

## Comparison of sorption coefficients between powder and intact solid phase: Case study with $^{133}\text{Cs}$ and $^{87}\text{Sr}$ on pumice tuff

\*Mohammad Rajib<sup>1</sup>, Chiaki T. Oguchi<sup>2</sup>

1. Graduate School of Science and Engineering, Saitama University/ Bangladesh Atomic Energy Commission, 2. Department of Civil and Environmental Engineering, Saitama University

Adsorption experiment was carried out with both powdered and block pumice tuff to compare the distribution coefficient ( $K_d$ ) values of stable cesium ( $^{133}\text{Cs}$ ) and strontium ( $^{87}\text{Sr}$ ) under variety of geochemical conditions. The main objective was to infer the  $K_d$  difference in laboratory and field condition. Cs and Sr were selected as two important nuclides from low and intermediate level radioactive waste, and pumice tuff as one of the host rocks of such waste repositories. Pumice tuff blocks were prepared for this study expecting that the block sample is nearly the 'natural condition', like what is found in the field- compact with intact pores. Considering minimum dependency of ionic strength, initial nuclide concentration and pH,  $10^{-4}$  mol/l Cs and Sr concentration was selected as initial experimental concentration (Rajib *et al.*, 2015, 2016) where 4, 8 and 12 were chosen as low, neutral and high pH conditions. Since ionic strength significantly influences  $K_d$  at less than 1.0 mol/l on pumice tuff (Rajib *et al.*, 2011, 2016), relatively high ionic strength of 1.0 and 3.0 mol/l was adopted. To keep the effect of particle size at minimum, homogenous grain size of 150-300  $\mu\text{m}$  was used for the powdered materials, whereas blocks were prepared at an equal size of 1  $\text{cm}^3$ . Conventional batch technique was carried out with a solid-solution ratio of 1:10. However, as the ratio cannot be kept fixed for block samples (the weight varied from 1.090 to 1.456 g), adjustment to 1:10 ratio was necessary to recalculate  $K_d$  values. After reaching equilibrium pH at the contact time of 14 weeks, Cs and Sr concentrations were measured by ICP-MS, and the  $K_d$  values were determined considering the natural dissolution of Cs and Sr during aging period (Rajib *et al.*, 2015). Pumice tuff samples were collected from below 50-100 m at subsurface where water-rock interaction occurred and a redox zone has been formed. Hence, both fresh and oxidized pumice tuffs were possible to use to observe any oxidation effect. To investigate surface area properties of pumice tuff, mercury intrusion porosimetry (MIP) and observation through scanning electron microscope (SEM) was conducted.

The results show that  $K_d$  values for block samples are considerably lower than powdered samples for both Cs and Sr. Almost all the samples showed similar tendency with the average lower values of 8.79% for Cs and 4.94% for Sr in block samples of fresh tuff. In oxidized tuff, block samples showed 14.58% and 13.65% lower values for Cs and Sr, respectively. The lower  $K_d$  values on block samples might be due to the use of lower surface area as sorption sites since many closed pores exist in intact solid which cannot be accessed. The destruction of smaller pores in oxidized tuff due to oxidation phenomena might be the cause of higher reducing  $K_d$  values. However, use of pumice tuff blocks for comparable batch adsorption experiment is possible due to its highly porous nature where much amount of surface area can be accessed. The  $K_d$  difference between block and powder samples is expected to be caused mainly by the effect of diffusion, taking very long equilibrium period, and the physical and mechanical properties of pores and fractures during experiment.

References:

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