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Time evolution of speciation of radionuclides adsorbed on specimens of host rock encasing a reactor

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Three major variants for decontamination of nuclear sites are considered in Russia: (i) dismantling; (ii) conversion; and (iii) burial in-place. The concept of decontamination of a shutdown industrial uranium graphite reactor (IUGR) by burial in-place was accepted by Rosatom in 2009. The safety of a buried IUGR is based on a system of protective barriers consisting of both engineered barriers and natural ones, the latter being the rocks that confine the reactor. The natural barrier capacity to function as a safety barrier depends on the geological and geochemical structure of the rock mass and its sorption properties with respect to different radionuclides.

In this work, radionuclide sorption characteristics are evaluated for rock samples taken from the location zone of a shutdown industrial uranium graphite reactor, using groundwater of a composition that is analogous to that of the groundwater near the reactor location zone, under both aerobic and anaerobic conditions.

Over the course of two years samples were investigated to determine the sorption kinetics for U(VI), Np(V), Pu(IV), Am(III), and some fission products: Co(II), Cs(I), and Sr(II). This study showed that after water-rock interactions for 1-2 months, the derived distribution coefficients (Kd) continued to increase in long-term experiments up to two years. The experiments showed that the rock masses around the IUGR have high sorption characteristics for the studied radionuclides.

To investigate the reversibility of radionuclide adsorption and the strength of the radionuclide bonds with the rock, the adsorbed radionuclide speciation and variability has been studied by the Tessier technique. The proportion of strongly fixed radionuclide species exceeds 50% of the total. The results showed that over time the speciation of radionuclides changes in two major ways: (i) the quantity of radionuclides that are strongly bound to the residual fraction and insoluble under acidic treatment increases with time, coupled with a decreasing proportion of radionuclides bound to exchange and carbonate fractions; and (ii) the quantity of adsorbed radionuclides bound to the Fe/Mn oxide fraction increased over time, at the expense of all other fractions.

This study demonstrated that the rock has reducing properties, reducing Np(V) to Np(IV) and U(VI) to U(IV), which decreases the mobility of these radionuclides in the deep geological environment. From the data obtained it is concluded that reduction by the Fe(II)-bearing minerals contained in the rocks confining the hosting massive of IUGR has occurred.

Thus, the rock mass will function as an anti-migration protective barrier with respect to the radionuclides contained in a decontaminated IUGR, including the irradiated graphite disposed in the rock mass.

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