Variations in Adsorption Coefficient of $^{133}$Cs and $^{87}$Sr Caused by Oxidation of Pumice Tuff

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Long term weathering processes in the geosphere surrounding the repository by contacting the surface/subsurface water can lead to the formation of redox front zone. This zone might have significant effect on the adsorption of certain nuclides that could be released from the radioactive wastes. Cesium (Cs) and strontium (Sr), which are common non-redox-sensitive elements released from such wastes, are strategically very important for the performance assessment of a radioactive waste repository. Thousands of radionuclide adsorption data are available in literature where different types of rocks/minerals are used for measuring adsorption quantity in terms of distribution coefficient values of many radionuclides in variety of experimental conditions. In recent years, adsorption characteristics of radionuclides in the oxidizing or reducing conditions have gained interest because of frequent presence of such environment in the subsurface where ground water-rock interaction occurs.

In the present study, an attempt has been carried out to investigate the effect of such redox phenomena on the adsorption coefficient of Cs and Sr on pumice tuff, which is already been selected for hosting low and intermediate level radioactive wastes in Japan. Powder of 150-300 micrometer size pumice tuff was used as solid phases of rock. Specific surface area of the fresh and oxidized pumice tuff were determined by mercury intrusion porosimetry. Mineralogical composition of pumice tuff was done by X-ray diffraction spectrometry and optical microscopy. Batch sorption study was carried at wide range of pH 4-12, varying nuclide concentration from $10^{-4}$ to $10^{-7}$ M and high ionic strength of 1.0 and 3.0. Solid-solution contact was made for 12 weeks with pH adjustment at every 2 weeks as necessary. Redox potential of the solutions was monitored to check the tendency of being oxidized. The Cs and Sr concentration in the aqueous phase was determined by ICP-MS and finally adsorption data-set were simulated by the surface complexation model with the help of IgorPro 6 software. Higher adsorption coefficient values were obtained in the oxidized part than fresh pumice tuff for both cesium and strontium. This is indicative to increase of available adsorption sites on the surfaces of tuffs due to oxidizing phenomena.

Keywords: Distribution coefficient, Cesium, Strontium, Redox zone, Pumice tuff