Free-radical reactions by mechano-chemical process within seismic fault zones: Results from fundamental experiments

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Fracture zones within active fault zones are known to be passages of fluids. The fluid penetration in the fracture zone leads into fluid-rock interaction that changes the physical and chemical properties of the fault zone materials, affecting the dynamic behavior of the fault zone. One of the evidence of the fluid-rock interaction is the generation of hydrogen gas along active fault zone (e.g., Wakita et al., 1980). Kita et al. (1982) first explained that the hydrogen gas was generated by free radical reactions between pure water and new surface of crushed quartz. On the other hand, the fluids are another important media, which react with the activated new surfaces of destructed fault rocks. Tanaka and Lockner (personal communication) focuses on generation of hydrogen ions (H+) during the tri-axial quartz sand deformation experiments and shows remarkable decrease of pH during the experiments. Generation of hydrogen ions would be potentially significant to invoke chemical reactions such as dissolution-precipitation of feldspars (Garrels and Christ, 1965; Faure, 1991) in fluid-filled damaged fault zone, which probably induces fault weakening (e.g., Chester et al., 1993; Caine et al., 1996) or strengthening (e.g., Olsen et al., 1998).

Thus, we perform batch-style crushing experiments of single crystals of quartz, alkali feldspar and biotite and granites within weakly buffered pH7 solutions (KH2PO4 + NaOH, 10-4 ionic strength) under argon-atmospheric conditions using a glove box at the ambient temperature and pressure. The reason of the usage of the weakly buffered solution is to overcome the difficulty to stabilize the pH of pure water. The attention is mainly directed what is the relations between the variation of hydrogen ions in the solutions and the formation of hydrogen gas for the natural granite and the each constitutive mineral. BET method was adopted for precise measurements of net surface areas of the samples. The results are the followings:

(1) pH variations: pH values decrease for the experiments of quartz single crystal with increasing crushing times (0.5, 1, 1.5, 2 hours). The lowest pH value is 6.35 for the 2 hours experiment. The other single crystal and granite experiments show the pH increases. The pH values are considered to present the solvation and saturation of alkali-metal ions, such as K+ and Na+, that is different from the experiments of quartz crystal.

(2) H2 variations: The amounts of hydrogen gas linearly increase with increasing the experimental duration time for all the crushing experiments.

(3) BET surface area: The BET surface area increases with increasing the experimental duration.

Thus, at the ambient temperature and pressure, both amounts of hydrogen gas and hydrogen ions increase proportionally with increasing the newly formed surface area for single quartz crystal experiments. However, it is not always for the other minerals and granite because the amounts of hydrogen ions decrease even though the amounts of hydrogen gas and the net surface area increase, probably suggesting the influence of the dissolution reaction with alkali-metal ions.