

Estimation of maximum metamorphic temperature from Al/Si-order parameter in sillimanite

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The polymorphs of Al_2SiO_5 (andalusite, kyanite, sillimanite) are valuable for metamorphic rocks as indicators of pressure and temperature. Moreover, crystal structure and micro-texture of sillimanite can provide more detailed information about thermal history. For example, Raterron *et al.* (2000) observed the micro-textures in sillimanite, which were formed by the partly transformation to mullite ($\text{Al}_2(\text{Al}_{2+2x}\text{Si}_{2-2x})\text{O}_{10-x}$, $x \cong 0.17-0.59$) at high temperatures. Furthermore, Zen (1969) and so on suggested that Al/Si-distribution in TO_4 tetrahedra of sillimanite should become continuously disordered with adding temperature. Although this Al/Si-disordering, in particular, has a possibility of a powerful geothermometer, the quantification of Al/Si-order parameter has been never succeeded. Recently, Igami *et al.* (JpGU, 2016; 2017) successfully determined the Al/Si-order parameter from micrometric region by HARECXs (High Angular Resolution Electron Channeling X-ray Spectroscopy, *e.g.* Soeda, 2000) method using TEM-EDS and obtained the relationship between the Al/Si-order parameter and temperature. In this study, HARECXs experiment was applied to natural sillimanite from metamorphic rocks, and the maximum metamorphic temperature of analyzed sillimanite was estimated.

Analyzed samples were sillimanite from Rundvågshetta, Lützow-Holm Complex, East Antarctica (sample No. RVH92011102A), and that from Mt. Riiser-Larsen, Napier Complex, East Antarctica (sample No. TH96123009). Both regions are assumed to be the UHT metamorphic regions (*e.g.* Kawasaki *et al.*, 2011; Hokada, 2001). No characteristic textures were observed in these sillimanite by optical microscope and SEM. Ultrathin sections were prepared from these sillimanite using focused ion beam (FEI Quanta 200 3DS or Helios NanoLab 3G CX), and then observed and analyzed by TEM-EDS (JEOL, JEM-2100F, JED-2300T).

In RVH92011102A, no characteristic textures were observed, and HARECXs analysis showed that the Al/Si-order parameter converged on ~ 0.88 . In TH96123009, on the other hand, abundant anti-phase boundaries (APBs) were observed, and HARECXs analysis showed that the Al/Si-order parameter slightly converged on around 0.90, but widely distributed ranging from ~ 0.6 to ~ 0.9 .

Generally, Al/Si-order parameter are assumed to have the lowest value at the peak temperature and to become somewhat higher during cooling. The HARECXs results of RVH92011102A implies that the peak temperature is > 1000 °C by comparison with those of experimentally heat-treated sillimanite (Igami *et al.*, 2016; 2017). This temperature is consistent with previous estimates (*e.g.* Kawasaki *et al.*, 1993; 2011; Harley, 1998; Fraser *et al.*, 2000). On the other hand, the lowest Al/Si-order parameter obtained from the HARECXs results of TH96123009 is ~ 0.65 , and this value implies that the peak temperature is much higher than RVH92011102A. However, this low Al/Si-order state is thought not to be formed by stoichiometric disordered sillimanite and to be formed by the transformation from mullite + SiO_2 to sillimanite, because mullite has similar framework with sillimanite and its Al/Si-distribution is disordered. The APBs observed in TH96123009 is also thought to be formed at the same time. Therefore, the peak temperature of this sillimanite is assumed to be above the transition temperature between sillimanite and mullite + SiO_2 , ~ 1200 °C. This result provides further constraint on previous estimates, > 1100 °C (Harley and Motoyoshi, 2000; Hokada, 2001).

Keywords: sillimanite, Al/Si-order parameter, geothermometer, Napier Complex, HARECXs, TEM

