

Application of Modified Clay for Sorption of Radionuclide Elements

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Abstract

The clay materials are distinguished on the basis of their different crystal structures. Due to their specific structure, they are applied in the nuclear industry for the removal of radionuclides from nuclear liquid waste as ion exchange material. This mini review presents some of the recent advances of various kinds of clay materials in this aspect. These inorganic materials continue to gain attention due to their chemical and radiation stability and effectiveness over wide range of conditions.

Keywords: Clay material; Radionuclide; Sorption; Ion exchange

Abbreviations

EXAFS: Extended X-ray Absorption Fine Structure; Kd: Distribution Coefficient; BCF: Boda (siltstone-) Claystone Formation; REE: Rare Earth Elements; ICP: Inductively Coupled Plasma; MS: Mass Spectrometry; SEM: Scanning Electron Microscope; TEM: Transmission Electron Microscope; XRD: X-ray Diffraction; ClayFF: Clay Force Field; SPC: Simple Point Charge

Introduction

The generated nuclear waste in the nuclear industry, including plutonium production, radionuclide removal campaign and relevant scientific research activities is stored for further treatment. Radioactive nuclides in the sludge fractions are the primary water soluble species for the disposition for the reduction of the radioactivity. Ion exchange has been a leading candidate for this separation.

Clay is a fine-grained natural rock or soil material that combines one or more clay minerals with traces of metal oxides and organic matter. Modified clay materials, produced through various synthesis art, can function as ion-exchange materials. Clay materials function as the natural barrier for the geological disposition for the decommissioned nuclear facilities. Therefore, it is necessary to discuss the behaviors of radionuclide metal ions on the clay materials or the modified clay materials.

This mini review article summarizes finding from the synthesis and performance of clay material or modified clay ion exchangers reported during the recent years.

Bentonite

Bentonite, an adsorbent aluminum phyllosilicate, is generally impure clay consisting mostly of montmorillonite. The interaction of layered silicates, such as bentonite, with polymers often results in two particular modifications that should be considered as character of modified bentonite materials. The first is the modification of the silicate layers through ion-exchange reaction, and the second is the ability of silicate layer to disperse as single layers.

Seliman AF collects Bentonite clay samples as a large stone from Egyptian mining at kilometer-95 of Cairo-Alexandria desert road. The adsorption of four radionuclides [1], Cs-134, Sr-90, Ba-133 and Eu-152 onto the collected Egyptian bentonite and its modified Na⁺ form is investigated by the application of batch equilibrium techniques. At lower concentration, the value of distribution coefficient (Kd) follows the order of Eu-152>Sr-90>Cs-134>Ba-133 for the Bentonite and Na-Bentonite. The effect of pH for these four ions is studied in the range of 2-12.

Boda (siltstone-) claystone formation (BCF)

Mell P studies the sorption of Co-60, Sr-85, Cs-137 and I -125 on samples of Boda (siltstone-) claystone formation (BCF) in the Hungary. BCF is the candidate media for the final disposal of high level radioactive waste in Hungary [2]. The main mineral components of BCF are: quartz (detrital component, 2–33%), albite (mostly authigenic origin, 10–60%), illite-muscovite (commonly predominant clay mineral, 0–25%), carbonates (calcite and dolomite) and hematite (common rock-forming mineral, 5–10%). The order of sorption of isotopes is Co-60>Cs-137>Sr-85>I-125.

Halloysite

Halloysite has a 1:1 layer structure with a layer of H₂O molecules in between when fully hydrated. Such 1:1 layer structure consists of a unit of one octahedral and one tetrahedral sheet. Th De Putter studies halloysite materials [3] for the immobilization of U, Th, Pb and other rare earth elements (REE) in the karst sedimentary filling during a Miocene weathering event. Special attention is paid to the trace element immobilization/trapping process. Analytical methods such as emission ICP, ICP-MS, XRD, SEM, TEM and other microprobes suggest that both the sandy sediments and the carbonate wall itself acted as geochemical barrier for the immobilization of these mentioned trace elements. It shows that the mentioned radionuclide (U, Th, REE, Pb) migration paths are often limited under such condition, which could prohibit the migration of radionuclides for the low level radioactive wastes.

Pyrophyllite

The simple structure of pyrophyllite is with 2:1 layer structure consisting of two tetrahedral sheets, with one bound to each side of an octahedral sheet. Lei studies the mechanism of the adsorption of aqueous uranyl species UO_2^{2+} to the basal surfaces of clay material Pyrophyllite by the molecular dynamics simulation. Uranyl ion could form various complexes with carbonates [4], such as $(\text{UO}_2(\text{H}_2\text{O})_5)^{2+}$, $(\text{UO}_2(\text{H}_2\text{O})_3(\text{CO}_3))$.

$(\text{UO}_2(\text{H}_2\text{O})_2(\text{CO}_3)_2)^{2-}$ and $(\text{UO}_2(\text{CO}_3)_3)^{4-}$. The simulations show that at aqueous clay interfaces, both uranyl species and surface type control the adsorption pattern. Sorption behaviors derived from this study could supply new insights into understanding the migration and enrichment of uranium and other nuclides. The noncarbonato and monocarbonato uranyl species can form outer-sphere complexes on siloxane surfaces through electrostatic interaction, but the dicarbonato and tricarbonato uranyl complexes rarely adsorb on the siloxane surfaces. The ClayFF force field is used to simulate Pyrophyllite [5], while SPC water model is for water molecules [6,7].

Mica

The Mica material has the 2:1 layer structure with Al^{3+} occupied one quarter of the tetrahedral sites. A Cota studies influence of temperature and time on the Eu^{3+} reaction with synthetic Na-Mica-n ($n=2$ and 4). The swelling high-charge micas [8] could be highly valuable for the decontamination of harmful cations such as Eu^{3+} . The ability of these micas to immobilize Eu^{3+} under subcritical conditions has been probed by adsorption technology. The adsorption process happens in both non-specific sites through the cation exchange mechanism and specific sites through chemical reaction or surface defects adsorption. Due to this point, the Mica material could also be applied to adsorb the radio cesium to reduce the influence from the transfer of radio cesium from soil to paddy rice around agricultural field of the Fukushima area since the accident of Daiichi Nuclear Power Plant [9].

Vermiculite

Vermiculite is with the similar 2:1 layer structure mentioned above. I Plecas studies the sorption character of Mn-54 and Sr-89 onto the cement modified Vermiculite materials. These radionuclides could escape from the waste matrix through diffusion migration in water-saturated conditions, although they are adsorbed onto the cement modified [10] Vermiculite. The leaching resistance is determined for the test of mechanically, physically and chemically stability of the sorption characteristics onto modified Vermiculite.

Chlorite

Like Vermiculite, Chlorite mineral has a 2:1 layer structure with an excess of negative charge which is balanced by a positively charged interlayer hydroxide sheet. A J Fuller studies the Sr sorption to Chlorite under hyper alkaline condition by Extended X-ray Absorption Fine Structure (EXAFS) [11]. The effect of changing pH and ionic strength

on the sorption of Sr to Chlorite soil materials are investigated by EXAFS techniques. It confirms that at highly alkaline pH (>12.5) Sr forms inner-sphere complexes on the surfaces of Chlorite. This specific adsorption of Sr explains the phenomenon of sorption on the mineral under high ionic strength conditions withstanding the existence of Na.

Conclusion

Significant research efforts have been undertaken in the recent years for the application of raw or modified various clay materials for the uptake of radionuclides, from the experimental synthesis art to the simulation work. Clay material family is a huge group of various minerals which serves for the immobilization of radionuclides in the environmental application or the nuclear industry field. This mini review only summarizes part of the minerals which belong to the clay family. Their applications in other fields like biomedicine or material science are out of the discussion of the mini review, although they have found their own application in this mentioned area. Perhaps during the next decade one will even further find the application of these clay materials.

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