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Ultrahigh-sensitive simultaneous determination of halogens and noble gases by an extension of Ar-Ar and I-Xe methods

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Noble gas isotope ratios in various geochemical components in the Earth are significantly different, making them useful tracers to constrain origin of volatiles in the mantle. The development of noble gas mass spectrometry during the last two decades has enabled us to detect less than 10000 noble gas atoms (e.g., [1]).

An extension of Ar-Ar and I-Xe dating methods allows us to simultaneously determine trace amounts of noble gases, halogens, K, Ca, Ba, and U by use of ultrahigh-sensitive noble gas mass spectrometry on neutron-irradiated samples. This method has several advantages: (i) detection limits for halogens are three or four orders of magnitude lower than those of other conventional analytical methods, (ii) several components of different origin can be distinguished based on their relations with specific noble gas isotopes such as mantle-derived ^{3}He and by using various noble gas extraction methods such as laser microprobe [2], and (iii) in-situ production of radiogenic noble gas isotopes (such as ^{4}He and ^{40}Ar) after the entrapment of the noble gas component of interest in the sample can be corrected by the simultaneous determined their parent elements, such as U and K, when the age of the entrapment is known or can be assumed.

We have developed a new noble gas mass spectrometric system for this method based on an Ar-Ar and I-Xe dating system [3]. Accuracy and precision of our method were examined by analyzing GSJ and USGS reference materials, their original rocks, and scapolite standards [4] and by comparing the halogen data with those obtained with ion chromatography and ICP-MS followed by pyrohydrolysis extraction [5].

By using this method, we analyzed halogens and noble gases in exhumed mantle wedge peridotites and eclogites from the Sanbagawa-metamorphic belt, southwest Japan and those in mantle-derived xenoliths from Kamchatka and N. Philippines, in all of which relicts of slab-derived water are contained as hydrous mineral/fluid inclusions. The striking similarities of the observed noble gas and halogen compositions with marine pore fluids [6,7] challenge a popular concept, in which the water flux into the mantle wedge is controlled only by hydrous minerals in altered oceanic crust and sediment (e.g., [8]).

On the other hand, halogen ratios of olivines in lavas from the northern Izu-Ogasawara arc [9] indicate insignificant contribution to the mantle wedge of pore fluid-derived halogens. This implies a relatively small amount of the pore water subduction fluids would be released from the Izu slab at a sub-arc depth resulting in further subduction to great depths in the mantle, possibly resulting in the seawater-like heavy noble gas composition of the convecting mantle [10].

Based on the relation with ^{129}Xe produced from decay of short-lived nuclide ^{129}I during stepwise heating noble-gas extraction of the Allende and Shallowater meteorites, intrinsic I and U to the meteorites were distinguished from those of terrestrial contamination origin.

These results demonstrate that simultaneous determinations of noble gases, halogens, K, Ca, Ba, and U in mantle-derived rocks and meteorites provide important information about their origins.

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