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Article

Hybrid composite matrices to prevent radionuclides from spreading into the environment

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Abstract. This paper presents a discussion of the effective creation of hybrid composite materials (HCM) with significant sorption abilities for radionuclides in liquid radioactive waste. These hybrid composite materials are made up of two sorbents, which are derived from an organic source and a natural mineral containing intercalated ferrocyanide complexes of transition metal ions. A substantial degree of radionuclide uptake is noted during the initial phase of sorption, which is linked to the plentiful active sites on the surfaces of the hybrid composition. In contrast, the subsequent stage of sorption shows diminished and slower radionuclide uptake, likely due to the gradual saturation of the remaining active sites within the HCM. Therefore, the rapid sorption can be associated with adsorption taking place on the outer surfaces of the materials, in contrast to the extended adsorption process observed in the macropores of the composite materials.

A thorough hydrodynamic-geochemical mathematical model was developed to represent the fixation and concentration. This model provides a theoretical framework for understanding the movement of radionuclides through artificial geochemical barriers associated with liquid radioactive waste.

Key words: hybrid composite materials, sorbents, migration, radionuclide, liquid radioactive waste, mathematical modelling, swelling.

Introduction

Technological progress in operational methods within the nuclear sector is designed to improve environmental safety by optimizing waste management practices, including the recycling of radioactive materials. Upgrading infrastructure for environmental safeguarding, which includes wastewater treatment, air pollution regulation, and the storage of radioactive waste in different formats, typically requires significant financial, energy, and material investments. The objective is to reduce the ecological footprint of human actions in ways that are economically advantageous, particularly for businesses in the nuclear energy field [1, 2].

The creation and implementation of engineering and geochemical barriers (EGB) must consider

their functional conditions. This pertains to every facet associated with the assembly of IGGB devices, encompassing material choice, composition, design, as well as manufacturing and operational processes. Selecting appropriate materials is vital for the progression of nuclear energy and for executing preventative strategies to lessen the negative effects of NPP operations. This selection is directly influenced by the physico-chemical, physico-technical, and nuclear characteristics of radioactive waste. It is important to take into account various engineering parameters, such as electrical, magnetic, sorption, and physical-chemical properties of materials, to guarantee the effective and long-lasting operation of EGB [3, 4].

In the current functioning of nuclear power facilities, establishing migration geochemical barriers and enhancing them through technology has emerged as a primary focus. This is due to the fact that the advantages of affordable electricity and elevated energy levels across multiple scientific domains come with the negative repercussions associated with technology and healthcare, leading to the generation of substantial amounts of radioactive waste in numerous physical and chemical forms that are difficult to manage [5,6].

Choosing the appropriate matrix to immobilize radionuclides and the suitable technology to fix them are both very important parts of the branch of insulation for liquid radioactive waste (LRW). Development of matrices for curing LRW has been ongoing in many countries in the world and international scientific centers for about fifty years. Proposals have suggested a wide variety of matrices, such as glass, ceramics, glass-ceramic materials, crystalline substances, and mineral-like materials that mimic natural minerals, among others [7, 8].

The study of a targeted synthesis of new high-performance sorbents for fixation and concentration of easily migrating radionuclide forms is important as it allows for the modification of the composition of the samples obtained and facilitates "programmable" modification of their structure and texture, thus guaranteeing the necessary functional properties and sorption attributes.

The study has been carried out across several fields, concentrating mainly on their use as geochemical barriers in engineering infrastructures at storage and disposal locations for low and medium-level radioactive liquid waste. The aims were as follows:

Methodological justification for the principles and techniques of directed synthesis of hybrid compositions incorporating various fillers through the intensification of chemical, physical,

and radiation processes at the interface of mineral phases, specifically PVC ferrocyanide and polymer solutions (or monomers), with or without the presence of cross-linking agents;

Examination of the origin of phase and structural transformations in the mineral sorbent matrix upon the incorporation of transition metal ferrocyanide micro particles during various stages of in situ polymerization and their impact on the filtration-capacitance properties of hybrid composite materials;

Creation of a model for the hierarchical structure of hybrid composite materials synthesized using in-situ polymerization utilizing thermal or radiation initiation for polymer matrix bonding processes.

The selection of geochemical membrane constituents was based on multiple factors. Initially, all hydrogel materials serving as the primary matrix for the fixation of active radionuclide ions consist of carboxylic, carbonyl, and amide groups capable of forming electrostatic (Sr^{90} , Cs^{137}) and ion-coordinating (U, Np, rare earth metals (REM)) bonds with metal ions or their oxygen complexes in both acidic and alkaline environments. The hybrid composites vary in structural arrangement, texture, and composition, enabling the creation of multi-layered geochemical membranes, each capable of selectively capturing a certain type of radionuclides within its volume. So, the hybrid composite materials are applicable for the sorption of alkaline and alkaline earth metal ions and heavy transuranium and light radionuclides.

To address the issues, a mathematical model was created to analyze the swelling of hybrid composite hydrogels in water and their contraction due to the diffuse migration of radionuclides within the cylindrical coordinate system.

Results and discussion

Mathematical model of swelling of hybrid composite matrices and diffuse migration of radionuclides

Mathematical modeling of the swelling characteristics of hybrid composite matrices and the diffuse transport of radionuclides within the cylindrical coordinate framework relies on the diffusion principle [9], enabling a comprehensive account of the diffusion phenomenon with minimal data requirements [10, 11].

The change in water concentration in the area under consideration is described by the following differential equation in private derivatives:

The variation in water concentration within the specified area is represented by the following differential equation in partial derivatives:

$$\frac{\partial C_w}{\partial t} = D\Delta C_w \quad (1)$$

where C_w – mass fraction of water in the medium.

D – water diffusion coefficient.

The expansion of the calculated environment is assumed to occur isotropically. The equation must consider the mathematical representation of the diffusion coefficient and the variation in sample volume during swelling. In addition, due to the nature of diffusion and water penetration,

the boundary between the sample and the water into which it is immersed, the swelling front, moves. Therefore, the field of solution of the diffusion equation changes in time and a moving boundary condition must be included. Since there is no analytical solution to this problem, the authors applied computational hydrodynamics methods [9-11].

The subsequent process of radionuclide ions interacting with the components of a hybrid composite can be exemplified as follows: (a) the formation of complexes with the carboxyl groups in the polymer matrix, leading to the release of protons, which explains the noticeable decrease in the pH of the medium, especially during interactions with Sr^{2+} ; (b) ion exchange processes between sodium and calcium ions sourced from a natural mineral and cesium and strontium ions present in an external solution, respectively; (c) isomorphous substitution reactions involving potassium cations within the complex intercalated with Cs^+ and Sr^{2+} ions (ref. to Figure 1). Furthermore, there is an incorporation into the cubic structure of copper ferrocyanide particles intercalated in clay, particularly with cesium ions. This is associated with the compatibility of the dehydrated Cs^+ diameter to the size of the entrance windows of transition metal ferrocyanides and the comparatively low solubility of cesium and strontium forms of ferrocyanide complexes [12].

This study was performed as a model system for further mathematical modeling and discussion of the results of radionuclide sorption from real liquid radioactive waste produced by the research reactor WWR-K.

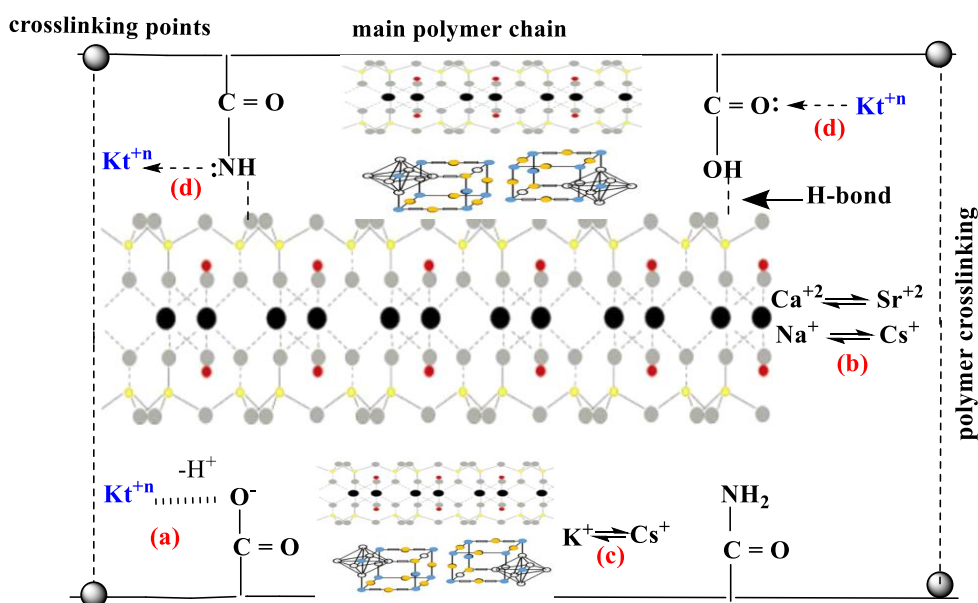


Figure 1. Schematic diagram of the sorption of cesium and strontium ions by percolated complexes $\text{P}[\text{AAm-AAc}]\{\text{BT}:\text{K}_{4-x}\text{Cux}[\text{Fe}(\text{CN})_6]\}$ [12]

The modeling of swelling caused by diffuse migration suggests that, under certain conditions, the sample undergoes a 400-fold increase in volume (see Fig. 2), which is consistent with experimental observations regarding the swelling kinetics of hybrid compositions [12]. The rate

of swelling is directly related to the amount of water present in the sample. The time needed to reach peak saturation is influenced by the size and shape of the sample, as well as the diffusion coefficient (D). The diffusion factor is established based on experimental findings.

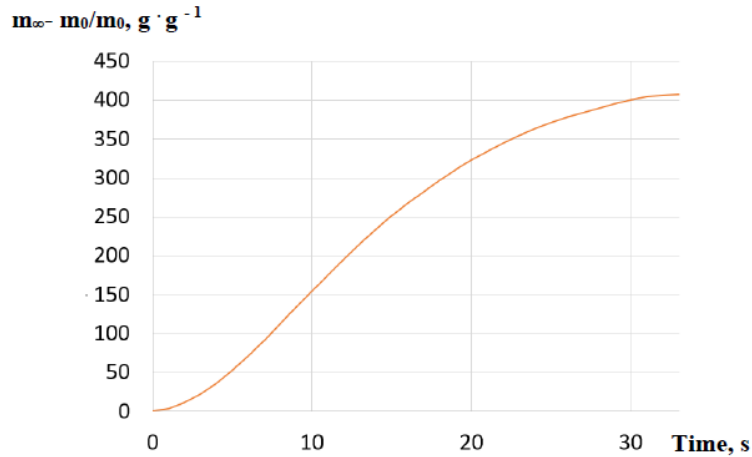


Figure 2. Change in mass of the sample of HCM over time due to swelling

Figures 3 shows that the change in sample volume is nonlinear. The lengthening of water migration time caused by a volume shift is due to the growth of the housing matrix and, in simulations, the computational grid. The behavior of this expansion depends on the current sample size.

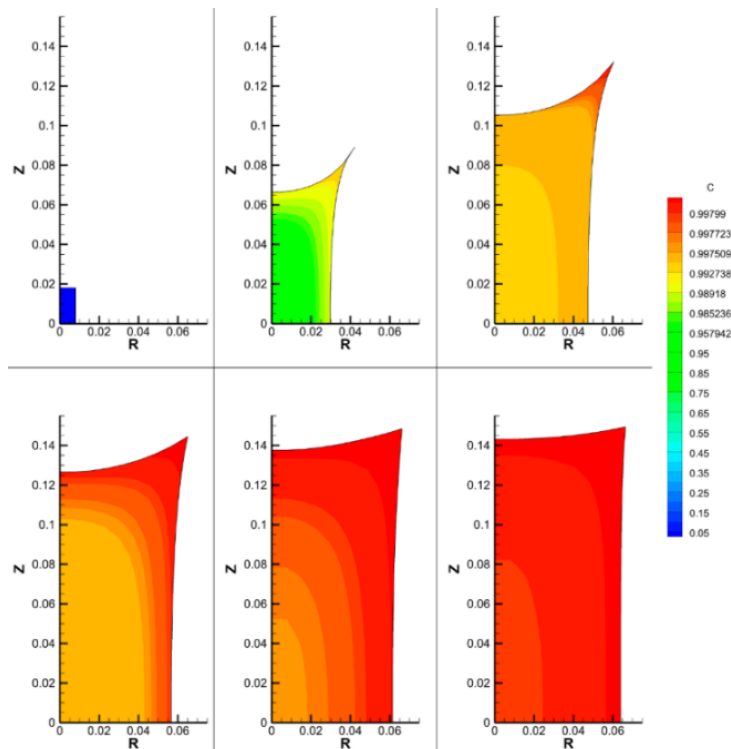


Figure 3. Deformation of the area and change in the concentration of water in the sample over time as a result of swelling

The top right corner (see Fig. 3) is extended due to heightened concentration of hydrated active reactive centers of HCM in this region resulting from an enlarged contact surface with water. Nonetheless, upon substantial water saturation of the sample, a partial restoration of the original shape occurs, as evidenced by the accompanying images.

The release of water takes place due to a chemical interaction between radionuclides and the active centers of polymer-mineral constituents of HCM. Utilizing the swellings model along with the law of active masses, we can outline the overall framework of the process as a set of differential equations with respect to partial derivatives, considering the kinetics of chemical reactions during the release of water and the capture of radionuclides:

– for components undergoing diffusion migration

$$\frac{\partial C_w}{\partial t} = D \left(\frac{\partial^2 C_w}{\partial r^2} + \frac{1}{r} \frac{\partial C_w}{\partial r} + \frac{\partial^2 C_w}{\partial z^2} \right) - H(C_{rl}) k C_{rl} C_w \quad (1)$$

$$\frac{\partial C_{rl}}{\partial t} = D \left(\frac{\partial^2 C_{rl}}{\partial r^2} + \frac{1}{r} \frac{\partial C_{rl}}{\partial r} + \frac{\partial^2 C_{rl}}{\partial z^2} \right) - H(C_{rl}) k C_{rl} C_w \quad (2)$$

– for solid component:

$$\frac{\partial C_{rs}}{\partial t} = H(C_{rl}) k C_{rl} C_w \quad (3)$$

where C_{rl} – concentration of migrating radionuclides;

C_{rs} – concentration of fixed radionuclides;

k – reaction rate constant,

$H(C_{rl})$ – The Heaviside step function defined as:

$$H(C_{rl}) = \begin{cases} 0, & C_{rl} < C_{rl}^* \\ 1, & C_{rl} \geq C_{rl}^* \end{cases} \quad (4)$$

where C_{rl}^* – the critical value of migrating radionuclides in the release process.

Figure 4 illustrates that the weight of a composite material sample is discharged on the "plateau" following its peak saturation with water. When the radionuclide solution is added to the system beyond its critical concentration, a change in the composite material is noticed, resulting in the expulsion of water molecules from the sample. The concentration of radionuclides introduced into the sample is denoted as C_{rlinj} . The critical concentration of radionuclide is defined by setting up a Donnan's equilibrium between the quantity of counter-ions within the composite volume and the concentration of radionuclides in the surrounding environment [13].

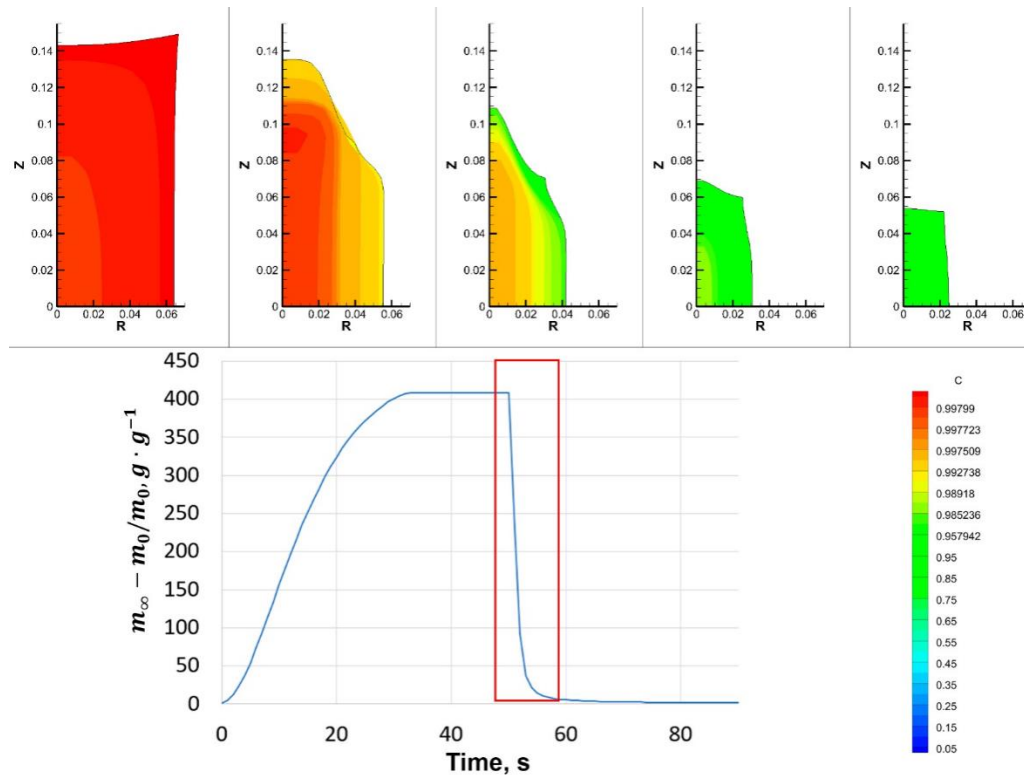


Figure 4. Change in mass of the sample over time due to swelling and release of water due to the diffusion of radionuclide molecules into the HCM

The water content in the sample subsequently decreases sharply, returning the sample to its initial value. The characteristics of the drop are directly influenced by the diffusion coefficient D , reaction rate constants k , and the concentration of C_{rl} radionuclides. The input and output data are derived from real experiments [12].

Besides, see fig. 4 also illustrates that the concentration of radionuclides remains at zero until maximum saturation with water molecules of HCM is achieved, then upon attaining the critical concentration of radionuclides in the mobile C_{rl} form, the sample volume diminishes as a result of their interactions with the functional groups of composite.

A mathematical model was developed to describe the migration and fixation processes of radionuclides in both natural and artificial geochemical barriers at radioactive waste burial sites. The model utilizes the diffusion equation, which is addressed on a time-deformed grid. The numerical model is addressed using the explicit Eulerian scheme through the finite difference method. The numerical results indicate that the model effectively captures variations in mass and volume of samples over time.

Geochemical barriers can be made up of multi-layered hydrogel membranes that are made on a base of polymer matrix (polyethylene, polypropylene, and polyvinyl chloride) so that they are easy to use in a range of conditions. They can be well placed in containers of different shapes (rectangular cassettes for use in storage tanks for HRC, cylindrical for creating

"traps" for radionuclides in wells), which can be extracted from the radiation environment by special discharge devices in automated software mode (ASM) as the maximum permissible concentration of LRW is reached in storage tanks and underground groundwater (see Fig. 5 and Fig. 6).

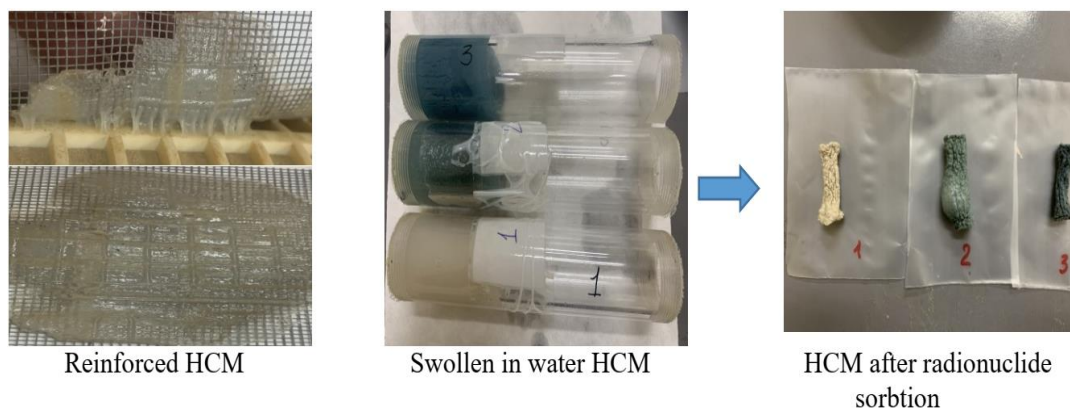


Figure 5. Design of hybrid composite materials in the creation of geochemical barriers

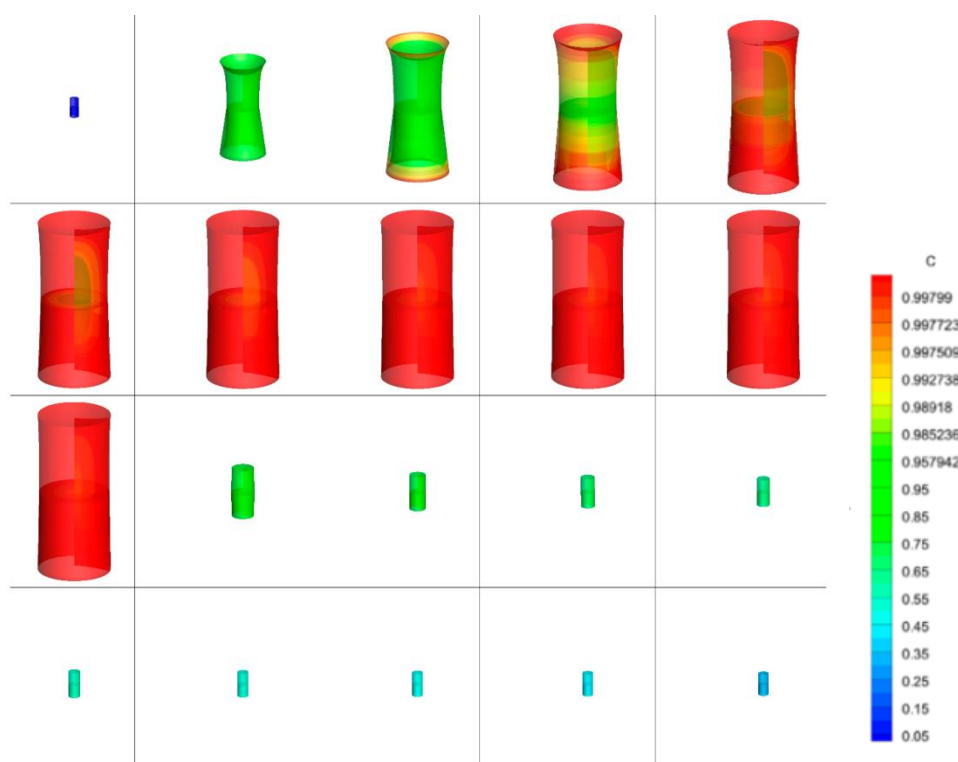


Figure 6. Three-dimensional deformation of the area and change in the concentration of water in the sample over time as a result of swelling and release under radionuclide sorbtion

Consequently, the following outcomes were achieved: simulation of sample swelling due to diffuse migration; formulation of equations governing the swelling state of samples during diffuse migration; creation of a mathematical model for diffusion migration in a cylindrical coordinate system; and development of a numerical model for diffusion migration. A model was constructed to represent the alteration of the computation grid due to diffuse migration in the transformation of the sample shape.

Thus, the hydrogel composite materials with intercalated mineral fillers for the development of a geochemical anti-migration barrier were discussed. Their effectiveness in tackling challenges associated with preventing LRW dispersion in soil and groundwater has been evaluated. This geochemical barrier will serve as a dependable safeguard against pollutants for extended durations, consequently lowering the expenses related to the remediation of nearby areas around industrial facilities.

Conclusion

The studies yield the following conclusions:

Synthesis and characterization of a novel class of hybrid composite materials consisting of intercalated complexes of natural minerals (bentonite, monthillonite, zeolite, and rutile) with ferrocyanide complexes of transition metals (FCTM) (phase I) and a volume-embedded polymer matrix (phase II) have demonstrated significant sorption efficiency for liquid radioactive waste.

The polymer matrix works well with different mineral fillers and their complexes that are intercalated with FCTM. This makes the hybrid composition more useful by working together better. This improves the efficiency of engineering structures during the design and construction of low- and medium-activity RN storage facilities.

A comprehensive hydrodynamic-geochemical mathematical model was formulated for the fixation and concentration processes, elucidating the migration of radionuclides via natural and manmade geochemical barriers at low-level and intermediate-level radioactive waste disposal sites. The model relies on solving the diffusion equation for water molecules and hydrated radionuclide ions in a time-deformed grid, which allows for the calculation of penetration depth through the HCM layer, the direction of radionuclide migration, and the prediction of the long-term performance of hybrid composite materials at liquid radioactive waste storage and disposal sites.

The contributions of authors

Mamytbekov G.M. – conceptualization, writing original draft preparation;

Shayakhmetov N.M. – methodology;

Kurmanseiit M.B. – software;

Issabayev N.B. – validation.

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Гибридные композитные матрицы для предотвращения распространения радионуклидов в окружающую среду

Аннотация. Обсуждается вопрос об эффективном создании гибридных композиционных материалов (ГКМ) с высокой сорбционной способностью по отношению к радионуклидам в жидких радиоактивных отходах. Эти гибридные композитные материалы состоят из двух сорбентов, которые готовятся из органического и природного минерального сырья, содержащего интеркалированные ферроцианидные комплексы переходных металлов. Высокая степень поглощения радионуклидов (РН) отмечается на начальной стадии сорбции, которая связана с наличием многочисленных активных участков на поверхности гибридного композиционного материала. Напротив, последующая стадия сорбции показывает снижение и замедление поглощения радионуклидов, вероятно, из-за постепенного насыщения оставшихся активных участков в НСМ.

Была разработана детальная гидродинамико-геохимическая математическая модель для представления механизмов фиксации и концентрирования радионуклидов, которая объясняет движение РН через искусственные геохимические барьеры в составе жидких радиоактивных отходов.

Ключевые слова: гибридные композитные материалы, сорбенты, миграция, радионуклиды, жидкие радиоактивные отходы, математическое моделирование, набухание.

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Радионуклидтердің қоршаған ортаға таралуын тежейтін гибридіті композиттік матрицалар

Андатпа. Сұйық радиоактивті қалдықтардағы радионуклидтерге сорбциялық қабілеті жоғары гибридіті композиттік материалдарды (ГКМ) тиімді алу тәсілдері талқыланады. Бұл гибридіті композиттік материалдар құрамында металдардың интеркаляцияланған ферроцианидіті комплекстері бар органикалық және табиғи минералды шикізаттан дайындалатын екі сорбенттен тұрады. Сорбцияның бастапқы кезеңінде радионуклидтердің (РН) сіңірілуінің жоғары дәрежесі байқалады, бұл гибридіті композиттік материалдың бетінде көптеген белсенді учаскелердің болуымен байланысты. Керісінше, кейінгі сорбция сатысы радионуклидтердің баяу сіңірілуін көрсетеді.

Сұйық радиоактивті қалдықтардың құрамындағы жасанды геохимиялық кедергілер арқылы РН қозғалысын түсіндіретін гидродинамикалық-геохимиялық математикалық модель әзірленді.

Түйін сөздер: гибриді композициялық материалдар, сорбенттер, миграция, радионуклидтер, сұйық радиоактивті қалдықтар, математикалық модельдеу, ісіну.

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