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Determination of 3H, 36Cl, 22Na, 85Sr and 133Ba by means of precipitation method

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Spent nuclear fuel from the nuclear power plants owned by TVO (Teollisuuden Voima Oy) and Fortum, is planned to be disposed at a repository at a depth of more than 400 meters in the bedrock of Olkiluoto (Eurajoki, Finland). The repository system includes multiple release barriers: the nuclear fuel, copper canister with a cast iron insert, bentonite buffer around the canister and backfilling of the tunnels. Furthermore, the surrounding rock is the last barrier if the man-made barriers fail during the passage of time. Safe disposal of spent nuclear fuel requires information about the radionuclide transport and retention properties within the porous and water-containing rock matrix along the water conducting flow paths.
The second in-situ experiment within ONKALO, the underground rock characterization facility in Olkiluoto, as part of the project "rock matrix REtention PROperties"(REPRO) was performed during 2013 using 3H, 36Cl, 22Na, 85Sr and 133Ba as tracer nuclides. The aim is to study the diffusion and sorption properties of nuclear compounds in the rock matrix under real in-situ conditions.
A straightforward way to investigate properties of rock matrix under in-situ conditions is to carry out a water phase matrix diffusion experiment (WPDE2) in a two-meter artificial flow channel along the perimeter of the drillhole. The volume and aperture of the flow channel are minimized by an impermeable cylindrical flow guide inside the packer system. The inlet and outlet positions of water are located at the opposite ends of the packed-off section. WPDE2 tracer test is performed using a slow flow rate that is generated using a piston pump. The experiment is executed using synthetic groundwater to carry the tracer solution.
This work presents the determination of radionuclides activities from the WPDE2 experiment giving the breakthrough curves of the radionuclides as a result. Rapid and precise determination of 3H, 36Cl, 22Na, 85Sr and 133Ba is of vital importance in the project. The activity of 22Na, 85Sr and 133Ba can be measured using gamma detector. Due to their electron emissions 22Na, 85Sr and 133Ba disturb the LSC measurement of 36Cl and 3H. So it is essential to seek a simple, convenient way to separate these radionuclides before spectrometric analyses.
From the initial water sample 22Na, 85Sr and 133Ba are measured by gamma spectrometry first. Then NaCl carrier is added in solution. AgCl precipitation is produced and Ag(NH3)2Cl solution is measured with liquid scintillation analyzer to obtain the 36Cl activity. Sr(NO3)2 and Ba(NO3)2 carriers are added to the supernate and 85Sr and 133Ba are precipitated as carbonates; SrCO3 and BaCO3. This supernate contains 3H and 22Na and remains of 85Sr and 133Ba. Gamma emitting nuclides are measured first and then 3H with liquid scintillation analyzer. 22Na, 85Sr and 133Ba disturb the counting of LSC measurement. Thus using the quench curves determined for each of gamma emitting nuclides in 3H window of LSC, the amount of counts from the disturbing nuclides in 3H window can be calculated and reduced from the actual 3H counts. Then the "clean" 3H activities are determined by using the 3H quench curve.

The separation procedure is optimized. The recovery of 36Cl is about 100% as well as the recovery of 85Sr while the recovery of 133Ba is slightly less, being about 85%. The detection limit for 3H in this solution is 1Bq/g. The precipitation procedure is easy and fast to separate 36Cl from the solution. 85Sr and 133Ba can be removed from the solution by carbonate precipitation in a way that 3H is measurable.

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