

# The sulfur isotopic ratio of Neogene granitoids in Kyushu

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The batholiths and small stocks of granitoids of Neogene are located in the whole area of Kyushu, and the area is divided into three zones, the Outer zone of Southwest Japan near the Pacific coast line (Outer zone 1 : reduced type : Osuzuyama, Takakumayama, Osumi and Yakushima bodies), the Outer zone of Southwest Japan near the mid-tectonic lines (Outer zone 2 : Okueyama, Ichifusayama, Shibisan, Mukaeyama, Suzuyama and Kinpouzan bodies), and the Inner zone of Southwest Japan (Inner zone : oxidized type : Tushima, Goto, and Koshikizima bodies), based on the kind of associated ore deposit, halogen content, redox condition, source materials, tectonics of magma emplacement, and assimilation and contamination of sedimentary rocks. In this study, we measured the sulfur isotopic ratio of these granitoids. There are many reports on the sulfur isotopic ratio of the granitoids in Japan (e.g., Sasaki and Ishihara, 1979).

Pyrrhotite occurs coexisting with biotite and other rock-forming minerals in the granitoids in the Outer zone 1. The sulfur content is correlated with SiO<sub>2</sub> of whole rock, decreases from 1400ppm to several ppm with the increase of SiO<sub>2</sub>. In contrast, sulfur content of the Inner zone varies widely. For instance, sulfur content of many granitoids of the Goto body is lower than several ppm, although, there are some rocks of which the sulfur contents reach thousand ppm. There is no correlation with SiO<sub>2</sub> content. The sulfur contents might be related to the mineralization. The Tushima body reaches to 2000ppm. The granitoids in the Outer zone 2 shows the similar features. At the Okueyama complex, sulfur content is hardly contained in the central batholith, but high in the stocks near the Obira and Mitate ore deposits. The Suzuyama stock, intruded into the center of mineralization zone, contains sulfur up to 2000ppm.

The behavior of sulfur in magma depends largely on the redox condition. H<sub>2</sub>S or HS<sup>-</sup> might be dominant in the reduced magma in the Outer zone 1. The existence of pyrrhotite as an ordinal rock-forming mineral suggests that magmatic sulfur had been consumed by the crystallization of pyrrhotite and had decreased in the residual magma. The addition of sulfur by the assimilation of country rocks might not be important compared to the subtraction of magmatic sulfur. In contrast, sulfur might exist as SO<sub>2</sub> as well as H<sub>2</sub>S in the oxidized magma in the Inner zone, and SO<sub>2</sub> is discharged from the magma. As a result, the sulfur content in the granitoid might become remarkably low. When the discharged SO<sub>2</sub> is cooled in the lower PO<sub>2</sub> condition than that of magnetite-hematite buffer, SO<sub>2</sub> changes to H<sub>2</sub>S and is precipitated as pyrite or pyrrhotite by the subsolidus reaction (Ohmoto and Goldhaber, 1997). This tendency might appear especially at the granitoids related with mineralization.

In the Outer zone, the sulfur isotopic ratios are all negative values from -1 to -13 per mill, while the Inner zone from +2 to +11 per mill. In the Outer zone 1, the sulfur isotopic ratio correlates with SiO<sub>2</sub> content, and becomes lighter with the increase of SiO<sub>2</sub>. A similar tendency is observed in the Inner zone.

As the fractionation factor of pyrrhotite to H<sub>2</sub>S and HS<sup>-</sup> are slightly positive, <sup>34</sup>S should fractionate into pyrrhotite. Therefore, the sulfur isotopic ratio of the granitoids in the Outer zone 1 decreased during crystallization. The evidence might be able to interpret without the influence of the country rock, although the assimilation and contamination might be effective. In contrast, in the magma in the Inner zone, both H<sub>2</sub>S and SO<sub>2</sub> might exist. As the sulfur isotopic ratio of SO<sub>2</sub> is always about 2 per mill higher than that of H<sub>2</sub>S at the magmatic temperature, H<sub>2</sub>S is always oxidized to SO<sub>2</sub> under the buffering condition by the mineral assemblages in granitoids. The light sulfur both in the magma and discharged SO<sub>2</sub> might increase during the cooling of magma by the Rayleigh effect.