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The experimental study of leaching rates of elements (Na, K, Al, Si) from framework aluminosilicates

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For the further development of nuclear-power engineering currently the problem of high level waste disposal is strategic. The existent methods of radioactive waste immobilization are far from optimum. Modern IAEA conceptions recommend to spend a long storage of radioactive waste in solidified form in stable geological formations of Earth crust. One cannot ignore the influence of external environment on matrix stability by its distribution in the Earth crust rocks. In such a manner it is necessary to investigate the stability of mineral matrix materials in the system mineral matrix – fluid–host rock. Moreover values of leach rates of different minerals of rock able to give an information on intencity of exogenous geologic processes and time of location of rock in present conditions.

The investigation of leach rates of Na, K, Al, Si in framework alumosilicates: alkaline feldspar, nefeline and sodalite at 90ºC and at 20ºC presented in this work.

The experiment on of leachability of samples at 90ºC is based on test IAEA MCC-1. The dimensions of samples for testing were 4-5×10×10 mm. Surfaces of sample tables were polished on fine pouncing paper and then on diamond paste (20 mkm). After polishing, cleaning and drying samples were weighing on analytic balance and their geometrical proportions were measured. Geometrical square of samples surface were up to 10 cm^2, weight up to 4 g. Leachability runs were carried out in teflon autoclaves with volume of 80 ml. Pressuretight closed autoclaves were located in thermostatically controlled chamber; water level in chamber was always rise above level in autoclaves. The temperature was automatically kept at 90±1ºC. Series of experiments was carried out with duration of 1, 7, 14 and twice of 28 days. The experiments at 20ºC were carried out using the same technique, but the duration in this case was up to 520 days.

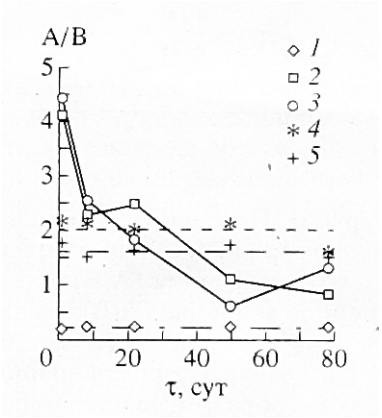
After every run period fluid was acidified with chlorine hydride and analysed by AAS and ICP methods. Sample and autoclave were carefully flushed out by triply distillate water and filled out by new portion of water.

For any element the leaching rate was estimated using following equation:

$$V_i=(l)_i \cdot W / [\,(c)_i \cdot \tau \cdot s]\ [g/(m^2 \cdot day)],$$

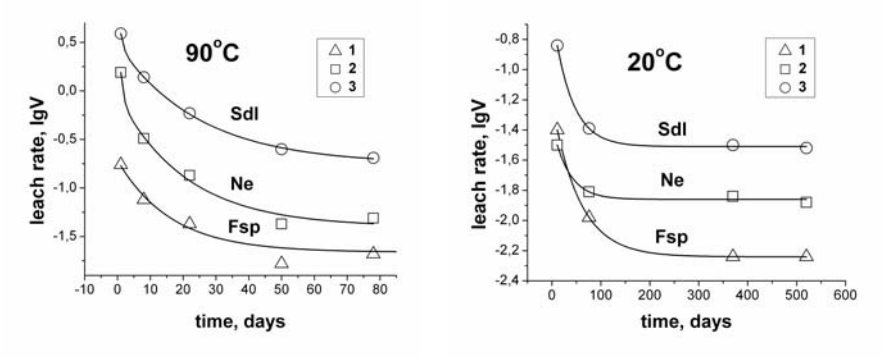
$(l)_i$ – quantity of i-element in fluid, $(c)_i$ – quantity of i-element in sample (in same unit of measurement as for fluid), W – weight of sample (gramm), τ – total run duration (days), s – Geometrical square of sample surface (m^2). Run results are presented on diagram in coordinates $\lg(V_i)$–τ (days).

It was shown that stability of minerals during leaching increase in the following chain: sodalite → nepheline → feldspar. The process of interaction of minerals with water is complicated: the leaching of feldspar is primarily incongruous, rates of enleaching of Na, K are greater then Al, Si. But after some time leaching process becomes close to congruous (fig. 1). As a result of investigation it was shown that matrix materials based on framework alumosilicates are exceed in leachability stability than borosilicate glasses and close by leaching rates to polymineral matrices Synroc (fig. 2).



A

Fig. 1 a. Kinetic relationships of enleaching elements in fluid (A/B), A – sum of alkaline and alkaline earth elements; B – (Si+Al); for minerals: orthoclase (1), labradorite (2), scapolite (3), nepheline (4), sodalite (5). Results of MCC-1 test at 90ºC



b c

Fig. 1 (b, c). Kinetic relationships of leaching rates of elements from natural minerals: 1 – feldspar, 2 – nepheline, 3 – sodalite. Results of MCC-1 test at 90ºC (b) and at 20ºC (c) in water fluid are demonstrated

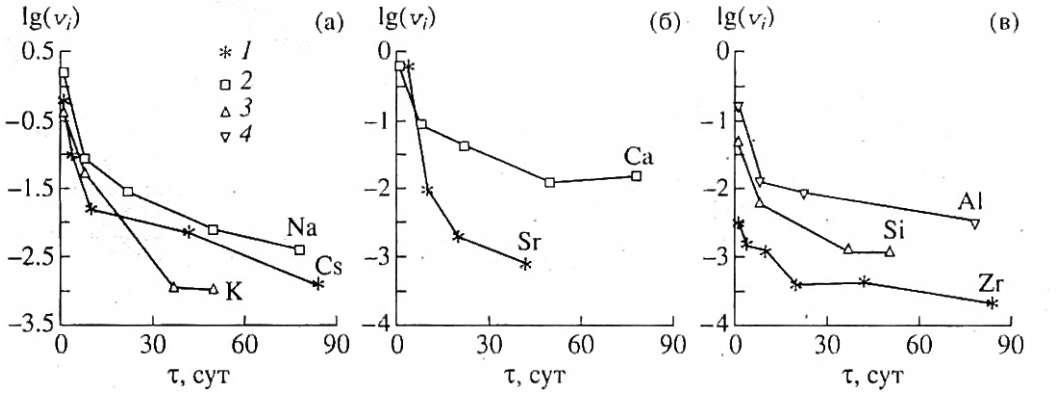


Fig. 2 (a, b, c). Kinetic relationships of leaching rates of elements from synthetic matrix materials and natural minerals. 1 – Synroc-C, 2 – labradorite, 3 – sanidine, 4 – orthoclase

Using the difference in leaching rates for different minerals the equations for estimation of water–rock sample interaction time (for 20°C) were concluded (tab. 1).

Table 1. Equations for estimation of water – rock sample interaction time (for 20°C)

Mineral pair	Equation
Feldspar - nepheline	$T\ (years) = 952.4 \cdot h_{leach}$ [1)]
Nepheline - sodalite	$T\ (years) = 344.8 \cdot h_{leach}$
Feldspar - sodalite	$T\ (years) = 253.2 \cdot h_{leach}$

[1)] h_{leach} – depth of leaching of one mineral relatively another mineral (mm)

On basis of this equations leaching rates of minerals in sample of nephelinic syenite (fig. 3), collected in coastal zone of Seydozero lake (Murmansk region) were estimated. The average values h_{leach} (for feldspar – nepheline pair) give 5±2 mm, which correspond to minimum water–rock sample interaction time for this samples about 5000 years.

Fig. 3. Nephelinic syenite collected in coastal zone of Seydozero lake (Murmansk region). Crystals of feldspar and relicts of nepheline grains are in a good light on the leached surface