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INVESTIGATION THE INTERACTION OF CESIUM IONS WITH DIFFERENT BARRIER MATERIALS

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Abstract: As a result of nuclear weapons testing, nuclear accidents and leaks from high level waste storage sites significant quantities of radionuclides have been released to the environment. Therefore, presence of radionuclides and toxic metals in wastes are a major environmental concern. Considering that cesium ($t_{1/2}$: 30.1 y) radionuclide has long half-life, radioactive wastes containing cesium are considered to be dangerous pollutants and their migration by groundwater is strongly affected by their adsorption on the geologic materials. Sorption processes of radionuclides help to understand prediction of their mobility. We investigated sorption of cesium in batch experiments for different barrier materials to examine concentration effect under ambient experimental conditions. The adsorption data was subjected to four different adsorption isotherms (Freundlich, Dubinin–Radushkevich, Langmuir and Temkin). Hence, various adsorption constants were obtained from the slope and intercept of isotherm graphs. The main energy of adsorption was also calculated from the adsorption isotherms.

Keywords: Nuclear waste, Barrier material, Sorption, Interaction, Isotherms, Cesium

1. Introduction

Since the chemical behaviour of radioactive isotopes is almost the same as that of their natural stable counterparts, non-radioactive stable isotopes may be used to explain the interaction of some important barrier materials in nuclear waste management with radioactive waste and to examine their structural behaviour against such wastes. In this way, the effects of interaction between temperature and time-dependent nuclear waste and barrier materials can be examined (Putman, 1956).

Cesium radionuclide in radioactive waste is considered to be dangerous pollutants and their migration by groundwater is strongly affected by their adsorption on the geologic materials of nuclear waste deposition areas. Considering that cesium ($t_{1/2}$:30.1 y) radionuclide has long half-life, presence of this radionuclide and toxic metals in wastes is a major environmental concern. Sorption processes of radionuclides help to understand prediction of their mobility (Ararem et al., 2013; Chen et al., 2016).

This work aims to investigate the ability of man-made barrier material to predict Cs mobility in aqueous solutions by testing the isotherm models, thermodynamic parameters and the diffusion parameters.

2. Materials and methods

Amino pyridine sulfonamide resin was used as a barrier material for simulated nuclear waste deposition area. Pure cesium nitrate (Merck) was used for adsorption experiments. Solutions of cesium were prepared by dissolving known quantities of cesium nitrate in distilled water.

For pH adjustment of cesium solutions, the buffers of pH 4 and 8 were used for the calibration of pH meter. Solutions of HNO_3 and Na_2CO_3–$NaHCO_3$ were added for pH adjustment.

0.1 g barrier material in a polyethylene tube was interacted with 25 mL of non-radioactive tracer solution of known concentration in a flask at ambient temperature in a thermostated shaker water bath for a known period of time. After equilibration, solid and solution phases were separated by filtration using Whatman No. 40 filter paper.

The cesium concentrations in solution were determined by ICP-OES (Perkin Elmer). The results are the average of at least duplicate independent measurements. Adsorption efficiencies (%) were calculated using mass balance equation.

3. Results

According to the results, the adsorption of monovalent ions in the barrier material showed a low concentration and the adsorption efficiency remained constant with the increase in concentration. It has been observed that the adsorption capacity of the barrier material against monovalent ions is low. Adsorption capacity for monovalent cesium ions was found as 0.12 g.

Sorption isotherms provide sufficient physicochemical information about mechanism properties and tendency of the adsorbent for target species by analysis of the experimental equilibrium data between the ion and solid surface. Adsorption equilibrium data were adapted to different adsorption isotherm models such as Langmuir, Freundlich, Dubinin-Radushkevich (D-R) and Temkin. The correlation coefficients (R^2) indicate the suitability of the adsorption data to the isotherms. According to the results, Dubinin-Radushkevich (D-R) (R^2=0.99) and Temkin (R^2=0.90) isotherms were found to be in good agreement for monovalent ions.

Thermodynamic considerations of an adsorption process are necessary to conclude whether the process is spontaneous or not. Gibb's free energy change, $\Delta G°$, is the fundamental criterion of spontaneity. Thermodynamic parameters such as enthalpy change ($\Delta H°$), entropy change ($\Delta S°$) and free energy change ($\Delta G°$) were estimated using the related equations. The positive value of enthalpy for cesium ions indicates that the interaction is an endothermic reaction which consumes energy.

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ИССЛЕДОВАНИЕ ВЗАИМОДЕЙСТВИЯ ИОНОВ ЦЕЗИЯ С РАЗЛИЧНЫМИ БАРЬЕРНЫМИ МАТЕРИАЛАМИ

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Резюме: В результате испытаний ядерного оружия, ядерных аварий и утечек из хранилищ высокоактивных отходов значительные количества радионуклидов попали в окружающую среду. Поэтому наличие радионуклидов и токсичных металлов в отходах является серьезной экологической проблемой. Учитывая, что радионуклид цезия ($t_{1/2}$: 30.1 y) имеет длительный период полураспада, радиоактивные отходы, содержащие цезий, считаются опасными загрязнителями, и их миграция подземными водами сильно зависит от их адсорбции на геологических материалах. Сорбционные процессы радионуклидов помогают понять прогноз их подвижности. Мы исследовали сорбцию цезия в периодических экспериментах для различных барьерных материалов, чтобы изучить влияние концентрации в условиях окружающей среды. Данные по адсорбции подвергались четырем различным изотермам адсорбции (Фрейндлих, Дубинин – Радушкевич, Ленгмюр и Темкин). Следовательно, различные константы адсорбции были получены из наклона и пересечения изотермических графиков. Основную энергию адсорбции также рассчитывали по изотермам адсорбции.

Ключевые слова: ядерные отходы, барьерный материал, сорбция, взаимодействие, изотермы, цезий.

SEZİUM İONLARININ MÜXTƏLİF BARİYER MATERİALLARI İLƏ QARŞILIQLI ƏLAQƏSİNİN TƏDQİQİ

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Xülasə: Nüvə silahlarının sınaqdan keçirilməsi, nüvə qəzaları və yüksək səviyyəli tullantı saxlama yerlərindən sızmalar nəticəsində ətraf mühitə ciddi miqdarda radionuklidlər yayılır. Buna görə də, tullantılarda radionuklidlərin və zəhərli metalların olması ətraf mühit üçün böyük bir təhlükədir. Sezyum ($t_{1/2}$: 30.1 y) radionuklidinin uzun ömürlü olmasını nəzərə alsaq, sezyum ehtiva edən radioaktiv tullantılar təhlükəli çirkləndiricilər hesab edilir və onların yeraltı sular vasitəsilə hərəkəti geoloji materiallar üzərində adsorbsiyasından ciddi şəkildə təsirlənir. Biz ətraf mühitin eksperimental şəraitində konsentrasiyanın təsirini araşdırmaq məqsədi ilə bir çox təcrübələrdə müxtəlif maneə materialları üçün sezyumun sorbsiyasını tədqiq etdik. Adsorbsiya məlumatları dörd fərqli adsorbsiya izoterminə (Freundlich, Dubinin-Radushkevich, Langmuir və Temkin) məruz qalmışdır. Buna görə, yamacdan və izoterm qraflarının kəsilməsindən müxtəlif adsorbsiya sabitləri əldə edilmişdir. Adsorbsiyanın əsas enerjisi də adsorbsiya izotermlərindən hesablanmışdır.

Açar sözlər: nüvə tullantıları, bariyer materialları, sorbsiya, qarşılıqlı əlaqə, isotermlər, sezyum