studies on the application of certan ion ex changers in nuclear fuel cycle

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This work had been done in an attempt to prepare inorganic ion exchangers that can be of help in the nuclear waste proplem. Also knowing information helping to understanding and improving the ion exchange process. This goal was achieved through the work done in the Chemistry Department, Faculty of Science, Benha, Zagazig University with the corporation of the atomic energy authority, at Inshas. This work was the preparation of the SnSb ion exchanger and used to exchange the different cations under investigation. The pillared layered materials showing a promissing results in the ion exchanger field, from the economy and the selectivity point of view. This work was studied in the Laboratory of prof. A. Clearfield, Chemistry Department, Texas A&MUniversity.The first part is the introduction which includes some aspects of the origin of the nuclear waste problem. According to the advantages of the inorganic ion exchangers over the organic resins, a literature survey was carried out to cover, almost, the last three decades related to the proposedsubject.Part I, contains the experimental and results and discussion for the SnSb materials. The experimental, includes the chemicals used and the method utilized for the preparation of the radioactive nuclides used in this study. The equipement was employed to collect the results, and the methods of the preparation was also given.The results and discussion, consists of four main parts:Characterization of SnSb was prepared using the XRD, DTA, TGA and IR spectrophotometer. The results showed that the SnSb has a poor crystallinity with thermal stability up to 600°C and the water content wasfound to be 13.7%.The ion exchange properties were carried out using three different cations, cobalt(II), uranyl(IV), and europium(III), representing the main different catagories of the nuclear waste solution. In this respect, the effect of the hydrogen ion concentration was studied. It was found that, for cobalt there is a steady decrease in the distribution coefficient with the increase of metal ion concentration. from the plot of log Kd vs the hydrogen ion concentration both, uranyl ion and europiun ion was found to follow the ideal ion exchange reaction mechaniem.Kinetics of exchange was studied as a function of particle radius and solution concentration. The Di values calculated for the nucleides under the investigation indicated that the Di value for cobalt(II), increases with increasing particle diameter up to 8.5X10-10cm2 s-1 at 0.45 mm particle diameter. Uranyl ion showed an appreciable increase of (Di) at SnSb sample heated at 400°C giving 31.20X10-9 cm2 s-1. This was referred to the increase in the SnSb crystallinity with the heating temperature. However, euorpium showed a slight increase in the (Di) values with increasing particle diameter. Comparing the obtained results with those given in the literature it was concluded that the interdiffusion coefficient strongly depends on the ion under the exchange process.The energy of activation was calculated as well as the entropy ofthe exchange. Cobalt(II) ion gave 5.74 kJ/mol, uranyl give 35.9 kJ/molandd euorpium give 15.95 kJ/mol.The saturation capacity was calculated from the Langmuir adsorption isotherm. The results indicates that the saturation capacity of uranyl ion is 0.5 meq/g, cobalt is 0.48 meq/g and of euorpium is 0.78 meq/g. In terms of the saturation capacities SnSb it seems to be a better sorbant for euorpium than the other cations used in this work.Part II, is concerning the pillared clay material. As mentioned in part I, the experimental included the chemicals used and the equipment as well as the different techniques employed. In details the preparations of both keggin ion and pillared clay was given. Kinetic measurements and the stability of the prepared pillared clay was given.Results and discussion, consists of the characterization of the keggin ion used for the intercalation process and the utilised NMR and XRD techniques. The results showed that the Al-13 keggin ion was as expected showing a 63 ppm NMR chemical shift and give a typical XRDpattern for Al-13 in the sulfate form.The surface area was measured with N2 at 77K showing the increase of surface area that was noticed for all pillared samples such as an increase of the surface area up to 400 m2/g of a sample pillared at room temperature and calcined at 400°C for 3h. The pillaring process was carried out at three different temperatures room temperature, 60°C, and 80°C. Considiring the -interlayer spacing (d-spacing) the pillared clay pillared at 60°C give the maximum d spacing up to 18.22 A for thesample heated to 500°C for 3h.The TGA carried out was given. The results indicated that thepillared clay was thermally stable up to 600°C. On heating up to 700°C the sample collapsed with loss of the micropore volume. The chemical analysis using microprob analysis was carried out. The chemical composition was calculated to be:[A11304(OH)27.11(420)8.8910.10[A11.61Mg0.389]Si4010(OH)2._The acid base stability was carried out using 0.01-0.1 M HCl or NaOH. The results indicated that the removal of aluminum from the pillared clay increased with the increases of the acid or base concentration. However the effect of NaOH on dissolution of Aluminum from the pillars shows a decrease with increasing heat treatment of the pillared material in the order 200>300>500°C.finally, the pH titration curve for Na+ and Cs+ was carried out on the pillared clay that had been heated to 500°C prior to exchange. The cation exchange capacity was 0.63 and 1.77 meq/g at pH 11.2 and 11.56 for Na+ and Cs+, respectively. At lower pH increasing amounts of aluminum were solubilized accompanied by a sharp DROP in ion uptake. In acid solution, chloride ion uptake increase started at pH=4.9 and reached a maximum of 1.55 and 1.54 meq/g for sodium and cesium ion, respectively, at pH=2.3. At a higher level of acidity increasing amounts of aluminum were solubilized accompanied bydecreased chloride ion uptake.