

INORGANIC AND BIO-MATERIALS IN THE REMOVAL/SPECIATION OF RADIOCESIUM AND RADIOSTRONTIUM : AN OVERVIEW

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Abstract : Cesium and strontium are two important fission products and the removal/speciation of these two cations with several inorganic/bio-materials is an interesting topic to discuss. It is to be noted that inorganic materials are found to be fairly effective and efficient in the removal/speciation of these cations. Moreover, these solids are to be found promising as they show fairly good radiation and temperature stability. Hence, they play an important role in the radioactive waste management studies. However, various studies reveal that in addition to inorganic materials various biosorbents can also be employed in the removal/speciation of these ions. But the radiation and temperature stability of these bio-sorbents and dead biomasses are still the topic lying among the researchers to be investigated.

Key Words : Sorption, Radiocesium, Radiostrontium, Fission Products, Biosorbents, Ion-Exchangers, Clay Minerals

INTRODUCTION

The development and welfare of modern society depends to a large extent upon the contribution of technology and industrial processes such as the generation and wide spread use of electricity. These processes are, in general, associated with the production of wastes, some of which are unavoidable, unrecyclable and hazardous. Such wastes require careful management to ensure adequate protection of humans and the environment. The timescales over which such protection is required can extend in the case of wastes containing toxic chemicals, elements or long-lived radioactive isotopes, well beyond the life spans

of current or forthcoming generation, i.e., many thousands of years into the future. Hence, there is an ethical importance to care about future generations and to act in such a way as to preserve, as much as possible, their options to enjoy and benefit from the earth resources. Hence, the World Commission on Environment and Development, the Brundland Commission 1987¹⁾ has observed such concern and suggested 'the protection of human health with sustainable development' has to be put forward. This concept which, is principally an ethical one, was defined as "satisfying the needs of present, without compromising the ability of future generations to meet their own needs". In this regard the potential health effects from the disposal of any type of wastes are dependent on a combination of several factors related to these differences,

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which poses a challenging problem for comparative assessment. The factors include the amount of the wastes, the toxicity and the environmental conditions and demographics/habits associated with people living near the disposal site.²⁾

Nuclear applications produce different types of radioactive wastes. In terms of volume, most of it is 'low-level radioactive wastes' that is disposal of in facilities just below the earth's surface. More than 100 of these shallow disposal facilities are already available and more than 30 are under development worldwide. They are able to receive low-level wastes from nuclear power plants/research reactors as well from medical, industrial and other research activities.

The situation is different for high-level wastes, either spent fuel or its reprocessed wastes, which must be safely isolated for millennia. The scientific and technical community generally agrees that disposal for this relatively low volume but highly radioactive waste can be carried out in stable geological formations, such as ancient salt domes or granite tunnels several hundred meters below surface. Multiple natural and engineered barriers would protect against human intrusion and ensure long-term confinement. However, no concept for the long-term disposal of commercial high-level-waste has been licensed in any country. One important step towards demonstrating the disposal concept was the opening of a Waste Isolation Pilot Plant (WIPP), March 1999, in the United States. Located 700 meters deep in a salt formation, WIPP is the World's first geological repository certified for disposal of long-lived radioactive wastes. The site is approved for receiving US defense-related wastes, and is not licensed for high-level-waste disposal. Technological developments in the nuclear fuel cycle may help address some concerns. At the La Hague reprocessing plant in France, for example, new volume reduction techniques is being adopted as the waste from spent fuel is to be converted into vitrified solid high-level wastes. If the World's annual generation of spent fuel could be reprocessed with equivalent volume reductions,

rough estimates are that the resulting vitrified solids would be of the order of 1,000 cubic meters, that is a cube of about 10 meters on each side per year of the World nuclear energy production.

Similarly it is to be reported that US Department of Energy is developing an integrated nuclear fuel cycle technology under its Advanced Fuel Cycle Initiative (AFCI) and under the AFCI, waste minimization is stressed. Engineered product storage materials will be required to store concentrated radioactive cesium, strontium, americium and curium for periods of tens to hundreds of the years.^{3,4)} The AFCI stressed towards the burned UO_2 fuel will undergo extensive processing to (1) recycle fissionable material for continued burning in-reactor, (2) recover long-lived radionuclides for transmutation in transmutation reactors being designed under AFCI, (3) isolate high-specific-activity cesium and strontium isotopes for decay storage in monitored low-level storage facilities, and (4) stabilization of the much-reduced volume of waste material designed for geological disposal within the Yucca Mountain underground repository.⁵⁾

The low and intermediate-level radioactive wastes arising from the operation of nuclear power plants can be immobilized by various solidification techniques prior to disposal. Liquid radioactive wastes have been treated with filtration, ion exchange resin, evaporation, and so on. The remaining liquid concentrate wastes from the evaporator have to be immobilized by solidifying agents such as cement, bitumen and polymer. Because waste forms, including radionuclides viz., radio cesium, cobalt, strontium mainly are immobilized with solidifying agents, they can be safely isolated from the biosphere for a long time in a radioactive repository. However, despite the engineering and natural barriers of radioactive waste repositories, the radioactive waste forms would eventually be in contact with groundwater, and the release of radioactive species from the waste forms would occur by the leaching mechanism. Therefore, acceptance criteria of radioactive

waste forms have to be developed to guarantee the long-term safety performance of a radioactive waste repository.⁶⁻⁸⁾

The spent fuel constitutes a significant amount of radio cesium and radio strontium and it contains more than 90% of radioactivity in the bulk of the spent fuel. It is to be reported that Cesium-137 and Sr-90 have the yield of *ca* 5.9 and 5.8% respectively from 30 days cooling of ²³⁵U fission at 1MW power level.⁹⁾ Hence, the cesium and strontium isotopes will require to be isolated from the spent fuel prior to the solidification and stabilization for prolonged decay storage of *ca* 300-500 years. They too have high-specific-activity from their decay (¹³⁷Cs $E_{\beta} = 0.514$ MeV; ⁹⁰Sr $E_{\beta} = 0.546$ MeV) and the decay of their short-lived daughter products (^{137m}Ba $E_{\gamma} = 0.662$ MeV; ⁹⁰Y $E_{\beta} = 2.28$ MeV). Because of the unique endpoint of these radionuclides the management of these radionuclides in the spent fuel is a greater concern for their handling and storage point of view. Further, it is also to be noted that these two radiotoxic ions have their wide applications in the area of medicines,¹⁰⁻¹¹⁾ food-irradiations¹²⁻¹⁴⁾ and sewage treatment technologies as well.¹⁵⁻¹⁷⁾ Hence, recently there seems to be a renewed interest as these are not waste products but are useful products hence their recovery from radioactive wastes is also needed.¹⁸⁾ Chemically these two alkali and alkaline earth metals are very stable in nature hence poses serious problems for their isolation from the radioactive wastes as by solvent extraction method. Therefore, the role of various inorganic/bio-materials is relatively an appropriate choice for such purposes¹⁹⁻²⁰⁾ and indeed various studies were already performed for the removal/speciation behavior of various such materials for cesium and strontium. Hence, with increased interest it prompted us to review the literature for the effective and efficient removal/speciation and sorption behavior of various kinds of inorganic/bio-materials for cesium and strontium and also to analyze critically their role in radioactive waste management studies.

RADIOCESIUM AND RADIOSTRONTIUM IN THE ENVIRONMENT

The nuclear weapon race during the cold war and the increased level of nuclear power plants resulted in several environmental concerns. Significant quantities of radioactive contaminants were not only stored at several sites but also released into the environment.²¹⁻²³⁾ One of the effective strategies to minimize the nuclear waste volumes for final disposal is the selective separation of ⁹⁰Sr and ¹³⁷Cs.²⁴⁾ However, the low concentration of these radioactive elements in a solution that has high sodium ion content (up to 6M) and alkalinity, renders this a daunting task.

Moreover, the April 26, 1986, one of the four reactors in the nuclear power station near the small Ukrainian town of Chernobyl exploded and started to burn. In the following days and weeks winds drove air layers containing radioactive elements from the collapsed reactor over a large part of Europe. With the rain these substances were washed out and deposited on plants and on the soil surface. Following the summer and autumn was reported to be the high level of radioactivity in fruits, berries, nuts and some processed fruit products in Switzerland.²⁵⁾ This increase in radioactivity is mainly due to the presence of Cs-137 and Cs-134. The literature shows that the interactions of cesium with soil particles are similar to those of potassium.²⁶⁾ Its availability for crop plants depends on soil properties, e.g., soil texture, clay mineralogy, contents of other cations, pH. Similarly, airborne radionuclides can be taken up by plants and thus transferred to the food chain. Radioactive particles can contaminate plants either by foliar deposition or by uptake from the soil *via* roots; during periods with high fallout rates, direct plant contamination might be greater than root contamination.²⁷⁻²⁹⁾ It is also to be noted that cesium is easily trans-located from the contaminated leaves to the edible parts of crop plants.³⁰⁻³²⁾ Since the radiocesium shows similar behavior to

potassium in the soil, therefore cesium availability in soil solution and its uptake into plant roots are quite low.³³⁻³⁵⁾ Similarly the strontium is almost biologically immobile; it is redistributed in the plant only to a very small extent.^{28,31,32,36-39)} Hence, the interactions of strontium with soil are comparable with those of calcium. Therefore, it is quite readily available to plants.⁴⁰⁻⁴¹⁾ The foliar contamination of *Phaseolus vulgaris* with aerosols of ¹³⁷Cs, ⁸⁵Sr, ¹³³Ba and ^{123m}Te were studied as an influence of plant development stage upon contamination and rain.⁴²⁾

Radioactive strontium and cesium also contained in the low level radioactive wastes hence as these wastes⁴³⁾ are usually disposed off in shallow land disposal sites, may perhaps cause for its possible migration into the biosphere through the diffusive mobilization through the soil layers. Similarly, it was reported that the United States accumulated large amount of the radioactive wastes at the Hanford site in south-eastern Washington⁴⁴⁻⁴⁵⁾, approximately 65 million gallons of highly radioactive wastes are contained in 177 single- and double-shell steel-lined tanks. Because some tanks are known to leak into the surrounding environment, and largely it contains the fission products ⁹⁰Sr and ¹³⁷Cs.

INORGANIC MATERIALS IN REMOVAL/SPECIATION OF CESIUM & STRONTIUM

Inorganic ion exchangers are found to be promising in the radioactive waste management studies as they show very high selectivity and also the capability to retain the radionuclide. Moreover, they possess fairly good radiation and thermal capacities.⁴⁶⁻⁴⁷⁾ Literature survey reveals that variety of inorganic materials employed for the removal/speciation of radio cesium and radio strontium. Hence, we try to focus the properties and their applicability of these materials in the following sections.

Oxides of Tetravalent Metals and Their Derivatives

Hydrous zirconium, titanium and silicon oxides are potentially used in the removal of various radionuclides as due to their exceptionally high stability, selectivity and resistance towards radiation hence, would find significant applicability in the radioactive waste management studies. Moreover, they can be vitrified or cermized to unleachable solid form for the ultimate disposal. The single phase assemblies for strontium fixation in zirconia,⁴⁸⁾ titania⁴⁹⁾ and conversion of zirconia to zircon were suggested for radioactive waste disposal. The presence of 10⁻² M NaCl and at acidic pH range suggested to be effective for the removal of these two fission products from the liquid wastes by titanium dioxide.⁵⁰⁾ Zirconium phosphate was assessed for the adsorption of ⁽⁸⁵⁺⁸⁹⁾Sr from aqueous solutions and it was reported that the irreversible type uptake is favored with the increase of sorptive concentration, temperature and pH.⁵¹⁾ ⁹⁰Sr was removed from the milk by using titanium phosphate, zirconium phosphosilicate and sodium titanate or the combinations of these.⁵²⁾ Further, the granular form of the materials are useful for the removal/speciation of these cations and also can be served as filter medium. Hence in a quest to prepare and apply the granular materials various investigators synthesized the derivatives of these metals and successfully applied for to achieve the better efficiency. In this line, the composites of zirconium *viz.*, zirconium molybdoarsenate⁵³⁻⁵⁴⁾, zirconium molybdoantimonate⁵⁵⁾, mixture of zirconium molybdate and zirconium tungstate⁵⁶⁾ were prepared and successfully employed in the removal of ¹³⁷Cs.

Silicates are widely employed for the speciation of these two fission products. The group of Clearfield at Texas A & M University, USA long back synthesizing and characterizing various kind of silicates and have reported their applicability in radioactive waste management studies. Titanium silicates, M₃HTi₄O₄(SiO₄)₃.4H₂O (M= Na⁺, K⁺) having three dimensional tunnel structure for the selective removal of strontium and cesium from simulated ground water conditions.⁴⁵⁾ These materials are structural analogs of the mineral pharmacosiderite for titanium and

silicon. Pharmacosiderite is a non-aluminosilicate molecular sieve with the framework composition $[\text{Fe}_4(\text{OH})_4(\text{AsO}_4)_3] \cdot 5\text{H}_2\text{O}$. For the titanosilicate analogs, the framework arrangement of silicate tetrahedral and titanium octahedral create three-dimensional structures with water molecules and charge-neutralizing cations located in the face-centres. Results obtained for distribution coefficient measurements for groundwater simulant for these two titanium silicates are shown in the Table 1 which clearly show that the sodium form has the higher K_d values however, the percentage removal values are almost found to be identical for both these exchangers i.e., ca 97-98%.

Table 1. Sr^{2+} and Cs^+ distribution coefficients (K_d s for the potassium (K26L) and sodium titanosilicates (Na26L) in groundwater simulant⁴⁵⁾

Sample	K_d (ml g^{-1})		pH_F	% Sr^{2+} removed	% Cs^+ removed
	Sr^{2+}	Cs^+			
K26L	7400 ^a	4600	9.7	97.4	95.9
Na26L	10800 ^a	9700	7.2	98.2	98.0

a Below detection limit. K_d values were determined using the lowest standard

In a line the selectivity of Nb-substitute titanosilicate with sitinakite topology was studied for ^{137}Cs and ^{89}Sr ⁵⁷⁾ and was demonstrated that enhanced uptake of ^{137}Cs was observed but lower for ^{89}Sr when it compared with the titanosilicates uptake behavior. Such difference in adsorption behavior was concluded mainly due to the different coordination environment of cesium and strontium in the eight-ring channel that results from various hydration sites in the tunnel. The sorption isotherm and NMR studies reveals that calcium titanate hydrate (CSH) is an effective choice for the cesium uptake.⁵⁸⁾ It was noted, cesium cations remains tightly bound to the surface sites of CSH forming, in dehydrated CSH, inner-sphere complexes, which can not be removed by alcohol washing and the chloride affinity is almost negligible even for high Ca/Si molar ratios, where the surface charge becomes positive. Similarly, semicrystalline potassium titanium silicate (KTS) ($\text{K}_3\text{HTi}_4\text{O}_4(\text{SiO}_4)_3 \cdot 4\text{H}_2\text{O}$) assessed

for the cesium and strontium.⁵⁹⁾ They reported that KTS is a moderately weak cation exchanger, possessing high ion exchange capacity (4-5 meq/g) and a moderate affinity towards heavy alkali and alkaline earth metals. It was reported that it has limited application for removal of radioactive cesium and strontium. Model based approach was presented for the removal of cesium by using crystalline silicotitanate (CST) and was proposed to be effective for the treatment of the Savannah River Site (SRS) wastes as a large amount of ^{137}Cs can be confined within a small volume of saturated CST particles for permanent storage.⁶⁰⁾

Silica gel is quite effective for creating hydraulic barriers to prevent the advective migration of radioactive contaminants in shallow permeable sediments. However, the effectiveness of silica gel grouted barriers in controlling transport of fission product radionuclides through sorption and diffusion was unknown. The work carried out by the Hakem et al.⁶¹⁾ was an attempt to assess the sorption of fission product radionuclides, ^{137}Cs and ^{90}Sr , by Savannah River sediments (South Carolina Site) impregnated with colloidal silica. The static batch type experiments conducted with and without the silica gel and it is to be reported that the retention of these radionuclides can be enhanced remarkably by the application of silica gel and also the data showed that the retention increased with decreasing radioelement concentration and it is fitted well for Freundlich adsorption isotherm.

Since early 1960 the synthesis and applications of various alkali metal titanates were started and at present several such titanates of the type $\text{A}_2\text{Ti}_m\text{O}_{2m+1}$ and $\text{A}_4\text{Ti}_n\text{O}_{2n+2}$ ($m = 1-9$ and $n = 1,3,5,7$ and 9) are known. These materials are found to have wider applications in the removal/speciation of various radiotoxic ions including strontium and cesium.⁶²⁻⁷³⁾ Similarly, the silicotitanates,⁷⁴⁻⁷⁶⁾ titanium antimonates (rutile),⁷⁷⁾ and titanium tungstates⁷⁸⁾ (amorphous) are assessed in a way to find their suitability in the removal behavior at high and medium salt concentration for radio cesium. Interestingly, the twenty

hydrous mixed metal oxides of Sb, Nb, Si, Ti and W with pyrochlore structures were prepared both by precipitation and by hydrothermal treatment (150-190 °C) and are applied for the removal behavior of Cs and Sr ions in the batch type experiments and it was observed that the selectivity and adsorption capacity was remarkably increased by the selective combination.⁷⁹⁾ In a line, recently the polyantimonic acid was precipitated in the pores of Amberlite XAD-7 (PAA-XAD) and studied for the uptake of Sr from the acidic and simulated spent fuel solutions. The batch type and column experiments reveal that the K_d of 6069 mL/g was obtained for this solid at 0.1M HNO₃ solutions.⁸⁰⁾ The same group also assessed the ion exchange and column separation of cesium, strontium and europium from acidic streams by employing the uranium antimonite.⁸¹⁾

Hydrous Ferric Oxide

Hydrous ferric oxide, which is a common iron oxide mineral in aquatic and soil environments shows a good adsorption capacity to adsorb the metal cations.⁸²⁻⁸³⁾ Generally the sorption by the hydroxides took place in two steps: a fast initial uptake and a slow subsequent sorption step that may prolong for days and months. The first step is attributed to fast sorption on the external surfaces of the oxide particles. The second step has been attributed to (i) intraparticle diffusion and sorption onto interior sites, (ii) slow redistribution of surface complexes (e.g., from mono-dentate to bi-dentate),⁸⁴⁻⁸⁵⁾ or (iii) surface precipitation⁸⁴⁾ Hydrous ferric oxide (HFO) as obtained with nano-particle size with diameter between 2 to 5 nm having the extremely large surface area in the order of 600 m² g⁻¹^{82,86-87)} can show the sorption of metal cations controlled by slow diffusion processes into the small pores of HFO aggregates.⁸⁸⁻⁹⁰⁾ The diffusion of strontium in the hydrous ferric oxide aggregates were discussed with the help of Donnan diffusion model.⁹¹⁻⁹²⁾ The basicity and FeO effects was presented on the retention of ¹³⁷Cs and ⁶⁰Co by performing the simulated non-metallic solidified

radioactive wastes (Al₂O₃-CaO-FeO-SiO₂) containing these radionuclides. The retention of ⁶⁰Co was almost independent of chemical composition of slag but the retention of ¹³⁷Cs was decreased with an increase of the basicity of slag, defined as a ratio of CaO to SiO₂ in weight and independent to FeO concentration.⁹³⁾

Clay and Minerals in Radioactive Waste Management Study

Clay and clay based materials are important engineered barriers in many waste disposal strategies designed to minimize contaminant migration from the waste to the surrounding environment. Studies related to the sorption/diffusion of radionuclides through such materials are quite important for designing the waste disposal repositories or to develop the safeguards for any radioactive hazard to the surroundings. Indeed, voluminous work has already been carried out with various kinds of clays and clay-based materials to assess their suitability/applicability for such studies. Naturally occurring swelling clays (e.g., montmorillonite, vermiculite) are regarded as backfill material in the engineered barriers of high-level waste forms⁹⁴⁻⁹⁷⁾ as due to its high cation exchange capacity, low hydraulic permeability, adequate thermal conductivity, etc. which, ensures the long lived radionuclides are to be retained for more than 10³ years, such that the radioactivity of these fission products decayed to its low value. The countries like Sweden, Finland, Switzerland etc. are extensively using the bentonite in the construction of repositories for the disposal of spent nuclear fuel and fission products.⁹⁸⁻¹⁰¹⁾

Cesium and strontium were sorbed on Wyoming bentonite and the data were analyzed with modified two-stage Freundlich adsorption model with primary factors such as temperature, pH value and nuclide concentration which, of course influenced the sorption behavior of bentonite.¹⁰²⁾ Similarly, sorption behavior of bentonite was assessed for ¹³⁷Cs, ⁸⁵Sr and ⁶⁰Co in a batch type experiments and showed that with the increase of pH, uptake retention of

these ions increased remarkably and the desorption studies with 0.01M CaCl₂ and ground water at low-metal loadings on bentonite showed that about 95% of Cs, 85-90% of Sr and 97% of Co were irreversibly sorbed.¹⁰³⁾ The sorption behavior of cesium on natural bentonite in synthetic groundwater (containing Ca, Mg, Na, K and carbonates) conditions.¹⁰⁴⁾ The thermodynamic and kinetic data were obtained for the retention of strontium by natural kaolinite and clinoptilolite minerals and it was reported that the sorption process followed pseudo second order kinetics, with faster sorption on kaolinite compared to clinoptilolite, where the uptake is affected by intra-particle diffusion. Freundlich and Dubinin-Radushkevich isotherm models described the data more adequately than Langmuir model, and clinoptilolite showed a higher strontium sorption than kaolinite.¹⁰⁵⁾ The different deposits of clinoptilolites from Bulgaria and the USSR in the different forms (i.e., natural, sodium, calcium and ammonium) were chosen to study the sorption behavior of these solids for cesium and strontium. Moreover, the mechanical characteristics which is important for the assessment of the technological use of these sorbents in dynamic conditions are determined.¹⁰⁶⁾ Red-clays from Sivrihisar-Eskisehir (Turkey) were used for the sorption studies of cesium and strontium ions using EDXRF spectrometry.¹⁰⁷⁾ Similarly, the kinetics was reported for the adsorption of strontium on natural clays.¹⁰⁸⁾ Carbonate-containing natural mineral tripolite was studied for the recovery of ¹³⁷Cs and ⁹⁰Sr from the model aqueous and real waste water solutions in a wide range of pH and in the presence of isotopic and nonisotopic carriers and organic impurities. They reported the saturation capacities of tripolite with respect to cesium and strontium were 0.3 and 1.0 mg-equiv g⁻¹.¹⁰⁹⁾ Spectral characterization was made by XRPD, SEM/EDS, XPS and DRIFT analysis for the sorption of Sr²⁺ on natural minerals of kaolinite and magnesite. XPS data showed magnesite is more effective for Sr²⁺, EDS mapping indicate that while the sorbed Sr is equally distributed on surface of natural kaolinite, it was associated -

to a larger extent - with the regions richer in Mg in the case of natural magnesite.¹¹⁰⁾

It is noted that while carrying out the transport modeling of the contaminants, more realistic parameters are required as the distribution coefficient K_d (for low concentration) and the maximum adsorption capacity S_T (for high concentrations). Hence, in view of that Gutierrez¹¹¹⁾ obtained the possible data for the competitive sorption of strontium, cesium and cobalt at various pH values and for two background electrolytes (i.e., NaCl and CaCl₂). In a line, granite and diorite, as a host rock in Taiwan area, were investigated for the sorption of various radionuclides viz., Co, Sr, Cs, U, I and Am in the deionized and seawater conditions by performing the batch tests and polar-microscopy/autoradiography. It was reported that biotite in the rock sample is the principal mineral component which is responsible for the sorption of ¹³⁷Cs and ⁹⁰Sr onto granite and diorite.¹¹²⁾

In an attempt to increase the suitability and efficiency of a particular clay or mineral for these radionuclides as well for anionic type of radionuclides like long lived ¹²⁹I and ⁹⁹Tc, these clays were modified by replacing natural inorganic interlayer cations with quaternary alkylammonium ions of the form [RN(CH₃)₃]⁺, where R is an alkyl or aromatic hydrocarbon. The resulting organo-clays are capable of sorbing non-ionic organic compounds as well as iodide. In this regard the hexadecylpyridium (HDPy⁺)-Vermiculite system, makes it efficient to sorb comparable amounts of anions (I⁻) and cations (Cs⁺, Ca²⁺) simultaneously.¹¹³⁻¹¹⁴⁾ Similarly, the MX-80 Wyoming-bentonite treated with different amounts of hexadecylpyridinium (HDPy⁺) were used for the uptake of I⁻, TcO₄⁻, Cs⁺ and Sr²⁺. Using bidistilled water, synthetic ground water (SGW) and sea water with half of the ionic strength (Sea/2) as equilibrium solutions. It was reported that the increasing alkyl-ammonium ion (HDPy⁺) incorporation in the clay suppress the sorption of Cs⁺ and Sr²⁺.¹¹⁵⁾ Sivaiah et al.¹¹⁶⁾ modified the vermiculite with zirconia for strontium ion adsorption. They attempted actually to place pillars in the

interlayer of vermiculite by classical method and partially they were able to swell or prop apart the lamella of vermiculite to some extent by the tetrameric $[Zr_4(OH)_8(H_2O)_{16}]^{8+}$ ion that results in a zirconia modified product, ZrMV, which seems to be adequate for the rapid sorption of strontium than the parent clay. The comparison of results obtained with vermiculite and zirconia modified vermiculite are presented as a function of pH and in the presence of Na^+ and Ca^{2+} ions concentrations (*cf.* Tables 2 and 3) which clearly shows that the modified vermiculite with zirconia has remarkable enhanced suitability and are to be a superior candidate over vermiculite as a back fill material, and it can ensure the retention of radioactive strontium if leached out from the waste forms.

Table 2. Variation of percentage sorption of strontium tracer on vermiculite and ZrMV as a function of pH¹¹⁶⁾

pH _{eq}	Percentage Sorption (%)	
	Vermiculite	ZrMV
1.00	0.00	3.4
2.00	20.2	37.3
3.00	70.6	99.2
4.00	87.5	99.3
4.95	87.5	99.7
5.86	88	99.8
6.67	89.1	99.9

V/m = 200 mL/g, T = 301 K, equilibrium time = 6h, $[Sr^{2+}]$ = trace

Table 3. Variation of distribution Coefficient (Kd) of strontium in the presence of sodium and calcium ion¹¹⁶⁾

$[Mn^+]$	Kd of strontium (mL/g) in the presence of			
	Na^+		Ca^{2+}	
	Vermiculite	ZrMV	Vermiculite	ZrMV
0.1	42	55	2	8
0.05	120	225	51	64
0.01	411	2896	114	220
0.001	566	22178	301	1242

$[Sr^{2+}] = 1.14 \times 10^{-4}$ M, V/m = 200 mL/g, $[HNO_3] = 0.001$ M, equilibrium time = 6h, T = 301 K.

Compacted clay based materials are important engineered barriers in many waste disposal strategies designed to minimize contaminant migration from the waste to the surrounding

environment. Moreover, the disposal concept being developed in Canada, for example, both compacted 1:1 bentonite/sand (buffer material) and 1:3 clay/crushed granite (backfill material) mixtures are being considered as barriers and seals in a vault for disposal of nuclear fuel wastes. These clay-based barriers have low permeability, and hence, diffusion is the principal mechanism of transport of most contaminants through these materials.¹¹⁷⁻¹¹⁸⁾ Hence, the data related to diffusion of these radionuclides through the porous media are found to be more realistic in the radioactive waste management studies. The diffusion of $^{22}Na^+$, $^{85}Sr^{2+}$, $^{134}Cs^+$ and $^{57}Co^{2+}$ in bentonite clay compacted densities were performed experimentally along with modeled with the computer code ANADIFF.¹¹⁹⁾ It was reported that the diffusivities of Cs^+ and Sr^{2+} decreased as the density of bentonite increased. The diffusivity of Cs^+ decreased from 1.5×10^{-7} to 1.1×10^{-8} cm²/s as the clay dry bulk density increased from 0.4 to 1.8 g/cm³ and the diffusivity for Sr^{2+} varied from 4.3×10^{-7} to 9.0×10^{-8} cm²/s. Sorption and diffusion behavior of Cs and Sr was carried out using the Jih-Hsing (China) bentonite. They applied the compacted bentonite samples with the densities of 1.8, 2.0 and 2.2 g/cm³ and with these density samples the observed apparent diffusion coefficients were found to be respectively $(2.83 \pm 0.75) \times 10^{-13}$, $(1.97 \pm 0.02) \times 10^{-13}$ and $(1.91 \pm 0.12) \times 10^{-13}$ m²/s for cesium and $(1.33 \pm 0.13) \times 10^{-13}$, $(1.51 \pm 0.15) \times 10^{-13}$ and $(1.34 \pm 0.10) \times 10^{-13}$ m²/s for strontium.¹²⁰⁾

Diffusion Through Rock & Rock Materials

The ultimate disposal of nuclear wastes in crystalline rock has been proposed in many countries and interest in the ability of the geosphere to retard radionuclides released from such repositories has increased.¹²¹⁻¹²²⁾ The retardation processes in the geosphere are believed to be sorption onto the fractured rock and diffusion into micropores in the rock matrix, so called matrix diffusion.¹²³⁾ Earlier, the intra-particle diffusion of cesium and strontium cations into rock materials were carried out by using the

crushed rock samples in a batch adsorption technique.¹²⁴⁾ The diffusion coefficient values for these two cations ranges from 10^{-8} - 10^{-7} cm²/s and the diffusion proceeded into two competitive processes i.e., surface and pore diffusion. Charnockite rock, intact and fractured rock samples were studied for the diffusion behavior of iodide, cesium and strontium. It was seen that despite the sorbing nature of cesium and strontium, they diffuse comparatively faster than iodide (non-sorbing ion) through the intact and fractured rock samples.¹²⁵⁾ In a line several work reported on the natural and synthetic zeolites in their retention property for cesium and strontium.¹²⁶⁻¹³¹⁾

Interesting data were obtained by carrying out *in-situ* migration experiments at Aspo Hard Rock Laboratory, Sweden for the radioactive tracer migration through the fractures over a distance of 5 m and at a depth of 400m at the laboratory site.¹³²⁾ The experiment lasted for nearly 1.5 years, with single experimental time upto 10,000 hrs. The relative retardation sequence obtained in the field experiment was Na<Ca \approx Sr<K<Ba \approx Rb<Co \approx Cs, which was the same as the relative sequence of the sorption coefficients obtained in the laboratory experiments using crushed rock material. Silica sand-filled Lysimeter was used to note down the upward migration of ¹³⁴Cs, ¹³⁷Cs and ⁹⁰Sr at the Field Lysimeter Investigations: Low Level Waste Data Base Development Experimental Site at Oak Ridge National Laboratory, USA. They have found Cs and Sr migrated upward in the evapotranspiration stream of the root and as the sectioning of the sand core collected from directly above the buried waste form it was discovered that a fine root from an unidentified plant was present throughout all but the upper few centimeters of the core.¹³³⁾ Hence, they suggested further study must be undertaken to confirm this phenomenon i.e., the upward migration of radionuclides as observed here could result in direct exposures and offsite release from underground storage facilities.

The low level waste disposal facility at the Hanford site, Washington was chosen for the simulated batch and column experiments as the

borehole sediments were collected from the Hanford site (200 East Area) and it was reported that among the studied radionuclides (⁹⁹Tc, ¹²⁹I, ⁷⁹Se and ⁹⁰Sr), ⁹⁰Sr showed highest sorption affinity following with ⁷⁹Se and ¹²⁹I and ⁹⁹Tc showed the least affinity for the sediments. The data are useful for more defensibility to the past performance assessment predictions.¹³⁴⁾ Similar results were also shown earlier as radiostrontium and radiocesium strongly sorbed by the Hanford soil.¹³⁵⁾

Role of Soils as Natural Barrier

The role of soils in radioactive waste management studies is found to be important, as it is one of the natural barriers of the migration of radionuclides nearby the disposal sites. Therefore, it is worth considering the data and scientific inputs related to the sorption/desorption behavior of soil samples for various radionuclides. The migration of radionuclides through soils mainly depends on groundwater movement and interactions with soil component.¹³⁶⁻¹⁴⁰⁾ Ion exchange and sorption kinetics of cesium and strontium in soil samples (Jishin soil and Lanyui Soil of Taiwan) were studied *via* batch method. Ion exchange is mainly responsible for the sorption of the Cs and Sr on both the soils and the monovalent ions are exchanged by Cs and similarly the divalent ions are exchanged by Sr in addition the film diffusion is reported to be the rate-limiting step.¹⁴¹⁾

The sorption/desorption behavior of sandy soils for strontium was investigated with reference to the minerals of the soil components with 0.1 M LiCl solution and revealed that the additivity rule can not directly be applied to predict the sorption behavior of strontium on this sandy soil but the distribution ratio (Kd_s) of strontium for sandy soils is found to be expressed by the following equation:

$$Kd_s = \alpha \sum_i Kd'_i \cdot p_i$$

where Kd'_i and p_i are the distribution ratio for a reference mineral of the i -th soil component and the content of the i -th soil component, re-

spectively, and α is the ratio of a measured cation exchange capacity (CEC) of a sandy soil to that estimated on the basis of CECs of the reference minerals. The equation indicates that the CECs are important for predicting the sorption characteristics of strontium on sandy soils.¹⁴²⁾ Competitive adsorption of cesium, cobalt and strontium were performed with soil samples from Hudspeth County, Texas and the laboratory measurements revealed that the competition coefficients vary from 0.15 to 0.20 for Co in the presence of Sr and 1.0 to 1.3 in the presence of Cs; 0.3 to 0.6 for Cs in the presence of Sr and 0.4 to 0.8 in the presence of Co; 3.0 to 6.3 for Sr in the presence of Cs, and 4.5 in the presence of Co. These values are indicative of the heterogeneous interactions between ions and adsorption sites and also to be concluded that Co and Cs do not compete for adsorption sites as much as Co does with Sr, or Cs with Sr.¹⁴³⁾ Similarly, Yasuda et al.¹⁴⁴⁾ tried 36 agricultural soils (paddy and upland) collected in Japan and measured the K_{ds} for five radionuclides (⁵⁴Mn, ⁶⁰Co, ⁶⁵Zn, ⁸⁵Sr, ¹³⁷Cs) using the batch type experiments. Strontium and Cesium behavior on soil from the Radioactive Waste Management Complex (RWMC) of the Idaho National Engineering and Environmental Laboratory (INEEL), located on the Snake River Plain of Southern Idaho, USA, was investigated using sequential aqueous extractions and batch sorption methods. Strontium sorption was reported to be reversible, while Cs was not, except at very low concentrations. Slight increase can be obtained for both the ions as experiments were repeated on these soils that was treated to remove carbonates.¹⁴⁵⁾

Bioaccumulation and Role of Biosorbents in the Radioactive Waste Management

Biosorbents and dead biomasses showed an impetus role in removal/speciation of heavy metal toxic ions from aqueous solutions as due to their extremely good adsorption capacity and also showed comparatively good selectivity for these heavy metal toxic ions. With this it is interesting to assess various such biosorbents/dead biomasses

in the radioactive waste management studies and also their removal/speciation behavior towards the several radionuclides from the aqueous solutions. The suitability/selectivity of these biosorbents depends largely upon their stability towards radiation and temperature. Very scanty of the work has been reported in this regard. Mishra and Tiwari¹⁴⁶⁾ studied various dead biomasses particularly the agricultural by product rice hulls and bark samples of *Mangifera indica* and *Azadirachta indica* trees for their uptake behavior towards the Sr(85+89) from aqueous solutions. The batch type experiments show that the increase in sorptive concentration, temperature and pH favors the uptake of Sr(II) ions. Further, these solids are also being assessed for their radiation stability by irradiating it towards a 300 mCi Ra-Be neutron source with the moderate neutron dose i.e., 3.85×10^6 n/cm²/sec associated with nominal γ -dose of about 1.72 Gy/h and it was found that these solids are stable towards the employed radiation doses at least for the removal of Sr(II). The group also tried to find out the suitability of these dead biomasses for their selectivity towards the ¹³⁴Cs and was reported that relatively less uptake of this ion from aqueous solutions showed good radiation stability towards the above mentioned radiation doses.¹⁴⁷⁻¹⁴⁸⁾ Earlier, the detection and isolation of several radionuclides viz., Co-60, Sr-89, Ru-106, I-131, Cs-134 and Ce-144 from radionuclides-accumulating bacteria by autoradiography was discussed.¹⁴⁹⁾ The feasibility of cesium removal was assessed by using various derived biosorbents originally obtained from marine algae. They modified the biosorbents to ferrocyanide type1(FAS1) and type2 (FAS2) and these modified biosorbents are more efficient than the original one as they can remove almost all cesium from aqueous solutions (370 mg/L cesium solution) just within 30 minutes of contact and the uptake process well suited at a wide pH range (1 to 10) and unaffected with the sodium and/or potassium ion concentrations 0.5 and 1 mM.¹⁵⁰⁾ ²⁴¹Am was removed from the aqueous solutions using the *Candida sp* biomass.¹⁵¹⁾

The results showed that the adsorption of ^{241}Am onto *Candida sp.* was efficient. ^{241}Am could be removed by *Candida sp.* of 0.82 g/L (dry weight) from ^{241}Am solutions of 5.6-11 MBq/L (44.3-877.2 $\mu\text{g/L}$) (C_0), with maximum adsorption rate (R) of 98 and maximum adsorption capacities (W) of 63.5 MBq/g biomass (dry weight) (501.8 $\mu\text{g/g}$). Retention of strontium, cesium, lead and uranium by bacterial iron oxides from a subterranean environment was reported by Ferris et al.¹⁵²⁾ They collected bacteriogenic Fe oxides (BIOS) and groundwater samples at 195 m underground at the Strassa Mine in central Sweden having the prominent bacteria *Gallionella ferruginea* and *Leptothrix sp.* in the BIOS samples, and the retention of the Sr, Cs, Pb and U in filtered groundwater was ranging from 0.002 to 1.8 μM . Solid phase concentrations of these ions, in the BIOS spanned the 0.04-2.23 m mol/kg range. Moss (*Funaria hygrometrica*) a phyto-sorbent was used for the removal of ^{137}Cs and ^{90}Sr in a batch laboratory experiments. It was reported that very rapid uptake of these radionuclides further increased with increasing the solution pH and also with pre NaOH treated moss. They proposed that if the large quantities of this moss could be grown under artificial conditions, phyto-sorption using moss might emerge as a viable technology for the routine cleanup of LLW (low level waste) solutions. The recovery of carrier free $^{137}\text{Cs}/^{90}\text{Sr}$ also appears to be possible.¹⁵³⁾ Although the application of bio-sorbents/dead biomasses in the removal/speciation of these two toxic ions are found to be very effective and can play an alternative role in the removal/speciation of these toxic ions however, very scanty of the work has already been done hence, more studies particularly the stability of these bio-materials are to be assessed as exposed with higher radiation doses in addition to higher temperatures in order to find their suitability in the real radioactive waste system.

Mostly the work related to biosorption is focused on the active uptake of these radionuclides if they occurred in the environment either by

nuclear accident or by weapon tests. Uptake of these radionuclides through the plants are reported to be through three main pathways: root uptake, direct deposition onto the foliar system and resuspension of contaminated soil particles.⁴¹⁾ Several studies revealed that the uptake of the radionuclides by the plants and it has been discussed in terms of the various factors viz., the rain effect, plant species and their growth parameter, fallout rate, chemical form of deposited radionuclide, environmental conditions, etc.^{32,41,154-161)} As such studies are beyond the scope of this review, hence not much attention has been paid in this direction.

CONCLUSIONS

Radiocesium and radiostrontium are two very important fission products and are present in significant amount in the radioactive wastes. Relatively long half-lives and very high radiotoxicity of these radionuclides poses high radioactive hazard towards the human being as if it enters into the biosphere even at a trace level concentration. Hence, the removal/isolation of these radionuclides from the wastes is one of the necessary steps prior to the ultimate disposal/storage. Moreover, the concept of radioactive wastes has significantly been changed recently as these waste products are sometimes useful products because of their potential applications in the waste treatment plants, food preservation, medicines etc. hence are required to be isolated from the radioactive wastes. Hence, either due to radioactive hazard or due to its usefulness it has to be removed/isolated from the radioactive wastes before the final disposal. The role of various inorganic materials including the clay minerals, soil samples for their interaction and sorption behavior are important part of the studies. These solids showed extremely good selectivity, suitability and also the high radiation and temperature stability finds them useful materials for handling of these radionuclides. On the other hand, the role of biosorbents in such studies is very scarce and much more is required

for their future applications in the radioactive waste management studies.

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