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## Utility of apatite as a proxy for solidification processes of magma revealed in the Nosappumisaki intrusion

Yumi Hisaoka<sup>1\*</sup>, Tsuyoshi Iizuka<sup>1</sup>, Naoto Takahata<sup>2</sup>, Kazuhito Ozawa<sup>1</sup>, Hiroko Nagahara<sup>1</sup>, Yuji Sano<sup>2</sup>

<sup>1</sup>Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo, <sup>2</sup>Atmosphere and Ocean Research Institute, the University of Tokyo

We have examined behavior of volatile elements during cooling and solidification of a magma in the crust based on morphology, chemical composition, and hydrogen isotopes of apatite in the Nosappumisaki intrusion, which has been extensively studied on the processes of crystal differentiation and solidification.

The Nosappumisaki intrusion is a sheet-like intrusion intruded into the Nemuro formation in the Nemuro peninsula, Hokkaido. The basic structure is a lower melanocratic cumulate zone overlain by an upper leucocratic monzonite zone. This structure is inferred to have formed by a quick and massive crystal settling from a magma initially laden with crystals in a sheet like magma body, (Simura and Ozawa, 2006) followed by a slow stage of fractional crystallization caused by compositional convection in a closed system (Simura and Ozawa, 2011).

In the intrusion, apatite occurs in all of the lithologies except for chilled zones showing short columnar to acicular euhedral morphology. The aspect ratio and number density of apatite increase from the lower cumulate zone to the upper chilled zone, suggesting that the cooling from the upper contact of the intrusion is more effective than that of the lower contact.

Apatite crystals in three rocks representing earlier and later stages of solidification respectively were analyzed with NanoSIMS to determine the water content and hydrogen isotopic composition. The obtained average isotopic composition and water content are -109 per mill and 1.26 wt.% for the earlier stage of solidification and 18.2 per mill and 1.29 wt.% for the later stage of solidification. We also analyzed apatite with an EPMA to determine F and Cl contents. The Cl contents of apatite in the upper monzonite zone and the lower cumulate zone, which solidified earlier, are comparable. The Cl content decreases toward near the contact between the monzonite and cumulate zones, which solidified later. In contrast, the F contents of apatite are inversely correlated with the Cl contents. There are also zoned apatite grains, in which Cl content decreases from the core to rim with oscillation. Melt/fluid composite inclusions are present localized in a zone with lower Cl content.

The most important result of our study is the increase of F/Cl and D/H of apatite with solidification with minor variation of the water content. If the fractionation and solidification processes took place in a closed system, F/Cl in the magma decreases keeping D/H constant, because minerals crystallized with apatite after the intrusion are anhydrous minerals and biotite (Mg/(Mg+Fe)<sup>~</sup>60), to which F is more preferentially partitioned than Cl relative to silicate melts (Icenhower and London, 1997). However, the F/Cl and D/H increase contrary to this expectation, indicating that the variation of F/Cl of apatite was not caused by crystal fractionation. The most plausible scenario to account for the behavior is vesiculation/degassing at a certain stage of solidification of the intrusion. This inference is supported by followings: (1) F is more preferentially partitioned into melt than into gas as compared with Cl (Signorelli and Carroll,2000;Webster, 1990), (2) the D/H of apatite has a variation, (3) melt/fluid inclusions selectively occur in Cl-poor zones in zoned apatite grains, suggesting vesiculation caused depletion of Cl in the apatite. In addition, the increase of D/H with fractionation suggests that vesiculation/degassing occurred not as H<sub>2</sub>O but as CH<sub>4</sub> or H<sub>2</sub> (Kyser and O'Neil, 1983). This implies that differentiation and solidification of the Nosappumisaki intrusion took place under a reduced condition being udersaturated with H<sub>2</sub>O.

Information of vesiculation/degassing, gas speciation, and oxidation-reduction state of magma can be obtained using F/Cl and D/H of apatite. Apatite is a useful proxy for behaviors of volatile elements during the solidification of magma.