

ASSESSMENT OF SPECIATION AND MOBILITY OF URANIUM IN ABANDONED TAILING SITE IN UKRAINE

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For decision making in choosing remediation strategies on Uranium legacy sites it is vitally important to know whether the radioactive properties of the tailing material are stable in changing environment. This report presents the results of investigation carried out on Centralny Yar (CY) tailing, one of the oldest tailings of the Pridneprovsky Chemical Plant (PChP), Dniprodzergynsk, Ukraine, formed during 1950s. After filling the tailing reservoir located at the Dnieper River terrace ravine with radioactive waste of Uranium production, it was covered by a mixture of construction debris and soils, cultivated and planted with trees and bushes.

CY tailing (area 2.4ha) contains 0.22 million tons of waste material with estimated gross radioactivity as 104 TBq. Distinctive feature of this tailing is strongly acidic reaction (pH 2.5 - 4) within the tailing body due to filling with primarily processed uranium extraction residue reportedly without applying chemical neutralization. There was no special engineering protective cover designed for both lower and upper interface of the tailing body.

In order to understand how changing environment could affect Uranium migration potential within the tailing body a method of determination Uranium chemical speciation in stored material have been deployed.

In frame of 2012 Ukrainian National Program of PChP rehabilitation geological boreholes were drilled on CY for site characterization. Samples of core material collected during drilling were subjected for further radionuclide content analysis and set of characteristic over the vertical profile samples were chosen for extended assessment. This comprised of determination chemical speciation of radionuclides in the tailings material using modified method of sequential extractions known as BCR (developed after EC Bureau of Certified Reference Material). The 4-step experimental protocol is designed to evaluate water-soluble, acid soluble, reducible and oxidizable chemical speciation of radionuclides.

Measurements of gamma-emitting radionuclides in solid samples and extraction aliquots were made using low-background HPGe detector GMX40C, ORTEC. For validation the results some representative samples have been analyzed for Uranium content using radiochemical separation and alpha-spectrometry.

Vertical profile of the radionuclides distribution in boreholes evidence existence of clearly defined stratification within the tailing body due to differences of migration properties of chemical elements in certain geochemical conditions. It is primarily concerned with most mobile Uranium (measured as U-238) and least mobile Radium (measured as Ra-226). It was found that the ratio between the two isotopes in the upper part is 5, reaching 25-30 in the middle, and less than 1 at the bottom section of the tailing. Furthermore, it was found that the front of Uranium distribution is shifted down by ~2m against Radium and its maxima correspond to the most water-saturated layer.

Taking into account specific geochemical conditions developed within the tailing body, it is reasonable to combine water and acid soluble speciation of Uranium, derived by BCR extraction protocol, into a single group (readily exchangeable form). This fraction of radioactivity could be effectively washed out from the tailings body to the local aquifer by infiltration of rainwater. With next steps of the sequential extraction protocol less soluble form of uranium could be estimated. Reducible fraction represents Uranium bound to Fe, Mn oxides and hydroxides complexes and oxidizable - in sulfides and sparingly soluble organic complexes. Residual after extraction contains Uranium in practically insoluble form.

It is clearly seen that distribution of different forms of Uranium in tailing material follow general pattern when amount of exchangeable form increases towards the bottom of the tailings. Taken that in strongly acidic conditions the main constituent in groundwater composition within and under the tailings is sulphate-ion (up to 2.4 g/l of SO₄²⁻) one can assume that Uranium predominantly migrates in form of Sulphate.

Centralny Yar Tailing is one of the topic object of radioecological concern at former PChP. Strongly acidic reaction of the media within its body promote Uranium migration outside the core into the groundwater. According to the experimental results Uranium migrates in tailing mainly in form of Sulphate. Application of sequential extraction protocol BCR revealed that the soluble form of Uranium exceed 10% of the total content in present conditions, increasing to 20-40% in ultimate geochemical composition of harsh ion exchange.

Country or International Organization

Ukraine

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