PHASE TRANSFORMATION INTO CERAMICS OF ZEOLITES SATURATED BY ALKALINE AND (OR) ALKALINE-EARTH ELEMENTS

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It has been shown before [1], that minerals (feldspar and feldspatoids) can be perspective matrix material for capturing of radionuclides of strontium and cesium. However, a direct synthesis of aluminosilicates, especially from the impurities, containing radioactive isotopes, needs a complicated and expensive technology. So, simpler methods of synthesis are being searched. The purpose of the work was to study a possibility of capturing of radionuclides of alkaline-earth elements - strontium and cesium being in wastes of nuclear fuel, into ceramic matrix materials by a method of phase transformation of different zeolites, preliminarily saturated by the imitators of the corresponding elements by mean of their sorption on ion-exchange columns. Synthetic industrial zeolites of NaX and NaA, having a high selectivity for Sr and Cs as well as the ratio of Si/Al = 1-1,5 there similar to that in solid solutions of feldspar and feldspatoids served as a sorbent.

Phase transformation of Sr and Cs forms zeolites was performed by the following ways: 1) annealing (in sillite furnace during 3 days) of the samples preliminarily pressed into tablets at room temperature under the pressure of about 100 kg/cm^2; 2) hot pressing of the similar samples at the original laboratory device at 800-900^{o}C and axial pressure of 150-500 kg/cm^2 during 1-2 hours.

Proceeding from the data DTA, for a durable annealing in the furnace temperature 950^{o}C was chosen as a threshold at which all the zeolites have not had melting yet, but it is possible to have all the desirable structural changes started. A selection of condition of hot pressing was worked off the initial zeolite NaX. As one can see from the comparison of the curves of dependence on duration of leaching of leach rates of SiO_2 in Fig. 1 the lowest rates are reached in the runs done at 850^o and 475 kg/cm^2. Moreover, one can see that desilicatization takes place stronger from zeolites on the basis of NaA, what agrees well the data on stability of feldspars and nephelines to the processes of leaching: nephelines (product of phase transformation of NaA-zeolite) proved to be less stable then feldspar [2]. For hot pressing of the replaced zeolites we had higher temperatures (to 900^{o}C) what turned out to be quite justified, judging by the densities of the obtained samples of ceramics. However, the increase of the axial pressure proved to be more effective.

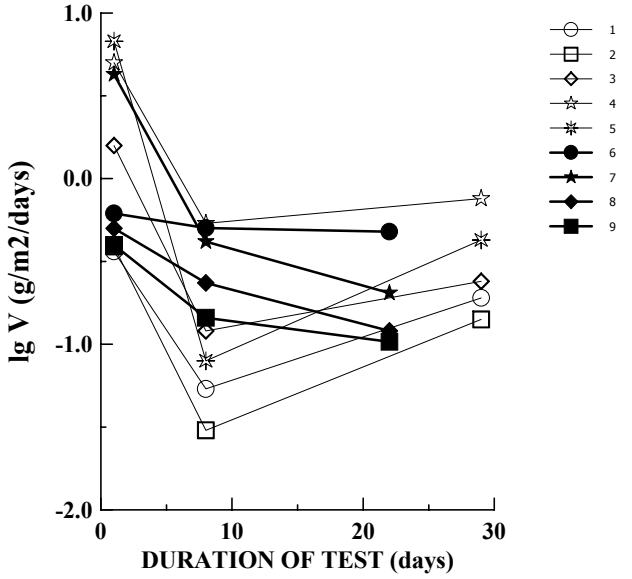

Fig. 1. Leach rates of SiO2 from different materials under static conditions used of test MCC-1 (IAEA)
Open symbol - for the ceramic samples prepared by high-temperature annealing of zeolites: 1 - NaX; 2 - SrX; 3 - CsX; 4 - NaA; 5 - CsA.
Solid symbol - for the ceramic samples prepared by hot-pressing of zeolite NaX under such technological conditions:
6 - 250 атм, 835 C; 7 - 522 атм, 800 C;
8 - 522атм, 805 C; 9 - 475 атм, 850 C.

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From the preliminarily replaced zeolites dense ceramic samples (2,1-2,7 g/cm^3) consisting of feldspar and feldspatoids, having strontium and cesium are obtained. Figures 1-3 represent the results of the investigation of these ceramics on rates of leaching (test MCC-1 IAEA). Judging by the densities of the products, as well as rates of leaching of Cs shown in Fig. 2, cesium replaced zeolites were badly pressed. Ceramic samples obtained by high-temperature annealing keep Cs better, whereas, Sr is equally slowly leached from the ceramics samples, obtained both by hot pressing and high-temperature annealing methods. The results on study of rates of leaching of Sr and Cs given in fig. 3 show that strontium is washed 10-15 times more slowly than Cs, this being observed in the runs done by both methods.

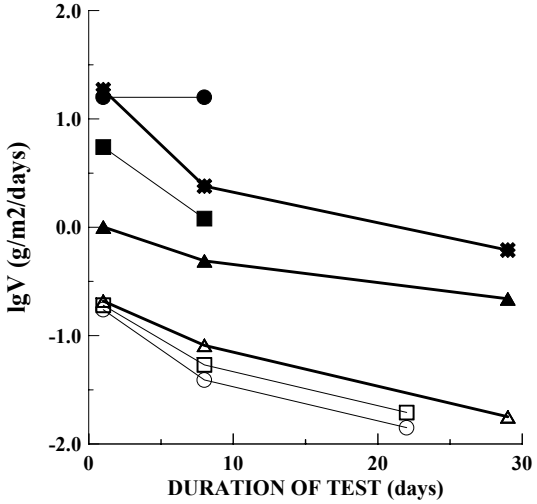

Fig. 3. The dependences on duration of test (test IAEA) of logarithms of leach rates of Sr and Cs from ceramics samples obtained by hot pressing (thin lines) or high-temperature annealing during 3 days (thick lines)

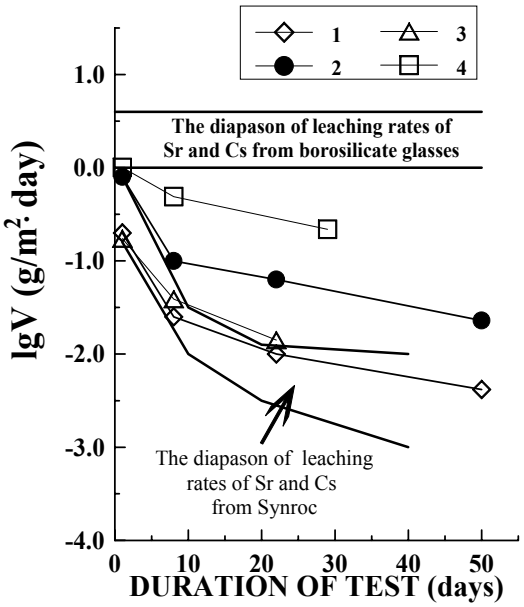

Fig. 4. The logarithmic dependences of leach rates of Sr and Cs from different ceramic materials obtained from zeolites on duration of test by method IAEA.
1 - Sr from (Na,Sr)- feldspar obtained by method of hydrotermal synthesis [4];
2 - Cs from Cs-calcisilite obtained by method of hydrotermal synthesis [5];
3 - Sr from (Na,Sr)-feldspar obtained by method of hot-pressing SrX-zeolite (runs GP-3, GP-14);
4 - Cs from pollucite-feldspar matrix obtained by method of high-temperature annealing Cs-replaced zeolite NaX (run Ft-3).

From fig. 3, representing dependences on duration of test of logarithms of leach rates of Sr and Cs from different ceramic materials one can see that the rate of leaching strontium from the matrix obtained by phase transformation of strontium form of zeolite NaX, is comparable with the rate of Sr leaching from the Synroc-C matrix [3]. Rates of leaching of Cs-bearing feldspars on cesium at short time intervals are comparable with the leach rate of Cs from borosilicate glasses [3], but within the time they decrease and in 29 days they become for cesium feldspar 20 times lower than those of glasses.

Conclusions

1. The possibility of synthesis of strontium- and cesium-bearing feldspars by two methods is shown. Here the density of synthetic samples up to 87% from theoretical one is reached.

2. Testing for leaching has show that Sr-bearing feldspars, constituting our ceramic matrices, are characterized by the "Synroc-like" form of the dependence of rate of leaching on time, i.e. in contrast to borosilicate glasses which leaching rate is constant, the of rate of leaching from feldspars decreases during the time: approximately by an order for 22 days.

3. The date of the runs on definition of rates of leaching testify to a perspective use of the these materials for immobilization of alkaline and alkaline-earth radionuclides.

References

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