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Iodine speciation and $^{129}\text{I}/^{127}\text{I}$ ratios in sedimentary rocks in Horonobe, Hokkaido

Yoko Shimamoto^{1*}, Yoshio Takahashi¹, Yuki Amano², Hiroyuki Matsuzaki³, Yasuyuki Muramatsu⁴, Teruki Iwatsuki²

¹Hiroshima Univ., ²JAEA, ³Univ. of Tokyo, ⁴Gakushuin Univ.

Introduction

Iodine-129 is one of the most problematic radionuclides, which is produced by nuclear related activities such as nuclear power generation and nuclear fuel reprocessing, because of its long half-life (15.7 million years) and high mobility. To investigate behavior of iodine in sedimentary rocks is important to evaluate the effectiveness of the natural barrier for nuclear waste repositories. In Horonobe area, iodine rich brine exists in deep underground. The region is underlain mainly by Neogene to Quaternary marine sedimentary rocks, the Wakkanai Formation (Wk Fm), and the overlying Koetoi Formation (Kt Fm): siliceous and diatomaceous shale. We determined iodine distribution, speciation, and isotope ratio ($^{129}\text{I}/^{127}\text{I}$), to investigate long-term migration of iodine in diatomaceous and siliceous shale.

Experiment

All core and groundwater samples were collected by at JAEA Horonobe underground research center. Iodine concentrations in groundwater samples were determined by ICP-MS. For core samples iodine was separated by pyrohydrolysis or alkaline extraction (using Tetramethyl ammonium hydroxide) and determined by ICP-MS. Iodine species in groundwater were separately detected by high performance liquid chromatography (HPLC)-ICP-MS, using anion exchange column and size exclusion column for separation of IO_3^- and I^- , and organic and inorganic iodine, respectively. Iodine species in core samples were determined by iodine K-edge XANES (SPring-8 BL01B1). Thin sections of core samples were prepared, and iodine mapping were obtained by micro-XRF analysis (SPring-8 BL37XU). $^{129}\text{I}/^{127}\text{I}$ ratios in groundwater and core samples were measured by accelerator mass spectrometry (MALT, Univ. of Tokyo).

Results and discussion

Concentration of iodine in groundwater varied widely and was much higher than that of seawater with high correlation with that of chlorine ($R^2 = 0.90$). Iodine in core samples decreased near the boundary between Wk and Kt Fm. Iodine existed as I^- in groundwater, while that in shale was a mixture of organic and inorganic I. Iodine mapping showed that iodine accumulated to micro region (< 30 micron). Iodine isotope ratios ($^{129}\text{I}/^{127}\text{I}$) were higher in siliceous shale compared with that of groundwater. According to these results, migration of iodine in this area can be expected as follows; 1) iodine accumulated as organic iodine in siliceous sediment, 2) iodine was released from layers deeper than Wk Fm during diagenetic process where I^- was dissolved into iodine-rich groundwater, 3) iodine-rich groundwater was distributed to Wk and Kt Fm due to the compaction of the layers, 4) both iodine and chlorine were diluted by freshwater from the surface.

It was suggested that I^- is released to the ground water during the maturation of organic matter. Dissociated I^- can move toward the surface because of the upward water flow driven by compaction. Thus, iodine rich brine is created by integration of iodine released from underlying formation. Because of high mobility of I^- , released I^- remains in solution phase, and concentration of the iodine in solution has increased during sedimentation history.

Keywords: Iodine, XANES, HPLC-ICP-MS, Iodine isotope ratio