

Water-rock interaction observed in the brittle-plastic transition zone

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Rock alteration and geochemistry of the fault rocks are examined to infer the characteristics of the fluid phase related to the ancient fault activity. The Hatagawa Fault Zone, northeast Japan, is an exhumed seismogenic zone which is characterized by close association of brittlely and plastically deformed fault rocks mostly derived from Cretaceous granitoids. Epidote and chlorite are dominant alteration minerals in both rocks. However, calcite is characteristically developed in the cataclastic part only. Decrease in oxygen isotope ratio and existence of epidote and chlorite, even in weakly deformed granodiorite, is evidence of water-rock interaction. The water/rock ratio is interpreted to be relatively small and fluid chemistry is buffered by host rock chemistry in the mylonite. The occurrence of calcite in brittle structures is explained by changes in water chemistry during shear zone evolution. CO₂-rich fluid was probably introduced during cataclastic deformation and increased CO₂ concentration resulted in precipitation of calcite.

1. Introduction

The presence of a fluid phase affects fault activity both mechanically and chemically (e.g., Hickman *et al.*, 1995). An increase in pore pressure decreases effective stress on the fault plane to make fault slip easier. Water-related species in crystalline defects decrease the strength of minerals such as olivine and quartz substantially (e.g., Mackwell *et al.*, 1985). Chemical alteration, including dissolution of primary minerals and precipitation of secondary minerals changes the mechanical properties of rocks considerably (e.g., Stünitz and FitzGerald, 1993). Understanding the fluid chemistry is essential for evaluating the effect of water-rock interaction on the fault activity. However, the nature of the fluid phase in the middle to lower crust, the important part for the generation of earthquake in the continental crust, is poorly understood. We have been studying the Hatagawa Fault Zone (Fig. 1) as an ancient seismogenic zone from geological and geochemical aspects to clarify the physical and chemical processes in the seismogenic zone. General geology and description of fault rocks are reported in Shigematsu and Yamagishi (2002) and Tomita *et al.* (2002). In this paper, we examine rock alteration to infer the characteristics of the fluid phase related to the ancient fault activity.

2. Outline of the Hatagawa Fault Zone

The Hatagawa Fault Zone is located in the eastern part of the Abukuma Mountains, NE Japan. It extends in a NNW-SSE direction for up to 100 km. It is a tectonic boundary

between Abukuma belt in the west and South Kitakami belt in the east (Kubo *et al.*, 1990). Various kinds of fault rocks derived mostly from Cretaceous granitoids occur in the fault zone. The granitoids intruded in the relatively shallow level (5 to 10 km depth) judging from the existence of cordierite in a contact aureole (Kubo *et al.*, 1990). Distributions of fault rocks are shown in Fig. 1. The main cataclasite zone, which is considered to be the core of the Hatagawa Fault Zone, extends continuously in an N-S direction and has a width of about 100 m (Watanabe *et al.*, 1953; Tomita *et al.*, 2002). Mylonite zones with a sinistral sense of shear partially surround the cataclasite zone and have a maximum width of 1 km (Shigematsu and Yamagishi, 2002). Small-scale shear zones, with widths ranging from a few mm to a few meters, are distributed in the surrounding granitoids (Shigematsu, 1999; Shigematsu and Tanaka, 2000; Takagi *et al.*, 2000). Deformation structure is well preserved in these small shear zones and pseudotachylyte bands sometimes occur (Kubo and Takagi, 1997). Plastic deformation and brittle deformation are often closely associated in the shear zones (Takagi *et al.*, 2000; Shigematsu *et al.*, submitted). Shigematsu and Yamagishi (2002) categorized quartz microstructure in the mylonites in the Hatagawa Fault Zone into two groups; microstructure A characterized by core and mantle structures and microstructure B characterized by sutured grain boundaries, smaller aspect ratios and less striking undulatory extinction comparing to A. The deformation conditions are estimated to be 260 to 310°C for the mylonite with microstructure A and 310 to 450°C for the mylonite with microstructure B based on the two feldspar thermometry (Shigematsu and Yamagishi, 2002). Some of the my-

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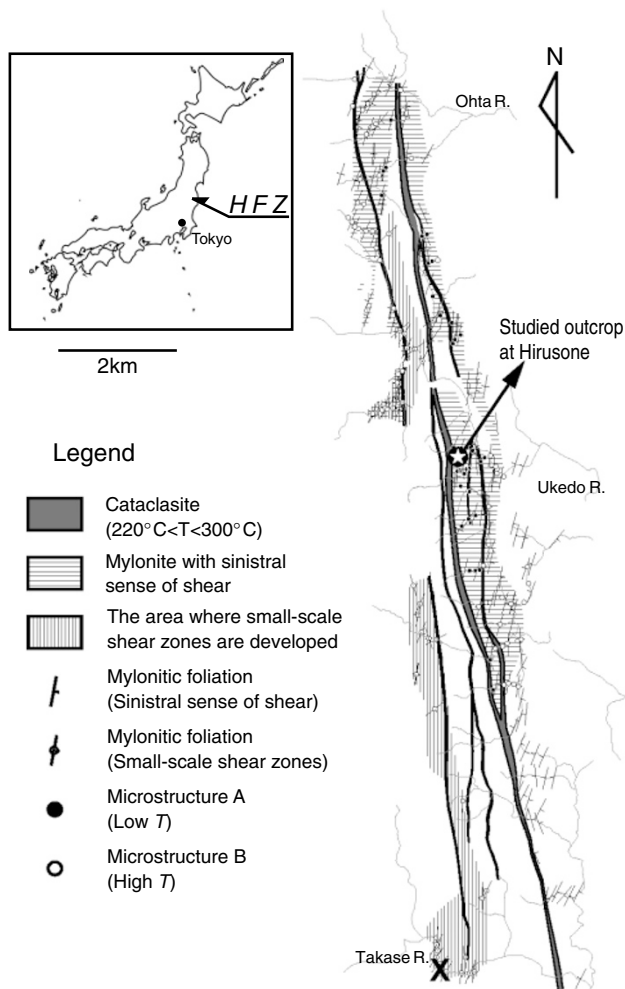


Fig. 1. Distribution of the fault rocks in the Hatagawa Fault Zone (HFZ) NE Japan. The point X along the Takase River (the south end of the map) indicates the studied outcrop by Tanaka *et al.* (2002).

lonites have undergone hydrothermal alteration during the deformation. In general, the mylonites deformed at higher temperature exhibit little alteration (Tsurumi *et al.*, in press), except chloritization of mafic minerals. The mylonites deformed at lower temperature are weakly altered especially close to the main cataclasite.

The cataclasite is usually intensively altered characterized by the mineral assemblage of chlorite + epidote + mica + calcite, suggesting that the alteration temperature is approximately 220–300°C (Tomita *et al.*, 2002). The alteration mineral assemblages of the mylonites are nearly the same, however, calcite is less common and prehnite sometimes occurs.

The temperature ranges estimated above cover the typical temperature of inland earthquake source region (e.g., Scholz, 1988). The Hatagawa Fault Zone is, therefore, considered to have been a possible Cretaceous seismogenic zone (Tomita *et al.*, 2002). The size of the main cataclasite zone is almost equal to that of the rupture zone of M7 class earthquake.

3. Samples and Results of Whole Rock Chemistry and Oxygen Isotope Ratio

The studied samples were collected from a quarry located at Hirusone (the location is shown in Fig. 1), about 200 m east of the main cataclasite zone. Weakly to strongly deformed granodiorites (deformed at lower temperature) are exposed in a quarry. Figure 2(a) shows a distribution of fault rocks over a distance of 60 m at a large outcrop in a direction almost perpendicular to the foliation strike. We divided the deformed granodiorite into three zones, based on the intensity of development of foliation. The deformation is heterogeneous even in hand specimen scale, however, deformation is more intense in the central to the western part of Fig. 2(a), which is nearer to the main cataclasite zone. There are numerous zones of localized deformation with a thickness of mm to cm scale. Such deformation localized zones are abundant in the central part of the outcrop, where deformation is strong. Sometimes the central part of deformation localized zones is crushed and epidote fracture-fill is present (Fig. 3). Some crushed fragments are elongated (Tomita *et al.*, 2002) and epidote veins seem to have been overprinted by later mylonitization, suggesting that the cataclastic and plastic deformations occurred intermittently.

Whole rock chemistry and oxygen isotopic ratio were analyzed as shown in Table 1. We selected five representative samples from each deformation zone as shown in Figs. 2(b)–(f). We collected the least deformed and altered sample as a reference from a few tens meters away from the outcrop because the intact part of the same granodiorite is not exposed. Two hand specimens showing heterogeneous deformation and alteration were also analyzed to check the heterogeneity within a sample. One is cataclasite (Fig. 2(f)) and the other is heterogeneously deformed granodiorite including an ultramylonite band and epidote veins (Fig. 3). The four representative deformed granodiorites show similar major element compositions (Fig. 4), indicating they were originally the same rock body. Figure 4 is an isocon diagram showing the compositional changes of deformed granodiorites compared with the reference sample. Most components except H_2O^- , LOI (loss of ignition) are on the line with the inclination of unity, suggesting that volume change and compositional change are small among deformed granodiorites. Oxidation state may be changed since FeO increases whereas total Fe content does not change. As for cataclastic part, CaO, Na_2O and K_2O seem to be changed probably due to alteration of feldspar.

Mineral assemblages in the reference sample and deformed granodiorites are quartz - plagioclase - potassium feldspar - biotite - hornblende - magnetite - ilmenite for primary minerals and chlorite - epidote - illite - (calcite) for secondary minerals. Alteration such as chloritization of biotite is more intense in strongly deformed granodiorite. Calcite is abundant as vein or matrix filling in the cataclastic part.

$\delta^{18}\text{O}$ and LOI show slight changes (Table 1). LOI represents total concentration of volatile elements. In this case, LOI is nearly equal to sum of H_2O and CO_2 . The $\delta^{18}\text{O}$ value is between 0.4‰ (SMOW) and 3.9‰ (SMOW). The value is considerably lower than the typical granitic rocks in Japan. According to Sakai and Matsuhsa (1996), $\delta^{18}\text{O}$ is between 5 and 8‰ (SMOW) for the magnetite series granitic rocks.

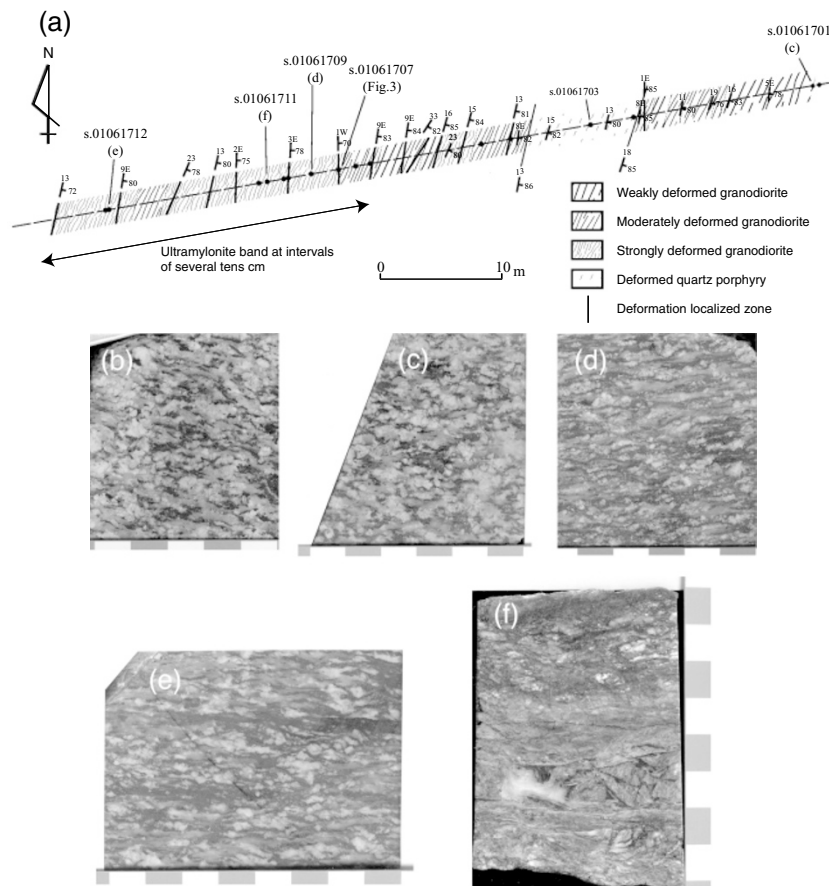


Fig. 2. (a): Lithologic map of the quarry at Hirusone, about 200 m east of the main cataclasite zone. (b)–(f): Granodiorites showing the different degree of deformation from the quarry at Hirusone. (b) Reference sample 01061713B, (c) weakly deformed 01061701, (d) moderately deformed 01061709, (e) strongly deformed 01061712, (f) cataclastic part 01061711. The divisions of the scales are 1 cm. Sampling points are shown in the lithologic map except (a). (a) is sampled from a few tens meters from the outcrop.

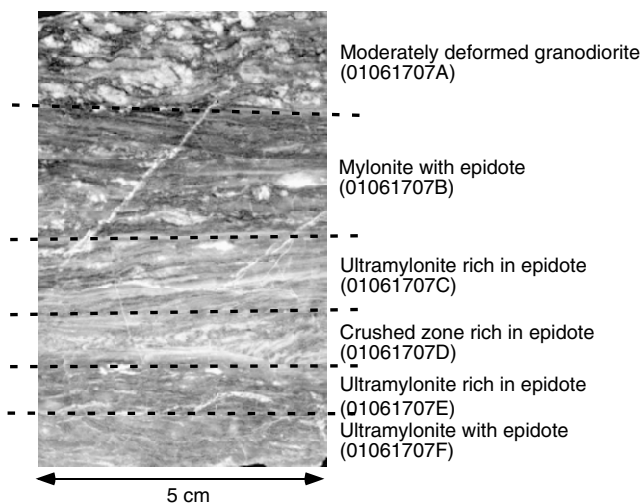


Fig. 3. Heterogeneously deformed granodiorite sample from Hirusone (01061707). Ultramylonite is closely associated with epidote veins (white bands parallel to the foliation).

The intact granodiorite (magnetite series), which is not exposed, probably has the $\delta^{18}\text{O}$ value more than 5‰ (SMOW). The most probable reason for the decrease in $\delta^{18}\text{O}$ is interaction with near-surface derived water, since $\delta^{18}\text{O}$ value is zero for sea water and usually negative for meteoric water (Sakai

and Matsuhisa, 1996).

Figure 5 shows the relation between LOI and $\delta^{18}\text{O}$. $\delta^{18}\text{O}$ value has a negative correlation with LOI except for samples from localized deformation zone rich in epidote (01061707A–F). A strongly deformed granodiorite sample (01061712) has higher LOI value and lower $\delta^{18}\text{O}$ value than that of the reference sample. This supports the idea that interaction with water reduces $\delta^{18}\text{O}$ value during plastic deformation.

Hydrous minerals in mylonite are biotite (3 to 4 wt% H_2O) and hornblende (less than 2wt% H_2O) as primary minerals and chlorite (10 to 15 wt% H_2O) and epidote (1 to 2 wt% H_2O) as secondary minerals. An increase in LOI probably results from formation of chlorite. In contrast, epidote-rich rock has a lower LOI value compared to the weakly deformed granodiorite, whereas, $\delta^{18}\text{O}$ value decreases. It is difficult to make sure that deformation and interaction with water is contemporaneous or not.

4. Occurrence of Calcite and Fluid Chemistry

As mentioned before, calcite characteristically occurs in the cataclastic part of the fault zone. The deformation temperature of the mylonites deformed at lower temperature is 260 to 310°C based on two-feldspar geothermometer (Shigematsu and Yamagishi, 2002). The formation temperature of cataclasite is 220 to 300°C (Tomita *et al.*, 2002).

Table 1. Major element concentrations and oxygen isotope ratio. Sample descriptions: 01061713B - very weakly deformed and altered granodiorite used as reference, 01061701 - weakly deformed granodiorite, 01061709 - moderately deformed granodiorite, 01061712 - strongly deformed granodiorite, 01061707A~F - heterogeneously deformed granodiorite including ultramylonite band and crushed zone rich in epidote, 01061711A~B - cataclastic part in strongly deformed granodiorite. The photos of the samples are shown in Figs. 2 and 3.

Sample	SiO ₂ %	TiO ₂ %	Al ₂ O ₃ %	Fe ₂ O ₃ %	FeO %	MnO %	MgO %	CaO %	Na ₂ O %	K ₂ O %	P ₂ O ₅ %	H ₂ O %	LOI %	total %	δ ¹⁸ O ‰ (SMOW)
01061713B	70.01	0.47	12.57	1.92	1.81	0.1	1.1	3.73	3.51	3.31	0.1	0.08	1.4	100.11	3.6
01061701	67.17	0.5	14.87	1.08	2.72	0.08	1.32	3.92	3.72	3.02	0.12	0.14	1.37	100.03	3.9
01061709	67.36	0.47	14.45	0.98	2.62	0.08	1.19	3.95	3.67	2.93	0.11	0.23	1.87	99.91	3.3
01061712	67.54	0.45	14.65	0.58	2.66	0.1	1.2	4.05	3.53	2.66	0.11	0.43	2.17	100.13	2.3
01061707A	69.74	0.43	13.4	1.29	2.17	0.09	1.11	3.72	3.38	3.03	0.11	0.09	1.02	99.58	3.2
01061707B	73.73	0.37	11.72	1.06	1.85	0.08	0.86	3.62	2.62	2.69	0.09	0.29	1.12	100.1	3.5
01061707C	70.38	0.36	14.21	1.02	1.75	0.08	0.78	4.11	3.48	2.87	0.09	0.11	0.9	100.14	3.6
01061707D	68.45	0.36	13.43	2.35	1.23	0.1	0.48	5.17	3.46	3.72	0.09	0.07	1.02	99.93	2.9
01061707E	68.29	0.41	14.08	1.13	2.2	0.1	0.99	3.62	3.77	3.84	0.1	0.2	1.67	100.4	3.8
01061707F	68.93	0.44	12.67	1.11	2.3	0.1	0.96	3.6	2.84	4.35	0.11	0.1	1.92	99.43	2.9
01061711A	55.68	0.6	19.27	1.44	2.98	0.1	1.47	5.38	7.29	2.81	0.15	0.13	2.57	99.87	0.4
01061711B-I	67.54	0.48	14.92	1.08	2.53	0.08	1.18	2.45	4.35	3.37	0.12	0.08	1.85	100.03	2.3
01061711B-II	65.37	0.41	14.11	0.86	2.53	0.1	1.09	5.85	3.36	3.81	0.1	0.12	1.97	99.68	2.1

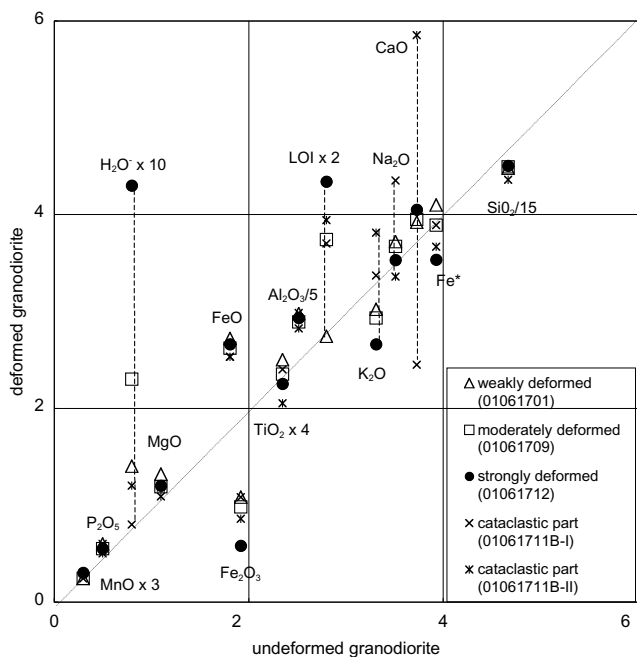


Fig. 4. Isocon diagram showing the compositional changes of deformed granodiorites with reference to the least deformed and altered one. Chemical compositions in wt% are scaled to fit on the plot.

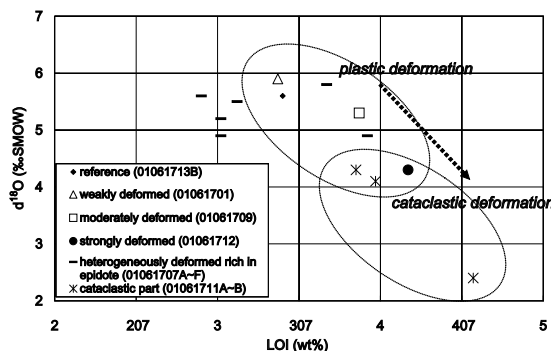


Fig. 5. Relations between LOI and oxygen isotope ratio.

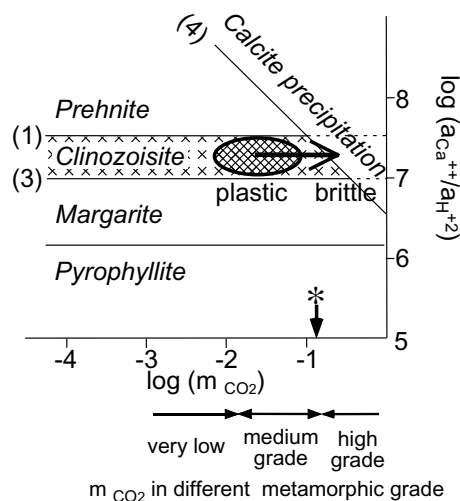


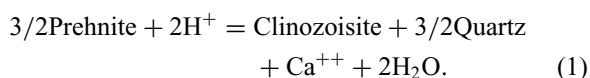
Fig. 6. Activity diagram of the system CaO-Al₂O₃-SiO₂-H₂O-CO₂ at 300°C, 100 MPa, showing the stability field of epidote (clinozoisite) and calcite. * indicates CO₂ concentration of the fluid inclusion in pseudotachylyte at the Nojima fault, SE Japan (Boullier *et al.*, 2001).

Thus, the temperature difference between the mylonite and cataclastite is not particularly large.

Here, we will examine the chemical equilibrium of Ca-bearing aluminosilicate minerals with respect to fluid chemistry, particularly, Ca⁺⁺/H⁺ activity ratio and CO₂ partial pressure using computer code SUPCRT92 (Johnson *et al.*, 1992). Temperature and pressure are assumed to be 300°C and 100 MPa respectively. Ca-bearing aluminosilicate minerals stable at temperatures of about 300°C are prehnite-clinozoisite-margarite (Ca-bearing mica). The fluid is assumed to be saturated with quartz, which is a reasonable assumption as the country rock is granodiorite, where quartz is one of the dominant minerals.

Figure 6 shows the activity diagram indicating the stability relations among prehnite (Ca₂Al₂Si₃O₁₀(OH)₂), clinozoisite (Ca₂Al₃Si₃O₁₂(OH)), margarite (CaAl₂(Al₂Si₂O₁₀)(OH)₂) and calcite (CaCO₃) and pyrophyllite (Al₂Si₄O₁₀(OH)₂). For

simplicity, epidote is assumed to be pure clinozoisite component, though the epidote group exhibits solid solution between clinozoisite and epidote. The prehnite-clinozoisite reaction is represented as follows.

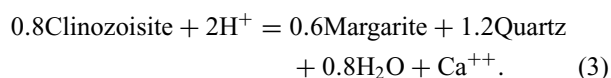


This relation leads to the following equation, assuming the activity coefficient of water and the solid phase to be unity.

$$\log K_{(1)} = \log(a_{\text{Ca}^{++}}/a_{\text{H}^+}^2) = -\Delta Gr_{(1)}/2.303RT. \quad (2)$$

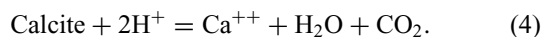
Here, $K_{(1)}$ and $\Delta Gr_{(1)}$ indicate the equilibrium constants and the changes in Gibbs free energy of the reaction (1). R and T indicate the gas constant and the absolute temperature respectively. The activity ratio of $a_{\text{Ca}^{++}}/a_{\text{H}^+}^2$ is fixed to 7.51 in logarithmic scale since ΔGr can be calculated based on the formation energy of each components.

Clinozoisite reaction to margarite is represented as follows:



Here, the activity ratio of $a_{\text{Ca}^{++}}/a_{\text{H}^+}^2$ should be 6.99 when the above minerals coexist. Prehnite and margarite do not coexist. Thus, the activity ratio of $a_{\text{Ca}^{++}}/a_{\text{H}^+}^2$ should be between 6.99 and 7.51 when clinozoisite is stable at 300°C. Solid solution expands the stability field relative to that for pure clinozoisite.

Then, the stability of calcite is calculated based on the following reaction.



This reaction leads to the following equation of calcite stability.

$$\log K_{(4)} = 6.54 = \log(a_{\text{Ca}^{++}}/a_{\text{H}^+}^2) + \log(m_{\text{CO}_2}). \quad (5)$$

This equation is shown as line (4) in Fig. 6. Calcite is oversaturated above the line and it is undersaturated below the line. The CO_2 concentration in the fluid at the deformation stage is difficult to be evaluated. However, Brown (1998) showed a relationship between CO_2 concentration and metamorphic grade based on fluid inclusion data. $X(\text{CO}_2)$ ranges between 0.015 and 0.148 in medium-grade and fluid is more CO_2 rich in high-grade rocks and it is less rich in low-grade rocks. If this value is applicable to the Hatagawa Fault Zone, the fluid might be undersaturated with calcite as shown in Fig. 6. This is consistent with our observation that epidote is common and calcite is rare in the mylonites. Epidote-chlorite-albite assemblage characterizes alteration under small water-rock ratio condition (e.g., Reed, 1997). This indicates that alteration in the mylonite might be rock-dominant and the fluid is rather stagnant. This is supported by little compositional change (Fig. 4). In order to precipitate calcite in such an environment, the fluid chemistry must change toward higher $a_{\text{Ca}^{++}}/a_{\text{H}^+}^2$ condition or higher m_{CO_2} condition. Considering that prehnite, which

is stable under higher $a_{\text{Ca}^{++}}/a_{\text{H}^+}^2$ conditions is not common in the Hatagawa Fault Zone, increase in m_{CO_2} might be plausible. It is possible that the fluid rich in CO_2 might have migrated through the fractured part of the fault zone. Boullier *et al.* (2001) discovered CO_2 -rich fluid inclusions in pseudotachylyte in the Nojima fault, Japan. The CO_2 concentration in the Nojima fault is about 15%, which may be rich enough to precipitate calcite. It is very interesting to clarify the timing of introduction of CO_2 -rich fluid to the fault zone. More careful study on the calcite occurrences and isotope analysis may be necessary to solve effect and origin of CO_2 . It is also an interesting problem whether fluid chemistry affects mechanical behavior of fault rocks or not, since physicochemical properties of water in grain boundaries depend on fluid chemistry such as CO_2 concentration and salinity and this could affect the mechanical behavior of crust (Nakashima *et al.*, 2001).

5. Concluding Remarks

In the Hatagawa Fault Zone, close association of brittlely and plastically deformed fault rocks is one of the characteristic features. Both rocks are interpreted to have formed at seismogenic depth. Epidote and chlorite are dominant alteration minerals in both mylonite and cataclasite, whereas, calcite is characteristically distributed in cataclasites. Decrease in $\delta^{18}\text{O}$ value and existence of epidote and chlorite, even in weakly deformed granodiorite, is evidence of water-rock interaction. The water/rock ratio is interpreted to be relatively small and fluid chemistry is buffered by host rock. Positive correlation between decrease in $\delta^{18}\text{O}$ and degree of deformation indicates the involvement of water during mylonitization. The occurrence of calcite is explained by the changes in fluid chemistry. Probably, CO_2 -rich fluid was introduced at the cataclastic deformation stage and increased CO_2 partial pressure resulted in precipitation of calcite.

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