USE OF ZEOLITES FOR DECONTAMINATION OF RADIOACTIVELY CONTAMINED WORKING SURFACES

A.Yu. Lonin[1*]***, A.P. Krasnopyorova***[2]
[1]*National Science Center "Kharkov Institute of Physics and Technology", 61108, Kharkov, Ukraine*
[2]*Kharkov National University, Kharkov, Ukraine*
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The possibility of using zeolites in decontaminants applied for cleaning up radioactively contaminated working clothes and surfaces is studied. It has been established that zeolites can be used for decontamination of working clothes, as well as, working surfaces presented by metals, ceramic tile, wood coated with paint or varnish, glass. The data on different zeolite-based decontaminant in comparison with a known detergent "Zashchita" are given.

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1. INTRODUCTION

During employment of nuclear power objects one of the methods providing the safety of personnel is radioactive decontamination of working clothes and surfaces followed by the radioactive waste utilization and disposal. Formerly, for decontamination of working surfaces and materials one used different reagents selected with taking into account the properties of surfaces and materials subjected to processing. For equipment decontamination used are alkali, permanganate of potassium and oxalic acid. Decontamination of rooms is performed by means of the water solution of oxalic acid with addition of hexametaphosphate and sulphonol. For decontamination of working clothes and footwear one uses the solutions containing salt-forming components: oxalic acid, sodium carbonate, sodium phosphate and detergents (sulphonol, soap).

Solutions, obtained after decontamination have had a large volume and required an additional treatment in the process of radwaste utilization and disposal [1-3].

In resent years, adsorption agents are widely used for radioactive decontamination. They are specified by the universal use in regard to working surfaces and materials and allow one to reduce substantially the decontamination sinks. At the most part of Ukrainian APPs ones use for decontamination of equipment, rooms and personnel an ion-exchange resin-based detergent "Zashchita" (produced in Russia). However, the use of adsorption agents has some disadvantages:

- low decontamination coefficient;
- presence of an abrasive effect for chrome-plated surfaces and surfaces of glass and organic glass;
- necessity in the additional charge for waste utilization associated with ion-exchange resin decomposition and subsequent immobilization of radioactive waste.

A material, used in this work, was natural zeolite - clinoptilolite and synthetic zeolites. The choice of zeolites was determined by the following factors:

Firstly, the authors of [1,2] regarded the ion-exchanging and adsorptive properties of zeolites considering a possibility of using them for decontamination.

Secondly, it was taken into account that the mining of natural zeolites, the resources of which in Ukraine are rather ample, is considerably cheaper than the production of ion-exchange resins [2].

Thirdly, Russia is carrying out a research into the use of clinoptilolite for radioactive waste utilization. At the State Scientific Center "Academician A.I.Leypunsky Institute for Energy Physics" a method of liquid radioactive waste condensation and reprocessing with the use of clinoptilolite as a sorbent was developed. Clinoptilolite is a component of the film-forming mixture, applied for radioactive decontamination of the constructional material surface, offered by the Academician A.A.Bochvar Research Institute of Inorganic Materials [4-6].

The purpose of this work was to create for decontamination of working clothes, equipment and rooms, a domestically produced zeolite-based decontaminant, having a high decontamination coefficient and being cheaper, in comparison with existing analogues, as well as, to develop a simplified utilization method.

2. MATERIALS AND METHODS

The sorption-selective properties of natural zeolite were studied in the Department of Radiochemistry and Radioecology at the Research Institute for Chemistry of the Kharkov National University. In this

*Corresponding author. E-mail address: lonin@kipt.kharkov.ua

work natural zeolite (clinoptilolite) from the Sokirnitsky zeolite deposit (Zakarpattya region) and synthetic zeolites of NaA, NaX, NaY type and erionite were used.

The capacity of zeolites to absorb isotopes of ^{137}Cs and ^{90}St was preliminary investigated. The sorption capacity of zeolites in relation to ^{137}Cs and ^{90}Sr was studied under statistic conditions as a function of pH (the pH range was from 2 to 8.5). The acidity of solutions was varied by addition of HCl and NaOH solutions.

In the experiment we used the solution of ^{137}Cs radionuclide with a specific activity of $3.18\Delta \cdot 10^6$ Bq/dm^3 without a carrier and the solution of ^{90}Sr with a specific activity of $1.76\Delta \cdot 10^6$ Bq/dm^3 without a carrier. Clinoptilolite was preliminary grinded. Such preparation greatly increases the sorbent - solution contact surface, and, as a result, the degree of clinoptilolite utilization becomes significantly higher. To investigate the sorption, a weighted amount of sorbent was mixed by means of a magnetic mixer with the solution to be studied [7,8]. For simulation of isotope contamination of working surfaces and clothes under laboratory conditions we used isotopes of caesium-137 and strontium-90, as well as, mixtures of isotopes of caesium-137 and strontium-90.

Radiometric measurements of a solid residue were carried out using an automatic $\alpha - \beta$-weigher NRR-610 "Tesla". The relative error of radioactivity measurements does not exceed 5%.

3. RESULTS AND DISCUSSION

As a qualitative characteristic of preliminary sorbent testing we used the radionuclide sorption coefficient (Ks, %) calculated by formula (1):

$$\mathrm{K}_s = \frac{\mathrm{I}_o - \mathrm{I}_p}{\mathrm{I}_o} \Delta \cdot 100, \qquad (1)$$

where I_o and I_p are the initial and equilibrium radioactivity of the solution, p/s. To estimate decontaminating properties of decontaminants we used a decontamination coefficient, characterizing the efficiency of the decontaminating composition in the process of removing the presented radionuclides from representative materials, calculated by formula (2):

$$\mathrm{K}_{dec} = \frac{\mathrm{I}_o}{\mathrm{I}_p}, \qquad (2)$$

where I_o and I_p are the initial and equilibrium radioactivity of the solution, p/s.

The results of preliminary investigations of the sorption with clinoptilolite and synthetic zeolites ^{137}Cs and ^{90}Sr are presented in Table1. The data obtained evidence on the fact that clinoptilolite possesses high sorption properties in relation to the radionuclide of ^{137}Cs at every pH values (from 97 to 99.5% of capture). It has been found that the sorption properties of synthetic zeolites in relation to ^{137}Cs have significantly lower values. The sorption properties of zeolites in relation to ^{90}Sr exhibit a distinct dependence on pH. The sorption properties of clinoptilolite in relation to ^{90}Sr are lower than the sorption properties of synthetic zeolites. The pH influence on the sorption capacity of zeolites is due to the fact that these sorbents, similarly to other aluminum silicates, are polyfunctional subacid ionites. The mechanism of interaction of zeolites with ^{137}Cs and ^{90}Sr is submitted in works published earlier [7,8].

In the course of preliminary investigations a high selectivity of clinoptilolite in relation to ^{137}Cs was found. However, to extend the list of radionuclides subjected to sorption it is necessary to introduce synthetic zeolite into the decontaminant.

In the course of preliminary investigations a high selectivity of clinoptilolite in relation to ^{137}Cs was found. However, to extend the list of radionuclides subjected to sorption it is necessary to introduce synthetic zeolite into the decontaminant. In the course of subsequent testing different clinoptilolite and synthetic zeolite ratios were studied.

A possibility of zeolite use for fabric decontamination was studied. Fabric articles to be decontaminatied were wetted in the zeolite-based decontaminant with periodic stirring. Experimental results have shown that the compounds offered are stable in four cycles of fabric decontamination, and that the deactivation coefficient of the decontaminant "Zashchita" is sharply decreased after repeated decontamination (Table 2).

For decontamination of solid surfaces (metal, ceramic tale, wood coated with paint or varnish) we applied a decontaminant with a small amount of water sufficient for the pasty mass formation. The decontamination wastes are easily mixing and drying that allows conducting radwaste disposal similarly to solid waste disposal.

Results on use of compounds offered for decontamination of solid surfaces demonstrate that these decontaminants excel, by the decontamination coefficient, the existing analogue the decontaminant "Zashchita" (Table 3). The highest efficiency was observed for compounds No 7 and No 8.

In the course of carrying out experiments on decontamination of chrome-plated surfaces and surfaces of glass and organic glass it has been established that there is no abrasive effect after processing such surfaces with a zeolite-based decontaminant.

CONCLUSIONS

The proposed composition of the decontaminant is characterized by the low cost since it contains a natural component clinoptilolite, the resources of which in Ukraine are very ample. Besides, the open mining of this mineral considerably reduces the manufacturing cost of the detergent. The solutions obtained after decontamination with the use of zeolite occupy a much smaller volume than the solutions obtained after application of the decontaminant "Zashchita". Moreover, these detergents provide appreciably lower expenses for radwaste utilization and disposal (clinoptilolite is strongly immobilized by radionuclides and can be utilized as a base for radionuclide disposal). Proposed compounds for zeolite-based decontamination have no limitations as for chrome-plated surfaces and surfaces of glass and organic glass due to the absence of an abrasive effect. The zeolite-based decontaminant is applicable for repeated decontamination of multiply contaminated fabric surfaces.

Table 1. *Coefficient of radionuclide sorption (Ks ,%) by zeolites with different pH values at a temperature of 293.15 K*

Zeolite	Coefficient of ^{137}Cs sorption (Ks), %			Coefficient of ^{90}Sr sorption (Ks), %		
	pH =2	pH =7	pH =8.5	pH =2	pH =7	pH =8.5
clinoptilolite	97.0±4.2	99.0±4.8	99.1±4.8	23.3±1.1	88.9±4.3	94.1±3.9
erionite	98.9±4.3	99.0±4.8	98.2±4.3	26.4±1.3	91.0±4.2	88.3±4.0
NaA	95.0±4.0	95.3±4.0	96.1±4.1	14.2±0.7	96.8±4.3	92.1±4.0
NaY	97.2±4.2	98.8±4.3	98.0±4.3	12.5±0.6	96.0±4.3	81.0±3.8
NaX	90.1±4.0	94.2±3.9	96.1±4.1	32.1±1.5	94.2±3.9	85.3±3.9

Table 2. *Coefficient of fabric decontamination after I - IY contamination-decontamination cycles*

Decontaminant	Contamination-decontamination cycles			
	I	II	III	IV
Compound No 1	19.6±0.7	17.5±0.8	17.0±0.8	11.7±0.5
Compound No 2	20.2±0.9	17.8±0.8	17.4±0.8	14.5±0.6
Compound No 3	20.6±0.9	19.2±0.9	19.0±0.9	15.3±0.7
Compound No 4	23.2±1.0	20.4±1.0	20.0±1.0	17.5±0.8
Compound No 5	21.5±1.0	19.4±0.9	19.0±0.9	16.2±0.7
Compound No 6	25.0±1.2	23.8±1.1	23.4±1.1	18.4±0.8
Compound No 7	25.8±1.2	25.0±1.2	24.6±1.2	19.3±0.9
Compound No 8	27.2±1.3	25.2±1.2	23.4±1.1	20.0±1.0
"Zashchita"	20.0±1.0	10.0±0.5	9.6±0.5	7.0±0.4

Table 3. *Coefficient of work surface decontamination (metal, ceramic tile, wood, glass)*

Decontaminant	Decontamination coefficient			
	For metal	For ceramic tile	For wood (coated with paint or varnish)	For glass
Compound No 1	53.0±2.6	47.3±2.4	45.5±2.2	53.0±2.6
Compound No 2	53.7±2.6	48.0±2.4	45.8±2.3	53.8±2.6
Compound No 3	56.2±2.7	50.4±2.5	48.7±2.4	56.4±2.7
Compound No 4	62.0±3.1	56.5±2.7	54.8±2.5	62.0±3.1
Compound No 5	60.0±3.0	54.7±2.5	51.6±2.4	60.4±3.0
Compound No 6	64.5±3.2	58.0±2.8	55.0±2.6	64.7±3.2
Compound No 7	66.7±3.3	60.0±3.0	58.2±2.8	66.8±3.3
Compound No 8	67.0±3.4	62.0±3.1	60.5±3.0	67.0±3.4
"Zashchita"	19.0±1.0	18.0±0.9	13.5±0.6	19.0±1.0

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ИСПОЛЬЗОВАНИЕ ЦЕОЛИТОВ ДЛЯ ДЕЗАКТИВАЦИИ РАБОЧИХ ПОВЕРХНОСТЕЙ ОТ РАДИОАКТИВНЫХ ЗАГРЯЗНЕНИЙ

А.Ю. Лонин, А.П. Краснопёрова

Исследована возможность использования цеолитов в средствах дезактивации спецодежды, рабочих поверхностей. Установлено, что цеолиты могут быть использованы для дезактивации спецодежды, а также рабочих поверхностей представленных - металлом, керамической плиткой, деревом покрытым краской или лаком, стеклом. Приведены сравнительные данные различных составов на основе цеолитов с известным дезактивирующим средством "Защита".

ВИКОРИСТАННЯ ЦЕОЛІТІВ ДЛЯ ДЕЗАКТИВАЦІЇ РОБОЧИХ ПОВЕРХОНЬ ВІД РАДІОАКТИВНИХ ЗАБРУДНЕНЬ

О.Ю. Лонін, А.П. Краснопьорова

Досліджено можливість використання цеолітів у засобах для дезактивації спецодягу, робочих поверхонь. Встановлено, що цеоліти можуть бути використані для дезактивації спецодягу, а також робочих поверхонь представлених - металом, керамічною плиткою, деревом покритим фарбою або лаком, склом. Наведені порівняльні дані різних складів на основі цеолітів з відомим дезоктивуючим засобом "Захист".