



Permeability and diffusion of tritium in bentonite

Galla Uroić¹, Želimir Veinović², Jadranka Barešić³, and Vjekoslav Herceg²

¹Fund for financing the decommissioning of the Krško Nuclear Power Plant and the disposal of Krško NPP radioactive waste and spent nuclear fuel, Zagreb 10000, Croatia

²Department of Mining Engineering and Geotechnics, Faculty of Mining, geology and Petroleum Engineering,

University of Zagreb, Zagreb 10000, Croatia

³Laboratory for Low-level Radioactivities, Division of Experimental Physics,

Ruđer Bošković Institute, Zagreb 10000, Croatia

Correspondence: Galla Uroić (galla.uroic@fond-nek.hr)

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Abstract. As a part of the Croatian RD&D (research, development, and demonstration) program within the Program for LILW (low- and intermediate-level waste) and SNF (spent nuclear fuel) disposal, research of permeability and the diffusion of tritium in bentonite was organized, as part of the research of the future engineering barriers for repositories of LILW and SNF. Two types of materials were prepared, namely pure sodium bentonite and a mixture of sodium bentonite (12%) with quartz sand (88%). Compaction of materials was conducted with a modified Proctor test, and the apparatus was designed and made at the Faculty of Mining, Geology and Petroleum Engineering at the University of Zagreb, utilizing fused deposition modeling 3D printer and PETG (polyethylene terephthalate glycol) material.

Water enriched with ³H has been prepared by dilution of the National Institute of Standards and Technology (NIST) ³H standard (4926e) to 6 L. The activity of the prepared enriched water was 49700 ± 180 TU. One part of the device was filled with 3 H-free water and the other with 3 H-spiked water (0.6 L volume for each part). In total, five devices with bentonite and five devices with a sand-bentonite mixture were prepared this way. The devices were thoroughly sealed to avoid loss of sample and isotopic exchange with the atmosphere and put horizontally in holders. The water for ³H activity measurements was sampled monthly or bimonthly. A total of 8 mL of the sample was taken by syringe and transferred to a measurement vial. Each vial was additionally filled with 12 mL of a scintillator (Ultima Gold LLT, PerkinElmer) and thoroughly mixed. ³H activity was determined by a liquid scintillation counter (LSC: Quantulus 1220) at the Laboratory for Low-level Radioactivities, Ruđer Bošković Institute, Zagreb. Due to the penetration of bentonite from the barrier into the water samples, tritium-free water has relatively high activity after 2 months of the experiment (average value was 5560 ± 240 TU). In the final sampling, after a 2-month time span, the activity reached 9670 ± 2490 TU. Later, sampling and measurement were not possible due to the high bentonite amount within the samples. Tritium-free samples from the sandbentonite mixture device were stable, and the first analyses (1 month after the experiment setup) showed an average activity of 533 ± 31 TU, while the final (5-month time span from the first sampling) average activity was $12\,800\pm3430\,\text{TU}$. The increase in the concentration from the beginning to the end of the experiment was almost exponential. Spiked samples from the bentonite device had an average activity of 36950 ± 2550 TU at the first sampling and 17580 ± 13320 TU after a 1-month time leg. Later, sampling was not possible. Spiked samples from sand–bentonite mixture showed a decrease in tritium activity from the first sampling $(43390 \pm 1310 \text{ TU})$ to 27210 ± 285 TU (5 months after the first sampling). Considering the penetration of bentonite material into the water samples, only the sand-bentonite mixture can be considered for further analyses through the calculation of the diffusion.

References

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