fraction was separated into different ranges by centrifugation. The centrifugation time required to separate this fraction into the size range was calculated by the modified Stokes' equation ($\sqrt[d]{\frac{18\eta \ln (R_2/R_1)}{(\rho_s - \rho_{\omega})\omega^2 t}}$) (Laidlaw and Steinmetz, 2005; McFadyen and Fairhurst, 1993; Puretz, 1979; Ross and Morrison, 1988), where d is the particle diameter (cm), $\boldsymbol{\eta}$ is the viscosity coefficient of the liquid (g cm⁻¹ s⁻¹), ρ_{ω} is the density of the liquid (g cm⁻³), ρ_s is the particle density (g cm⁻³), R_1 is the radius from the centrifugal center to the sample height inside the tube (cm), R_2 is the radius from the centrifugal center to the sample bottom inside the tube (cm), ω is angular velocity (rpm), and *t* is the ultra-centrifugation time (sec). The fault gouge density was measured by pycnometer method; the average gouge density is 2.58 g cm^{-3} .

The centrifugal steps was done using a Hitachi CR21 refrigerated centrifuge (Hitachi High-Technologies Corp., Tokyo, Japan), which had a R12A3 rotor with polycarbonate tubes (250 ml × 6) and settling sample height of 10 cm. For collecting the 450-to-2000 nm size fractions, the suspension (<2000 nm size fractions) was centrifuged at 980 ×g (3370 rpm) for 5 min by ultra-centrifugation at 4 °C. The settled particles were re-suspended in double deionized water (DDW) and were using ultrasonic dispersion at 170 W and 60 kHz for 1 min by NEY 300 Ultrasonic. The dispersed suspension was then repeatedly centrifuged and washed ten times by the same centrifugation and dispersion methods to rarefy the 450-to-2000 nm size fractions. We used the same process with different centrifugation speeds and time for separating different particle size ranges (100-to-450 and 50-to-100) by higher centrifugation velocity and time.

To collect the 25-to-50 nm fractions, <50 nm suspension was filtered by the AUD using 25 nm pore size Millipore membrane filter to separate the <25 nm size fraction. After, the <25 nm suspension was filtered by the AUD using the Sigma ultrafiltration disk membrane (NMWL: 1000 Da-equivalent to 1 nm in diameter) to collect the size of 1-to-25 nm fractions.

The flow chart of separation methods, centrifuge velocity, and time for different particle size fracture collections is shown in Fig. 2.

3.2. XRD and TEM analysis

The synchrotron XRD could provide higher X-ray counts and a small size X-ray beam for analyzing several milligrams of powder samples. The synchrotron powder XRD patterns were analyzed by a Wiggler Beamline BL17A W20 XRD with $\lambda = 1.334431$ Å at the National Synchrotron Radiation Research Center (NSRRC), Taiwan. It was operated at 1.5 GeV with a ring current of 300 mA using the top-up injection mode. The suspension was treated by freeze-drying for collecting powder (non-orientated) for synchrotron XRD measurement. For observing the occurrence of the particle, <100 nm fractures images were taken by using a FEI Tecnai G2 T20 TEM with energy dispersive X-ray spectrometer (EDX), which was operated with accelerating voltage at 200 keV in the Department of Materials Science and Engineering, National Taiwan University.

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