

Strontium adsorption behaviour of soils at NSDF location

Raghuveer Rao Palapati¹

¹Principal Research Scientist, Indian Institute of Science, Bangalore, India, email: pvr Rao@iisc.ac.in

ABSTRACT

Strontium being a major component of low level nuclear waste, its adsorption behaviour is studied at a Near Surface Disposal Facility in the presence of co-ions to understand its transport through vadose zone. The study conducted with soil at two locations of a proposed NSDF with depth through batch experiments with Strontium and in the presence of co-ions like Calcium, Magnesium, Sodium and Potassium which are commonly present in soil. The experiments conducted to study kinetics of adsorption (first and second order), adsorption intensity, free energy value, fitting the data in standard adsorption isotherms like Langmuir and Freundlich and evaluation of adsorption parameters. The experimental maximum adsorption values are compared with Isotherm fit parameters. The distribution coefficients for Strontium and in the presence of co-ions are evaluated to study the behaviour of adsorption. The study also includes different concentrations of co-ions and a combination of co-ions to study the combined effect. The relative replaceability of ions is studied based on their hydrated radii, gave the preference of ions for adsorption by soil studied. The study indicate Strontium adsorption is a favourable physical process based on isotherms, distribution coefficient, adsorption intensity. The study also brings out the reduction in Strontium adsorption due to presence of various co-ions individually and combined. The outcome of this study is important in the study of migration of radio nuclides to subsoil and groundwater in the event of leakages through Near Surface Disposal Facilities where low level nuclear waste is stored

Keywords: Strontium adsorption, isotherms, distribution coefficient, nuclear waste disposal, co-ions

1 INTRODUCTION

Radioactive strontium constitutes a major component in low level nuclear wastes (Huo et al., 2013). The long half-life of ⁹⁰Sr implies that contaminated soil and water will be threat to environment for decades (Zuo et al., 2009). The mobility of strontium depends on the adsorption-desorption behaviour. Strontium sorption by soil occurs via ion-exchange, precipitation and complexation reactions (Mell et al., 2006). Further, the extent of strontium sorption by the soil particles is influenced by pH of soil and groundwater, amount and type of clay fraction in the soil, organic content, metal oxides, co-ions and solids: water ratio (Bunde et al., 1997; Lee et al., 2000; Sahai et al., 2000; Bilgin et al., 2001; Murali and Mathur, 2002; Khan, 2003; Wang and An, 2004; Wang and Staunton, 2005; Tsukamoto et al., 2006; Zuo et al., 2009).

The solid-solute distribution coefficient (K_d) is also an important parameter in modelling transport of radionuclides in the subsurface (Zuo et al., 2009). Earlier researchers have measured strontium distribution coefficients through batch and column experiments. Porro et al. (2000) determined strontium distribution coefficients in crushed basalts through batch and column experiments. The researchers observed that the batch K_d values was lower than the column test values and decreased significantly with increasing solid:liquid ratio.

Earlier researchers have studied the adsorption of strontium ions on several geological media such as montmorillonite, kaolinite, goethite, hematite and silica colloids (Karasyova et al., 1999; Cole et al., 2000; Sahai et al., 2000; Jeong, 2001; Lu and Mason, 2001) and have observed that the strontium sorption capacities are dependent on the type of geological media under consideration. The clay mineralogy, pore-water chemistry and secondary mineral composition at each NSDF can vary depending on the local geological conditions. As strontium adsorption is intricately related to these factors, this paper examines the adsorption characteristics of strontium ions by NSDF soil samples. The adsorption isotherms, rate of adsorption and K_d parameters are evaluated for the soils using batch experiments.

The role of co-ions in influencing the strontium adsorption behaviour by the soils is also examined (Raghuvveer Rao, 2015).

2 MATERIALS AND METHODS

The study area is Kalpakkam which is situated about 70 km south of Chennai (12°33' N Lat and 80°11' E Long) along the east coast of India. Several of India's nuclear installations like Madras Atomic Power Station, Fast Breeder Test Reactor, Kalpakkam Reprocessing Plant and Indira Gandhi Centre for Atomic Research (IGCAR) are located in Kalpakkam. The proposed Near Surface Disposal Facility (NSDF) site at Kalpakkam is in the premises of IGCAR.

To evaluate the sub-soil at the proposed NSDF location for index, physico-chemical, mineralogical and geotechnical properties, two trial pits were excavated about 200 m apart, to a depth of about 3.2 m in the proposed NSDF area in May 2011. Soil samples were collected at every 1 m interval in each pit. A group of 3 thin walled 38 mm diameter tubes at spacing of 2 diameters were driven horizontally into the walls of the pit. Perforated wooden blocks were attached to the outer end of the tubes to prevent air pressure buildup and hit with a hammer. After driving the tubes, soil around the tubes was loosened with a sharp end steel rod. The soil collected around the tube is referred as representative sample and samples in the tubes are referred as undisturbed samples. Undisturbed samples were used to assess in-situ properties like density, degree of saturation, void ratio etc and representative sample for other tests like chemical analysis, pH, EC, adsorption and miscible displacement experiments. Ground water table was encountered at depth of 3.2 m during the collection of samples. The soil samples were preserved in light polyethylene bags which were sealed to prevent moisture loss and brought to the laboratory for testing. The physico-chemical properties are shown in Table 1.

Table 1 Physico-chemical properties of soil samples

	Sand (%)	Silt (%)	Clay (%)	pH	EC μ S/cm	TDS mg/L	CEC (meq/100g)	Organic matter (%)
Pit1-1m	66.08	20.02	13.9	7.72	543	348	11	1.45
Pit1-2m	62.22	22.28	15.5	7.71	716	459	15	1.66
Pit1-3m	66.39	18.51	15.1	7.80	809	519	21	1.74
Pit2-1m	62.07	24.23	13.7	8.40	250	160	11	1.19
Pit2-2m	64.33	20.47	15.2	7.86	190	122	11	1.33
Pit2-3m	71.33	16.27	12.4	8.44	344	221	20	1.71

3 STRONTIUM ADSORPTION STUDIES

3.1 Kinetics of Strontium adsorption

Figure 1 presents the kinetic plots showing equilibrium concentration with time (10, 30, 60, 120 and 240 minutes) for Pit 1 soil (initial strontium concentration = 300 mg/L) obtained for all the three depths (1 - 3 m). Though experiments are conducted on the soil from two pits namely Pit 1 and Pit 2, only the plots of Pit 1 are presented subsequently while the tables cover both the pits. The figure depict that the adsorption process is rapid and equilibrium adsorption is attained in 10 minutes of contact period.

As the first order fit had very poor correlation, second order rate equation was employed which can be represented as (Ho and McKay, 1999)

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (1)$$

where k_2 is the rate constant for second order sorption (g/mg.min), q_t is the amount of strontium adsorbed on sample at time t (mg/g) and q_e is the equilibrium sorption capacity (mg/g).

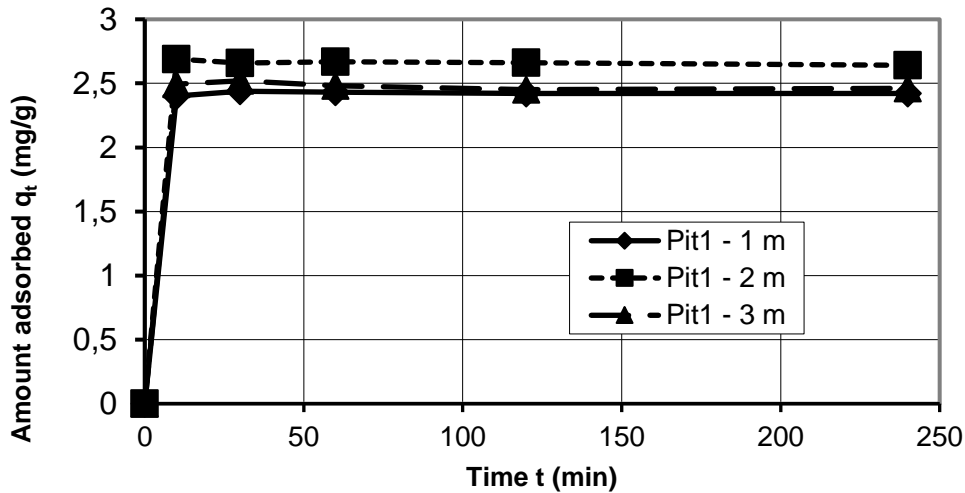


Figure 1. Variation of strontium adsorption with contact period for Pit 1

Figure 2 present the second order kinetic plots for strontium adsorption by the Pit 1 specimens from 300 mg/L strontium solution.

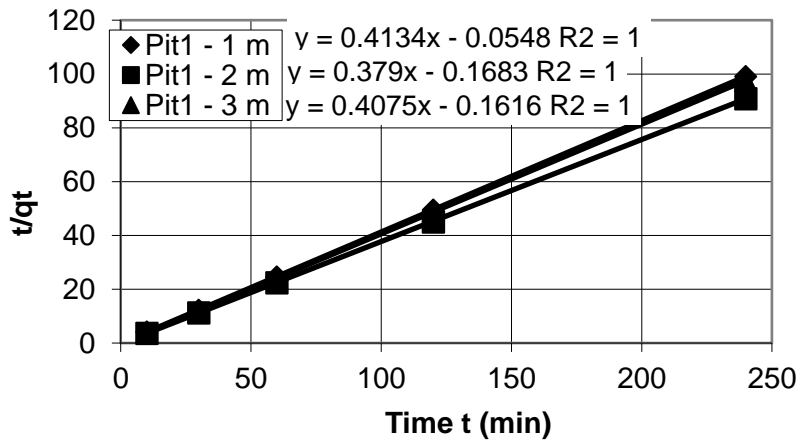


Figure 2. Second order kinetics fit plot for Pit 1

Obeying the second order model should yield a straight line with good correlation coefficient and secondly the graphical q_e should agree with the experimental q_e values. The graphical q_e values, rate constants and the corresponding experimental q_e values for Pits 1 and 2 soils are presented in Table 2. Plots (corresponding to strontium adsorption by Pit 1 soil) exhibit strong correlation coefficients of 1 for the Pits 1 and 2 soils. Consequently, the strong regression coefficients and the good agreement between graphical and experimental q_e values imply that strontium adsorption by Pits 1 and 2 soil specimens obey second order kinetics. There are two possibilities for second order reactions: the rate may be proportional to the product of two equal concentrations or to the product of two different ones (Laidler, 2011). As strontium adsorption depends on the number of exchange sites available on soil surface and the strontium ion concentration in solution, it is reasonable to assume that the rate is proportional to the concentration of two different reactants.

Table 2. Equilibrium adsorption from fit and experiment, second order rate constant and coefficient of determination

	q_e from fit plot (mg/g)	Rate constant k_2 (g/mg.min)	q_e from experiment (mg/g)	R^2
Pit1-1m	2.42	0.296	2.44	1
Pit1-2m	2.64	0.310	2.69	1
Pit1-3m	2.45	0.285	2.52	1
Pit2-1m	2.16	0.271	2.29	0.9997
Pit2-2m	2.40	0.278	2.44	1
Pit2-3m	2.25	0.275	2.30	0.9997

3.2 Adsorption isotherms

The strontium adsorption behaviour of the Pit 1 and Pit 2 representative soil samples is examined for the batch experiments data. Batches of representative sample of 5 grams was agitated with 100 mL strontium solution of concentrations 50, 100, 200 and 300 mg/L (solid:solution ratio of 1:20) for 240 minutes. The amount of strontium adsorbed at equilibrium, q_e (mg/g) is calculated as:

$$q_e = \frac{(C_0 - C_e) \times V}{M} \quad (2)$$

where C_0 is the initial concentration (mg/L), C_e is the equilibrium concentration (mg/L), V is the volume of the solution (L) and M is mass of soil (g).

The adsorption isotherms (plots of amount adsorbed vs equilibrium concentration) are presented in Figure 3 for Pit 1 specimens.

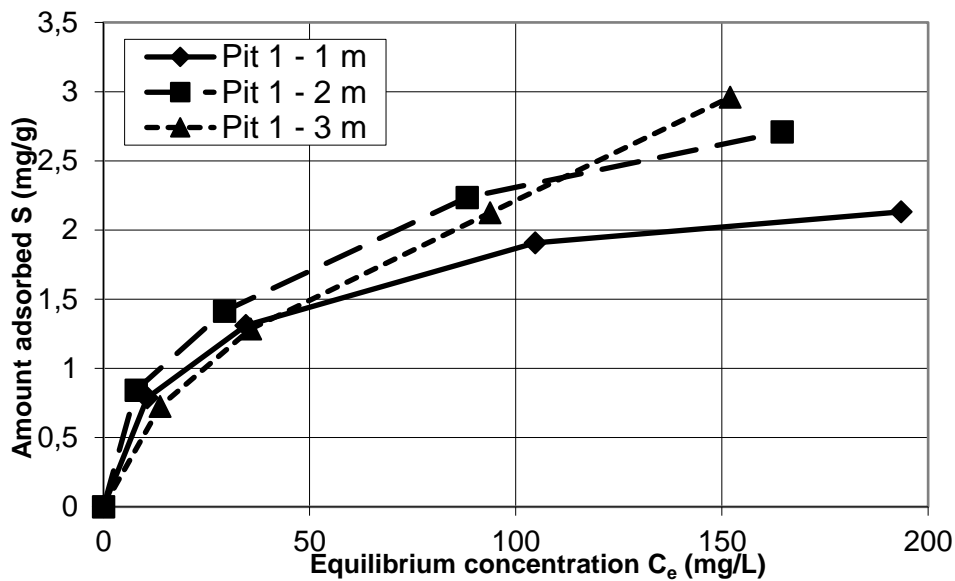


Figure 3. Variation of amount adsorbed with equilibrium concentration for Pit 1

The adsorption isotherms show that strontium adsorption by the soil samples tend towards equilibrium at C_e (equilibrium solute concentration) values > 100 mg/L ($C_0 = 200$ mg/L). The maximum strontium adsorption by Pit 1 soil samples range from 2.1 to 3 mg/g, while for the Pit 2 soils range from 2.1 to 2.5 mg/g. Huo et al. (2013) had performed batch adsorption studies using Chinese loess for much lesser range of initial strontium concentrations (0.76 to 27.8 mg/L); expectedly the authors observe much lesser strontium adsorption capacity (0.35 mg/g). Comparatively, Kamel (2010) had reported much larger strontium adsorption (12.5 mg/g) by Egyptian sandy clays ($C_0 = 250$ mg/L). The variability in strontium adsorption results of different researchers arise from variability in amount of strontium available for adsorption (C_0) and type of soil.

Langmuir and Freundlich Isotherms

The Langmuir isotherm (Langmuir, 1918) is represented as:

$$q_e = \frac{S_m b C_e}{1 + b C_e} \quad (3)$$

$$\text{or } \frac{1}{q_e} = \frac{1}{S_m} + \frac{1}{b S_m C_e} \quad (4)$$

where q_e is the amount adsorbed (mg/g), C_e is the equilibrium concentration (mg/L), b (L/mg) and S_m (mg/g) are the Langmuir parameters representing the adsorption equilibrium constant and the monolayer capacity.

The Freundlich isotherm is presented as:

$$q_e = K_f C_e^\varepsilon \quad (5)$$

where q_e is the amount adsorbed per unit mass of the adsorbent (mg/g), C_e is the equilibrium concentration of the solute (mg/L), K_f (mg/g) and ε are the Freundlich parameters.

The Langmuir parameters are determined by plotting $\frac{1}{q_e}$ vs $\frac{1}{C_e}$ and the Freundlich parameters are obtained by plotting $\log q_e$ vs $\log C_e$. The Langmuir isotherms plots are shown in Figure 4, while the Freundlich isotherm plots are presented in Figure 5.

It is observed from Table 3 that the maximum sorption capacities (S_m) from Langmuir isotherms range from 2.21 to 3.41 mg/g for Pit 1 soils and from 2.11 to 2.35 mg/g for the Pit 2 soils. The Langmuir sorption capacities (S_m) are close to the experimentally obtained maximum adsorption. The K_f values from the

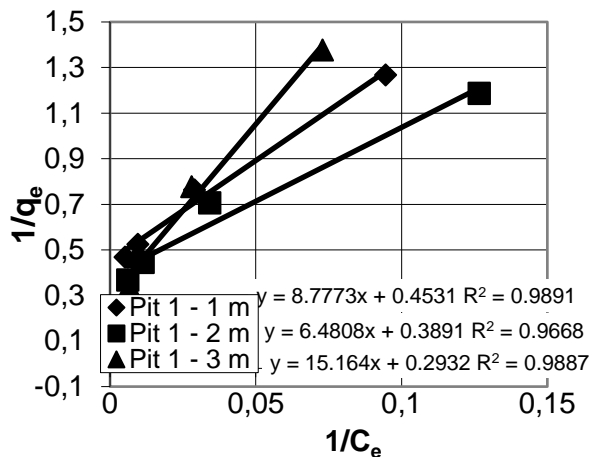


Figure 4. Langmuir isotherm for Pit 1

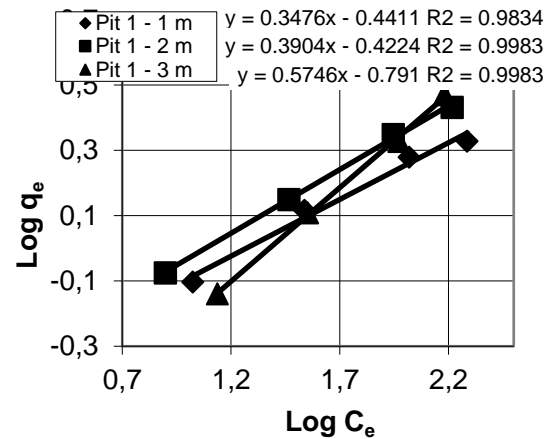


Figure 5. Freundlich isotherm for Pit 1

Table 3. Freundlich and Langmuir parameters for Pit 1 and 2 soils

	Langmuir parameters		Freundlich parameters		Max. adsorption experimental value
	S_m	b	K_f	ε	
Pit 1 - 1 m	2.21	0.054	0.362	0.348	2.13
Pit 1 - 2 m	2.57	0.060	0.378	0.390	2.71
Pit 1 - 3 m	3.41	0.019	0.162	0.575	2.96
Pit 2 - 1 m	2.15	0.064	0.395	0.333	2.16
Pit 2 - 2 m	2.11	0.061	0.406	0.319	2.03
Pit 2 - 3 m	2.35	0.039	0.251	0.438	2.51

Freundlich plots represent the adsorption capacities and range from 0.162 to 0.378 mg/g for Pit 1 soils and from 0.251 to 0.406 for Pit 2 soils. The Freundlich isotherms predict much larger range of sorption capacities than Langmuir isotherm and the experimental values. The ε values range from 0.348 to 0.575 for Pit 1 soils and 0.333 to 0.438 for Pit 2 soils. The range of ε values (< 1) indicate that the strontium adsorption is a favourable physical process (Crini et al., 2007).

3.3 Adsorption Intensity

The adsorption intensity (Separation factor) is defined as

$$R_L = \frac{1}{(1+bC_{ref})} \quad (6)$$

where b is Langmuir constant related to energy of adsorption (L/g) and C_{ref} is any initial strontium concentration. The value of R_L indicates the nature of adsorption: $R_L > 1$ implies unfavourable adsorption, $0 < R_L < 1$ implies favourable adsorption and $R_L = 0$ implies irreversible adsorption (McKay et al., 1982). The R_L values are plotted as function of initial strontium concentration for the Pit 1 (Figure 6) soils. The plot show R_L values < 1 indicating that the strontium adsorption is favoured process.

Further, the larger R_L values at lower C_0 imply that the adsorption is more favoured at lower surface coverage.

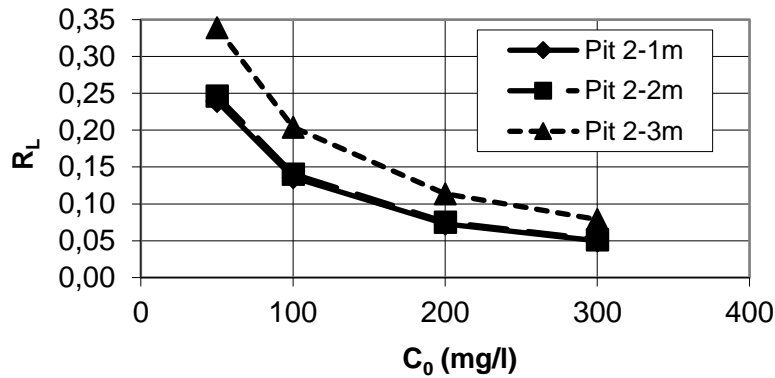


Figure 6. Variation of adsorption intensity with initial strontium concentration for Pit 1 soils

3.4 Free Energy Change

Free energy change (KJ/mol) is given by

$$\Delta G = -RT \ln S_m \tag{7}$$

where R is universal gas constant, T is temperature and S_m is Langmuir maximum sorption capacity. The ΔG values for the Pit 1 and Pit 2 soil specimens are shown in Table 4.

Table 4. Free energy change values, ΔG

	Free energy change KJ/mol
Pit 1 – 1m	-1961
Pit 1 – 2m	-2338
Pit 1 – 3m	-3039
Pit 2 – 1m	-1893
Pit 2 – 2m	-1853
Pit 2 – 3m	-2117

The negative ΔG values indicate the spontaneous nature of strontium adsorption and confirms the affinity of solute for the soil particles (Namasivayam and Ranganathan, 1995; Wang et al., 2011).

3.5 Distribution Coefficient

The distribution coefficient or partition coefficient K_d (L/g) is calculated as

$$K_d = \frac{q_e}{C_e} \tag{8}$$

where q_e is the amount adsorbed (mg/g) and C_e is equilibrium concentration (mg/L).

The variations of K_d (mL/g) as function of initial strontium concentration is given in Table 5.

Table 5. Variations of K_d (mL/g) with initial strontium concentration

	Initial concentration of strontium (mg/L)			
	50	100	200	300
Pit 1 – 1m	74.52	21.79	8.50	2.54
Pit 1 – 2m	106.58	26.75	13.96	6.19
Pit 1 – 3m	52.89	25.48	14.47	14.32
Pit 2 – 1m	86.87	20.01	9.15	2.55
Pit 2 – 2m	81.44	21.45	8.30	1.34
Pit 2 – 3m	63.96	16.68	10.07	8.99

The K_d values range from 106.58 to 2.5 mL/g for Pit 1 samples and from 86.87 to 1.34 mL/g for Pit 2 samples. The reduction in K_d with increasing C_0 values imply that higher adsorption is preferred at lower concentrations and the adsorption decreases with increase in initial concentration.

4 DISTRIBUTION COEFFICIENT IN PRESENCE OF CO-IONS

The strontium adsorption characteristics of the Pit 1 and 2 soil specimens are examined in presence of co-ions encountered in the soil pore water of the pit 1 and 2 specimens. The co-ion concentrations used in the batch experiments are based on the cation concentration in the soil-pore water extracts of Pit 1 and 2 soil samples (sampling depths 1-3 m, Table 1). The cation concentrations used in the batch experiments are provided in Table 6.

Table 6. Co-ion concentrations used in batch experiments

Calcium, mg/L	50	100	600	
Magnesium, mg/L	20	50	100	150
Sodium, mg/L	200	300	500	700
Potassium, mg/L	1	5	10	20

Figure 7 plot the strontium adsorption by Pit 1 soil samples in presence of calcium ions (50, 100 and 600 mg/L). The plot shows that strontium adsorption tendency reduces with increase in calcium ion concentration. The K_d parameter reduces from 27.2, 18.8 to 3.3 mL/g with increase in calcium ion concentration from 50, 100 to 600 mg/L for Pit 1 soil. Figure 8 plot the strontium adsorption by Pit 1 soil samples in presence of magnesium ions (20, 50, 100 and 150 mg/L). The K_d parameter reduces from 25.7, 16.9, 10.6 to 6.8 mL/g with increase in magnesium ion concentration from 20, 50, 100 to 150 mg/L for Pit 1 soil.

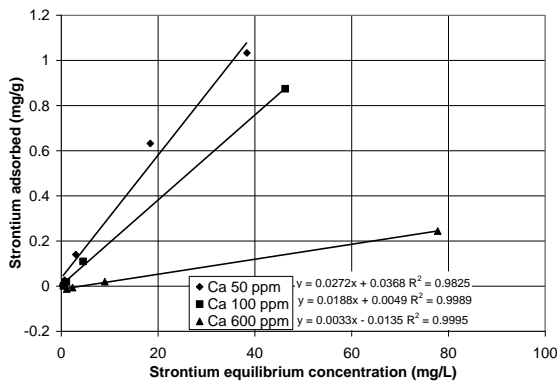


Figure 7. Strontium adsorption by Pit 1 soil in presence of calcium ions

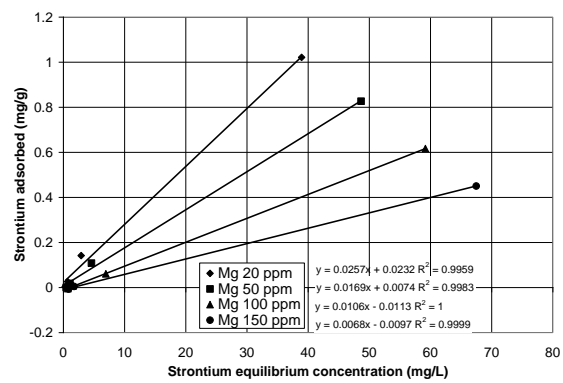


Figure 8. Strontium adsorption by Pit 1 soil in presence of magnesium ions

Figure 9 plot the strontium adsorption by Pit 1 soil sample in presence of sodium ions (200, 300, 500 and 700 mg/L). The K_d parameter reduces from 34.5, 27.3, 23.8 to 16.4 mL/g with increase in sodium ion concentration from 200, 300, 500 to 700 mg/L for Pit 1 soil. Figure 10 plot the strontium adsorption

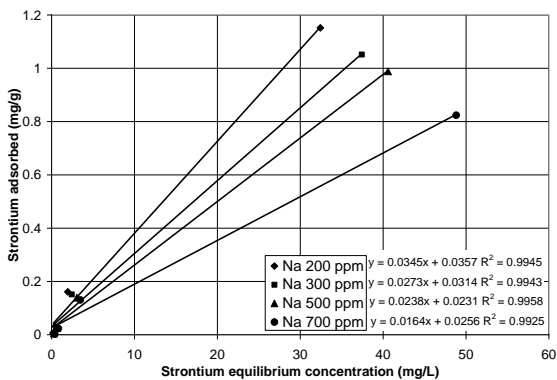


Figure 9. Strontium adsorption by Pit 1 soil in presence of sodium ions

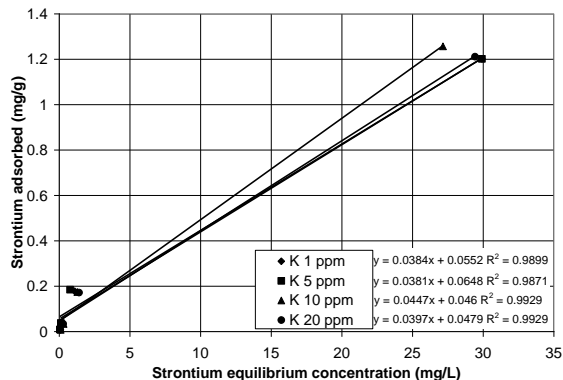


Figure 10. Strontium adsorption by Pit 1 soil in presence of potassium ions

by Pit 1 soil samples in presence of potassium ions (1, 5, 10 and 20 mg/L). The K_d parameter is unaffected in presence of potassium co-ions and range from 38.4, 38.1, 44.7 and 39.7 mL/g with increase in K ion concentration from 1, 300, 5, 10 to 20 mg/L for Pit 1 soil.

The relative replaceability of exchangeable cations is described by the lyotropic series and for calcium, magnesium and strontium ions, it follows the sequence, Mg^{2+} (0.428) > Ca^{2+} (0.412) > Sr^{2+} (0.412) where the values in parentheses represent the hydrated radii in nanometer (Essington, 2004). As calcium and magnesium ions are more favoured than strontium ions for adsorption by the soil surface, the reduction in K_d for strontium adsorption in presence of calcium and magnesium ions is understandable. Sodium ions have smaller hydrated radius (0.358 nm) than strontium ion (0.412 nm). Reduction in K_d for strontium adsorption in presence of sodium ions is ascribed to the concentration effect, where owing to the large concentration of sodium ions, the affinity of strontium adsorption by soil surface reduces.

Figure 11 summarize the variations in K_d with initial co-ion concentration for Pit 1 soil samples. In accordance with the lyotropic series, at given co-ion concentration, K_d parameter for strontium adsorption follows the sequence $K_{d, Sr,K} > K_{d, Sr,Na} > K_{d, Sr,Ca} > K_{d, Sr,Mg}$ for soil samples from Pit 1 and similar observation made for Pit 2 also. The findings of the study attach significance for the safety assessment of NSDF in case of accidental conditions.

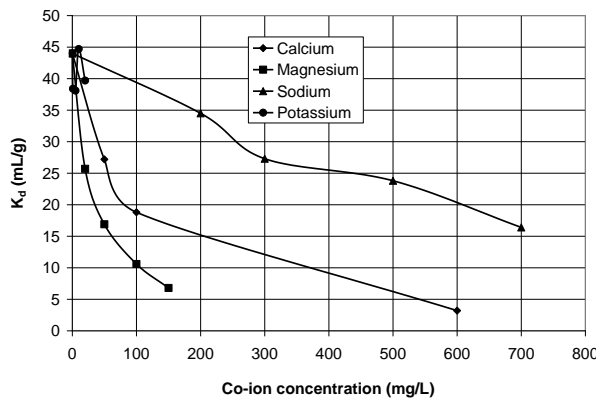


Figure 11. Variation of K_d with initial co-ion concentrations for Pit 1 soil

Figure 12 illustrate the influence of mixture of co-ions on strontium adsorption ability of Pit 1 soil. Two co-ion mixes were considered, representing low and high cation concentrations that can be encountered in the NSDF soil pore-water. The low co-ion mix comprised of 20, 200, 200 and 1 mg/L of Mg, Ca, Na and K ions respectively. The high co-ion mix comprised of 350, 600, 700 and 20 mg/L of Mg, Ca, Na and K ions respectively.

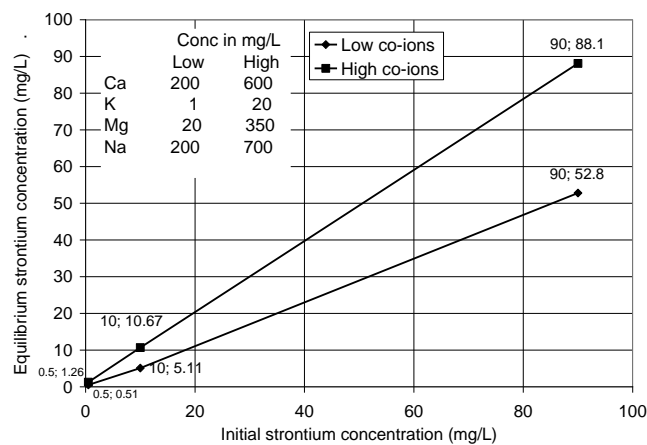


Figure 12. Strontium adsorption by Pit 1 soil in presence of co-ion mix: Values on plots indicate initial and equilibrium strontium concentrations in solution

Plots in Figure 12 show nearly 50% strontium adsorption at initial strontium concentrations of 90 and 10 mg/L in low co-ion matrix. No strontium adsorption by Pit 1 soils occur in high co-ion matrix. Earlier Deepthi and Sasidhar (2012) have also observed that the strontium distribution coefficient (K_d) for Kalpakkam soils decreases in presence of calcium and sodium ions. The authors attributed the reduction in K_d to charge neutralization of the kaolinite surface.

5 CONCLUSIONS

The present paper examines the adsorption behaviour of strontium ions by NSDF soil samples. The adsorption isotherms, rate of adsorption and K_d parameters are evaluated for the soils using batch experiments. The role of co-ions in influencing the strontium adsorption behaviour of the soils is also examined. Strontium adsorption by the soil specimens obeyed second order kinetics and the rate is apparently influenced by the number of exchange sites available on soil surface and available strontium ion concentration in solution. Validation of second order kinetics was probed by fitting the experimental data into non-linear and linear forms of second order equation. The strontium adsorption at equilibrium (q_e) ranges from 2.5 to 2.8 mg/g for Pit 1 soil specimens and from 2.4 to 2.5 for Pit 2 soil specimens. The experimental maximum strontium adsorption of Pit 1 soil samples range from 2.1 to 3 mg/g, while for the Pit 2 soils range from 2.1 to 2.5 mg/g; the experimental values compare reasonably with theoretical values from second order equation. The second order rate constants range from 0.285 to 0.31 (g/mg.min) for Pit 1 soils and from 0.271 to 0.278 (g/mg.min) for Pit 2 soils.

Strontium adsorption by the Pit 1 and 2 soils also obeyed the Langmuir and Freundlich isotherms. The maximum sorption capacities (S_m) from Langmuir isotherms range from 2.21 to 3.41 mg/g for Pit 1 soils and from 2.11 to 2.35 mg/g for the Pit 2 soils. The Langmuir sorption capacities (S_m) are close to the experimental maximum adsorption. The Freundlich isotherms predict much larger range of sorption capacities than Langmuir isotherm and experimental values. The range of ϵ values (0.348 to 0.575 for Pit 1 soils and 0.319 to 0.438 for Pit 2 soils) indicated that strontium adsorption is a favourable physical process. The R_L (adsorption intensity) values ranged from 0.5 to 0.07 for strontium adsorption by Pit 1 soils and from 0.34 to 0.05 for Pit 2 soils. R_L values less than unity imply that strontium adsorption by the NSDF soils is favourable process; further, the larger R_L values at lower C_0 (initial strontium concentration) implied that the adsorption is more favoured at lower surface coverage.

The free energy change for strontium adsorption ranged from -1961 to -3039 KJ/mol for Pit 1 soils, and from -1853 to -2117 KJ/mol for Pit 2 soils, implying that strontium adsorption by the soils is spontaneous process.

The K_d (distribution coefficient) parameter for strontium adsorption by NSDF soils were examined for samples collected at 2 m depth from Pits 1 and 2 respectively. The tendency for strontium adsorption reduces with increase in Ca ion concentration. The K_d parameter reduces from 27.2, 18.8 to 3.3 mL/g with increase in Ca ion concentration from 50, 100 to 600 mg/L for Pit 1 soil. Likewise, the K_d parameter reduces from 23.2, 15.5 to 3.1 mL/g for similar increase in Ca ion concentration for Pit 2 soil. The K_d parameter reduces from 25.7, 16.9, 10.6 to 6.8 mL/g with increase in Mg ion concentration from 20, 50, 100 to 150 mg/L for Pit 1 soil and reduces from 22.9, 14.6, 9.4 to 6.1 mL/g for similar increase in Mg ion concentration for Pit 2 soil. The K_d parameter reduces from 34.5, 27.3, 23.8 to 16.4 mL/g with increase in Na ion concentration from 200, 300, 500 to 700 mg/L for Pit 1 soil and from 32.2, 26.1, 25.6 to 21.4 mL/g with similar increase in Na ion concentration for Pit 2 soil.

Reduction in K_d for strontium adsorption in presence of Ca and Mg ions is attributed to the more favored adsorption of these divalent cations than strontium ions by the soil surface. Reduction in K_d for strontium adsorption in presence of Na ions is ascribed to the concentration effect, where owing to the large concentration of Na ions, the affinity of strontium adsorption by soil surface reduces.

The K_d parameters for strontium adsorption by the NSDF soils were also evaluated in the presence of mixture of co-ions. The low co-ion mix comprised of 20, 200, 200 and 1 mg/L of Mg, Ca, Na and K ions respectively. The high co-ion mix comprised of 350, 600, 700 and 20 mg/L of Mg, Ca, Na and K ions respectively.

Experimental results showed nearly 50% strontium adsorption at initial strontium concentrations of 90 and 10 mg/L in low co-ion matrix, while, no strontium adsorption by Pit 1 and 2 soils occurred in high

co-ion matrix. The K_d values ranging from 14.1 to 19.1 mg/L for pit 1 soils and 13.4 to 18.1 mg/L for pit 2 soils were observed for batch experiments in low co-ion mix.

REFERENCES

- Bilgin, B., Atun, G. and Keçeli, G. (2001) Adsorption of strontium on illite. *Journal of Radioanalytical and Nuclear Chemistry*. 250:323-328.
- Bunde, R.L., Rosentreter, J.J., Liszewski, M.J., Hemming, C.H. and Welhan, J. (1997) Effects of calcium and magnesium on strontium distribution coefficients. *Environ. Geol.* 32:219–229.
- Cole, J. E., R. B. Dunbar, T. R. McClanahan, and N. A. Muthiga, (2000) Tropical Pacific Forcing of Decadal SST Variability in the Western Indian Ocean over the Past Two Centuries. *Science*. 287:617– 619.
- Crini, G., Peindy, H.N., Gimbert, F. and Robert, C. (2007) Removal of C.I. basic green 4 (Malachite green) from aqueous solutions by adsorption using cyclodextrin-based adsorbent: Kinetic and equilibrium studies. *Separation Purification Technology*. 53:97-110.
- Deepthi Rani, R. and Sasidhar, P. (2012a) Geochemical and thermodynamic aspects of sorption of strontium on kaolinite dominated clay samples at Kalpakkam. *Environ. Earth Sci.* 65:1265–1274.
- Essington, M.E. (2004) *Soil and water chemistry*, CRC Press, Boca Raton, FL.
- Ho, Y.S. and McKay, G. (1999) Pseudo second order model for sorption processes, *Process Biochem.* 34:451-465.
- Huo, L., Qian, T., Hao, J. and Zhao, D. (2013) Sorption and retardation of strontium in saturated Chinese loess: experimental results and model analysis. *Journal of Environmental Radioactivity*, 116:19-27.
- Jeong, C.H. (2001) Mineralogical and hydrochemical effects on adsorption removal of Cs-137 and Sr-90 by kaolinite. *J. Environ. Sci. Health A Toxicol. Hazard. Subst. Environ. Eng.*, 36:1089-1099.
- Kamel, N.H.M. (2010) Adsorption models of ^{137}Cs radionuclide and Sr(II) on some Egyptian soils. *J. Environ. Radioact.* 101:297-303.
- Karasyova O. N., Ivanova L. I., Lakshatanov L. Z. and Lovgren L. (1999) Strontium sorption on hematite at elevated temperatures. *J. Coll. Interf. Sci.* 220:419–428.
- Khan, S.A. (2003) Sorption of the long-lived radionuclides cesium-134, strontium-85 and cobalt-60 on bentonite. *Journal of Radioanalytical and Nuclear Chemistry* 258:3-6.
- Laidler, K.J. (2011) *Chemical Kinetics*, 3/e Pearson Education.
- Langmuir I. (1918) The adsorption of gases on plane surfaces of glass, mica and platinum. *J. Am. Chem. Soc.*, 40(9):1361-1403.
- Lee, M.H. and Lee, C.W. (2000) Association of fallout-derived ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ with natural organic substances in soils, *Journal of Environmental Radioactivity*, 47:253-262.
- Lu, N.P. and Mason, C.F.V. (2001) Sorption-desorption behavior of strontium-85 onto montmorillonite and silica colloids. *Applied Geochemistry* 16:1653-1662.
- McKay, G., Blair, H.S. and Gardener, J.R. (1982) Adsorption of dyes on chitin: 1 Equilibrium studies. *J. Appl. Polym. Sci.* 27:3043-3057.
- Mell, P., Megyeri, J., Riess, L., Máthé, Z., Csicsák, J. and Lázár, K. (2006) Sorption of Co, Cs, Sr and I onto argillaceous rock as studied by radio tracers. *Journal of Radioanalytical and Nuclear Chemistry* 268:405-410.
- Murali, M.S. and Mathur, J.N. (2002) Sorption characteristics of Am(III), Sr(II) and Cs(I) on bentonite and granite. *Journal of Radioanalytical and Nuclear Chemistry* 254:129-136.
- Namasivayam, C. and Ranganathan, K. (1995) Removal of Cd (II) from wastewater by adsorption on waste Fe (III)/Cr (III) hydroxide. *Water Research*, 29:1737-1744.
- OECD (2003) *Engineered barrier system and the safety of deep geological repositories. State of the art report*, ISBN 92-64-18498-8.
- Porro, I., Newman, M.E. and Dunnivant, F.M. (2000) Comparison of batch and column methods for determining strontium distribution coefficients for unsaturated transport in basalt. *Environmental Science and Technology* 34:1679-1686.
- Raghuveer Rao, P. (2015) Factors influencing contaminant transport in vadose zone of near surface radioactive waste disposal facility. Ph D thesis, Indian Institute of Science, Bangalore, India.
- Sahai, N., Carroll, S.A., Roberts, S. and O'Day, P.A. (2000) X-ray adsorption spectroscopy of strontium (II) coordination. II. Sorption and precipitation at kaolinite, amorphous silica and goethite surfaces. *J. Colloid Interf. Sci.* 222:198-212.
- Tsukamoto, M., Fujita, T. and Ohe, T. (2006) Sorption and diffusion behaviours of strontium in sodium-type bentonite bed. *Czechoslovak Journal of Physics* 56:119-128.

- Wang, G. and Staunton, S. (2005) Evolution of Sr distribution coefficient as a function of time, incubation conditions and measurement technique, *J. Environ. Radioact.* 81:173-185.
- Wang, S., Nan, Z., Cao, Z., Liao, Q., Liu, J., Wu, W., Zhou, T., Zhao, C and Jin, W. (2011) Sorption and desorption behaviour of lead on a Chinese kaolin. *Environmental Earth Sciences*, 63:145-149.
- Wang, Z.M. and An, Y.F. (2004) Migration characteristics of ⁸⁵Sr in unsaturated loess. *Atomic Energy Science and Technology* 38:29-34.
- Zuo, R., Teng, Y.G. and Wang, J.S. (2009) Sorption and retardation of strontium in fine particle media from a VLLW disposal site. *Journal of Radioanalytical and Nuclear Chemistry* 279:893-899.