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Rate-limiting process and degree of disequilibrium of garnet-forming reaction

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Water released by dehydration reaction in metamorphic rocks will take a major role in rheology, mass transport, and reactivity of metamorphic rocks. Rate-limiting process of the dehydration reaction can be divided into the following three steps, such as reaction at interface, diffusion, and heat-flow. Garnet is one of the common minerals produced by dehydration reaction in metamorphic rocks. The rate-limiting process of garnet-forming reaction is usually assumed to be diffusion-controlled reaction. However, there are few cases where clear evidences for diffusion-controlled reaction were proposed. In addition, degree of disequilibrium of the diffusion-controlled reaction is not well known. Here, I report evidence of diffusion-controlled reaction and estimation of degree of disequilibrium of garnet-forming reaction in the Tsukuba Metamorphic Rocks.

Garnets in the Tsukuba Metamorphic Rocks are formed by the dehydration reaction of biotite + sillimanite + quartz = garnet + cordierite + K-feldspar + water. Biotite-depleted region surround the irregular shaped garnet. The depleted region of reactant is typically expected for diffusion-controlled reaction, but is rare for natural garnet-forming reaction. Irregular shape of garnet is also expected for diffusional instability of growing interface (Mullins & Sekerka, 1963). These sets of observations strongly suggest that the garnets were formed by diffusion-controlled reaction.

Spherical shape of growing particle under diffusion-controlled reaction becomes unstable due to diffusional instability, but interfacial energy will reduce the instability. Using spherical harmonics function, instability of small perturbation from spherical shape can be evaluated. Assuming interfacial energy of garnet, dominant wavelength in unstable regime is predicted with degree of super-saturation under diffusion-limited reaction. Dominant wavelength of the irregular garnet suggests that degree of super-saturation is less than 0.1. This value can be translated to degree of disequilibrium temperature with entropy change of the garnet-forming reaction. The disequilibrium temperature $\triangle T$ is less than 5 ℃, and is very small. This small value of disequilibrium temperature suggests that spherical or euhedral garnets that are more common than irregular garnets, should be produced near equilibrium condition. Otherwise, such common spherical or euhedral garnets should be produced by interface-controlled reaction associated with influent fluid.

Keywords: garnet, dehydration reaction, disequilibrium, metamorphism, metamorphic rock, metamorphic reaction