

# CERAMIC MATRIX FOR IODINE RADIONUCLIDES IMMOBILIZATION FORMED ON THE BASE COPPER-BEARING ZEOLITES

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Among the demands for matrix materials for radionuclide immobilization the main thing is stability under the conditions of storage, i.e. matrices must interact minimally with the enclosing rock and not dissolve in porous and crack solutions. At the same time, a technological scheme of their production must be maximally simple, but the initial raw material for their production must be cheap and available.

Iodine is one of the most dangerous elements of RAW for biosphere. It is shown that in liquid wastes of RAW the basic form of iodine is iodide (80-98%) [1]. A potential concentrator of iodine can be sodalite, quite stable to leaching [2]. Earlier we synthesized ceramics for iodine radionuclide immobilization, one of the components of which is iodine-bearing sodalite [3]. However, this synthesis was performed in platinum capsules under high temperatures and pressures.

So we suggested radionuclide sorption method for zeolites, having cations of copper with their subsequent transformation into rock-forming minerals (sodalite, feldspar) by hot pressing method.

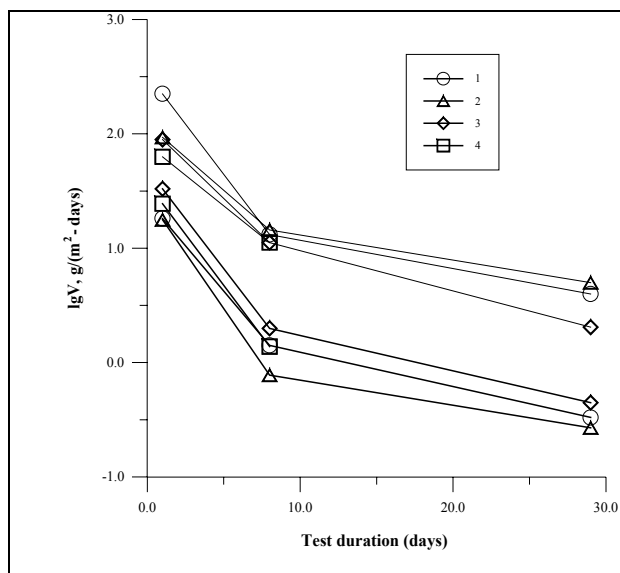
As the initial copper sorbent we chose industrial zeolite NaA. The choice was due to high selectivity of this zeolite for Cu and the relationship of Si/Al=1 in it, as in sodalities. In its turn the choice of copper for cation replacement of Na in zeolite is determined by a great similarity for the electron and a strong polarizing action of its cation at cation-radical formation. An industrial copper zeolite CuZeol, obtained in research corporation IONEX, Colorado, USA, was also used.

Before the runs the initial zeolites were ground to powder at 300°C during 3-4 hours to a constant weight. copper-bearing zeolite CuNaA was made according to the following technique: 1 M solution of HNO<sub>3</sub> was slowly added to powder suspension of the heated zeolite in water (in relationship 1:5), reducing pH to 6, to prevent Cu(OH)<sub>2</sub> formation, and then solution Cu(NO<sub>3</sub>)<sub>2</sub> was introduced with the chosen copper content for zeolite charge. After careful mixing the suspension was left to realize a replacement of Na in zeolite for Cu cations. In 2-3 days the sediment was decanted, washed by distilled water and dried at 110°C during ~20 hours. A total content of copper in the matrix is about 5 wt. %.

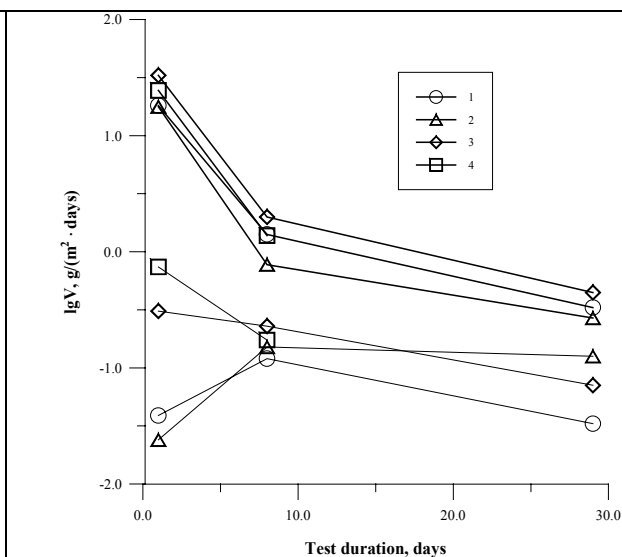
Iodine-bearing ceramics was produced as follows: the powder of zeolite CuNaA or industrial zeolite CuZeol made according to our technique, were ground with dry salt NaI or mixed with its aqueous solution. Sodium's iodide was taken in the amount, providing a total reduction of Cu<sup>2+</sup> ions, entering the structure of the initial copper-bearing zeolites up to ion Cu<sup>1+</sup>, entering CuI composition. In the second version the obtained suspension was left for the night at room temperature, then filtrated and dried at 100-110°C to powder. Then powder, preliminarily formed into tablets at hand press, was subjected to hot pressing at 815°C and axis pressure 100-150 kg/cm<sup>2</sup> at a special vessel having press form as cutting sphere. The conditions of hot pressing were chosen on the initial zeolite NaA and its copper-bearing analogue. After hot pressing the samples of the obtained ceramics were washed from the unbound iodine by ethanol (the solvent was changed to vanishing of the colouring, caused by elementary iodine).

Qualitative analysis of all samples, added by a comparison of the experimental and calculated X-ray diffraction profiles, has shown for the samples, made with dry NaI, the presence of the following I-bearing phases: sodium's iodide (NaI), iodine-sodalite (Na<sub>4</sub>Al<sub>3</sub>Si<sub>3</sub>O<sub>12</sub>I) and marshite (CuI). The reflections of Na-alumosilicate Na<sub>8</sub>Al<sub>4</sub>Si<sub>4</sub>O<sub>18</sub> are present on the diffraction diagrams of the other samples of ceramics instead of the reflections of I-sodalite.

While doing tests of the obtained ceramic matrices for the rate of leaching V<sub>i</sub>, we used the most widely spread test MCC-1 recommended by MAGATE for investigation of a wide number of matrix elements [4]. The results of the tests are given in Figs. 1 and 2 within the coordinates lg (V<sub>i</sub>) - τ (days).



**Fig. 1.** Logarithms of rates of leaching of iodine and sodium from different materials and dependences on duration of testing by technique MAGATE: 1- ceramics, obtained from the mixture of zeolite CuZeol and dry NaI; 2 - ceramics, obtained from the mixture of zeolite CuNaA and dry NaI; 3- ceramics, obtained from the mixture of zeolite CuNaA and solution NaI; 4 - ceramics, obtained from the mixture of zeolite CuZeol and solution NaI. The dependences for iodine are denoted by thick lines, for sodium - by thin lines



**Fig. 2.** Logarithms of rates of leaching of iodine and copper from different materials and dependences on duration of testing by technique MAGATE: 1- ceramics, obtained from the mixture of zeolite CuZeol and dry NaI; 2 - ceramics, obtained from the mixture of zeolite CuNaA and dry NaI; 3- ceramics, obtained from the mixture of zeolite CuNaA and solution NaI; 4 - ceramics, obtained from the mixture of zeolite CuZeol and solution NaI. The dependences for iodine are denoted by thick lines, for copper - by thin lines

Testing for leaching of I, Cu and Na has shown that the obtained ceramic matrices with the copper introduced there, are characterized by a “Synroc-like” form of dependence of leaching rate on time, i.e. in contrast to borosilicate glasses, which rate of leaching of elements is constant [4], here it decreases with time: for 29 days approximately by an order of 2 and attains by iodine (at the initial content of 20-29 wt%) the values 0,2-0,3 g/(m<sup>2</sup>·days). These values are comparable with the values of rates of leaching of this element from I-sodalite, synthesized from the mixtures of caoline clay and iodide-iodate Na/K at 1080<sup>0</sup>C [5]. A tendency of decrease of rates and degrees (a relationship of the amount of the element in the leaching for that in the initial sample) of leaching gives the right to consider that during the subsequent time the degree of leaching of iodine from these samples will not be more than 0,5%. It allows a durable preservation of iodine radionuclides, solidified into ceramics.

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