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Gas phase chemistry of the volatile chloride compound of Hf isotopes

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Gas-phase chemistry is one of the most utilized techniques to study chemical properties of superheavy elements. An adsorption enthalpy of volatile compounds of these elements can be determined with their adsorption-desorption processes on a gas chromatographic column surface. The gas phase chemistry for group 4 elements, Zr, Hf, and Rf has been performed by several groups, and it is reported that the order of the volatility of their tetrachlorides is $Zr \gg Rf > Hf$ [1, 2] and $Zr \sim Hf \sim Rf$ [3]. However, in macro-scale, tetrachloride of Hf is known to be more volatile than that of Hf. Until now, we have investigated gas chromatographic behavior of volatile chloride compounds of Zr and Hf, and the transfer process in the column using Hf radioisotopes of various half-lives. The results show a similar behavior with nonvolatile compound, oxychloride which is formed by oxygen in a system [1]. Therefore, we investigated in detail oxygen effects on a gas chromatographic behavior.

Hf isotopes were produced in the $^{19}F(xn)$ and $^{18}O(xn)$ reactions using 20 MV tandem accelerator at Japan Atomic Energy Agency (JAEA). The beam energy of ^{19}F and ^{18}O were 122 and 108 MeV, respectively. Produced Hf isotopes were ^{165}Hf (76 s), ^{166}Hf (6.8 min), ^{167}Hf (2.05 min). To compare volatility with a Zr compound, ^{85}Zr (7.9 min) was also produced in the $^{19}F(xn)$ and $^{18}O(xn)$ reaction. Nuclear reaction products were transported to the gas chromatographic apparatus with a carbon cluster in a helium gas flow. The transported products were collected on quartz wool plugged in a quartz tube where HCl gas added to form volatile chloride compounds. Oxygen gas was also introduced in order to examine an oxygen effect on a gas chromatographic behavior. The volatile compounds through the isothermal column were re-transported using a He/KCl gas-jet system, and collected in a cold trap where gamma-rays were measured to obtain the yield of each Hf isotope which passed through the column.

The relative yields for $^{165}, ^{166}, ^{167}Hf$ and ^{85}Zr are regarded as 100% at the column temperature of 450 C. In the condition of oxygen free, the yields of all nuclides increased with the temperature of the column from 200 C up to 300 C. When oxygen was added into the system, the yields at 300 C were very low and the temperature to which the yield reaches to 100 % was 400 C. Since this result agrees with our previous results, it is found that oxygen had actually influenced the chromatographic behavior reported until now. In the model generally used for analysis of the gas-phase chemistry, it is explained that the yield of the volatile compound changes according to the half-life of the nuclide used. But our experimental results show that nuclides of different half-lives exhibit almost the same breakthrough curves in an isothermal chromatography. The detailed reason is under investigation.

[1] A. Tuerler et al., J. Radioanal. Nucl. Chem. 1992, 160, 327-339

[2] B. Kadkhodayan et al., Radiochim. Acta 1996, 72, 169-178.

[3] T. K. Sato et al., J. Nucl. and Radiochem. Sci. 2005, 6, N1-N3.

Primary author: Prof. KUDO, Hisaaki (Niigata University)

Co-authors: Mr MURAKAMI, Masashi (Niigata University); Dr GOTO, Shin'ichi (Niigata University)

Presenter: Prof. KUDO, Hisaaki (Niigata University)

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