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Supplement of

Permeability and diffusion of tritium in bentonite

Galla Uroić et al.

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

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PERMEABILITY AND DIFFUSION OF TRITIUM IN BENTONITE



Galla Uroić¹, Želimir Veinović², Jadranka Barešić³, 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, Heinzlova ulica 70a, Zagreb

²Faculty of Mining, Geology and Petroleum Engineering, University of Zagreb, Pierottieva ulica 6, Zagreb

³Institute Ruđer Bošković, Laboratory for measuring low radioactivity, Bijenička Cesta 54, Zagreb
galla.uroic@fond-nek.hr

I. PROJECT GOALS:

- Define potential materials for the production of sealing protective layers;
- Design and set up permeability test—transport of radionuclides through sealing layers;
- Analyze the mechanisms of radionuclide transfer through sealing (buffer) material;
- Suggest materials for the production of sealing protective layers.

II. MECHANISM OF RADIONUCLIDE TRANSFER

Diffusion

The transport process in which the movement of a substance is a result of the gradient of its concentration, although the actual driving force is the gradient of the chemical potential of the dissolved substance. Hydraulic gradient is not necessary for the transport of pollutants through diffusion.

The flow of the observed substance within a solution sampled by diffusion can be described by Fick's law:

$$q_m = -D_M \nabla c$$

(q_m - the flow; D_M - molecular diffusion coefficient; c - the concentration of the observed substance, ∇ - Hamiltonian operator)

In mineral sealing layers, the transfer of substances caused by diffusion is slower than in a solution because the surface through which the transfer takes place in the porous medium is smaller, and the pores are curved, which increases the tracer's path. The mobility of transported substances can also be reduced due to the interaction of substances and the rock porous medium.

For the above reasons, it is necessary to define the effective D_M^E diffusion coefficient, which can be written in the form:

$$D_M^E = D_M f(n)$$

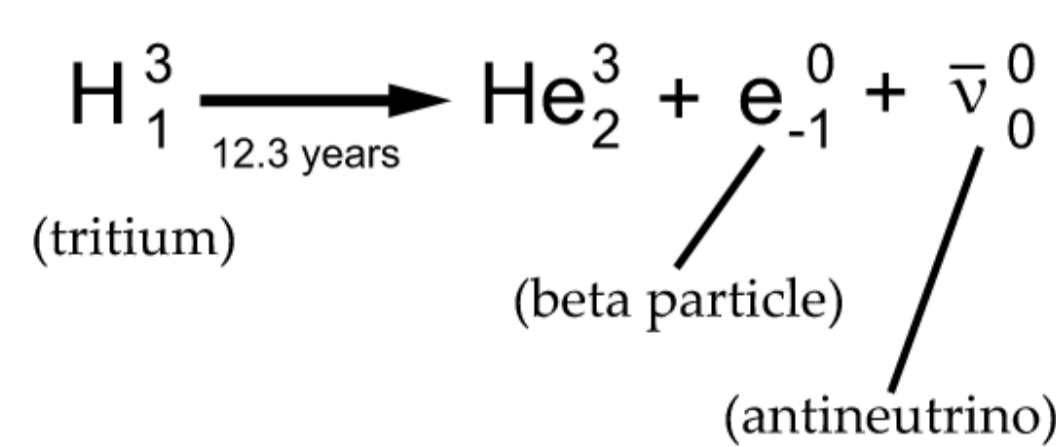
(D_M^E - effective molecular diffusion coefficient in a porous medium; $f(n)$ - function depending on the character of the porous medium)

III. MATERIALS USED IN THE EXPERIMENT

Tritium

Tritium was chosen as a tracer material due to its high mobility, low activity and favorable half-life (12.323 +/- 0.004 years) which makes it an optimal material for laboratory work.

Tritium is a radioactive isotope of hydrogen whose nucleus is made up of two neutrons and one proton. It has relatively low activity, belongs to β emitters, with decay energy 0-0.018590 MeV (average 0.0057 MeV):



Experiments with tritium were carried out in cooperation with Laboratory for measuring low radioactivity, Department of Experimental Physics, Institute Ruđer Bošković in Zagreb.

Low permeability materials

Two types of materials were prepared, namely **pure sodium bentonite** and a **mixture of sodium bentonite (12 %) with quartz sand (88 %)**. Compaction of buffer materials was conducted with a modified Proctor test.



Compacted clay in the cell



Sample compaction (Proctor test) and compacted mixture of sodium bentonite and quartz sand in mold

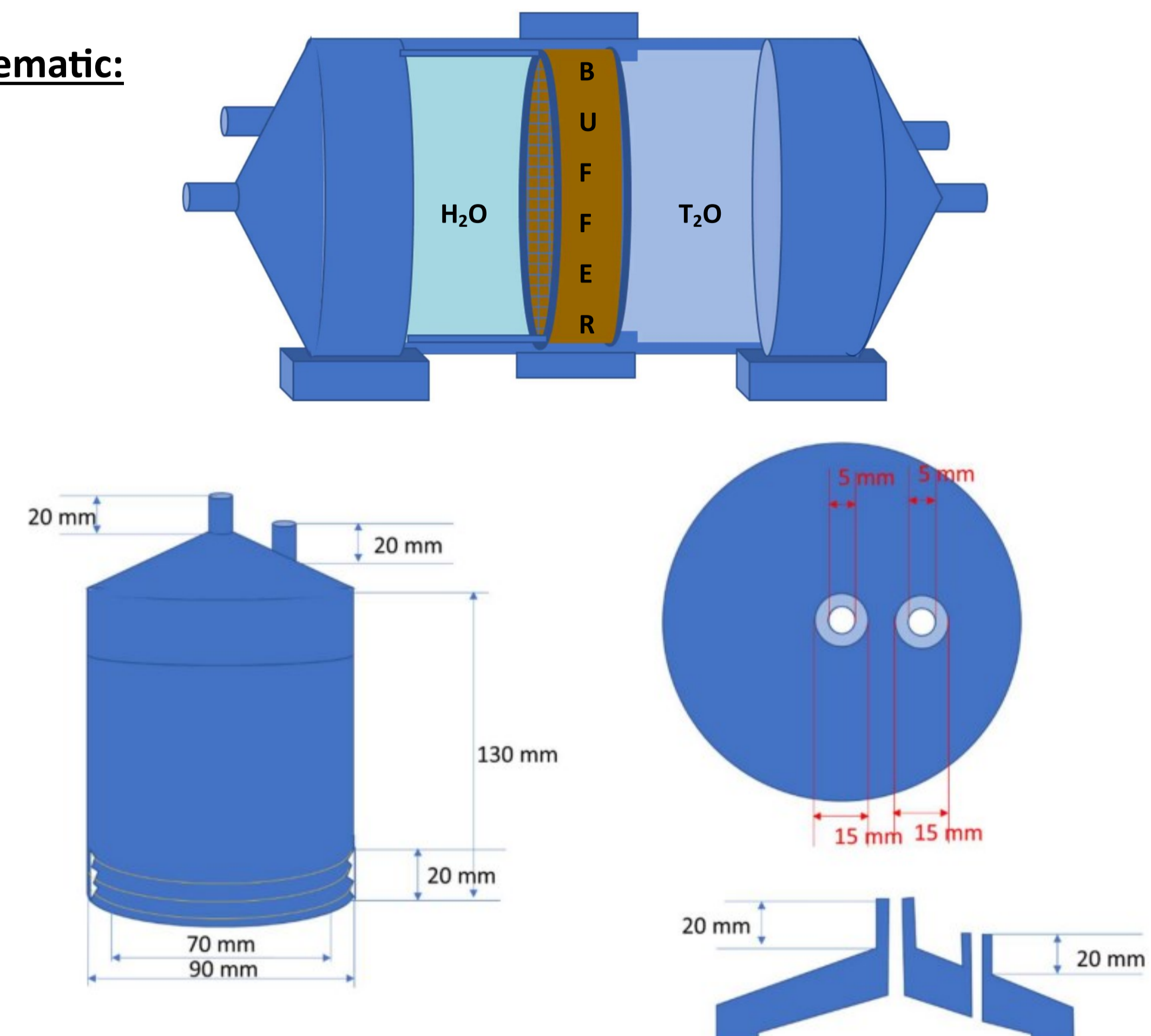


Tritium sample used in the experiment

IV. PERMEABILITY TEST CELL

The cell design was developed at the Faculty of Mining, Geology, and Petroleum engineering and underwent several modifications. The final iteration of the permeability measurement cell for tritium through the sealing layers was entirely produced using the Prusa i3 3D printer.

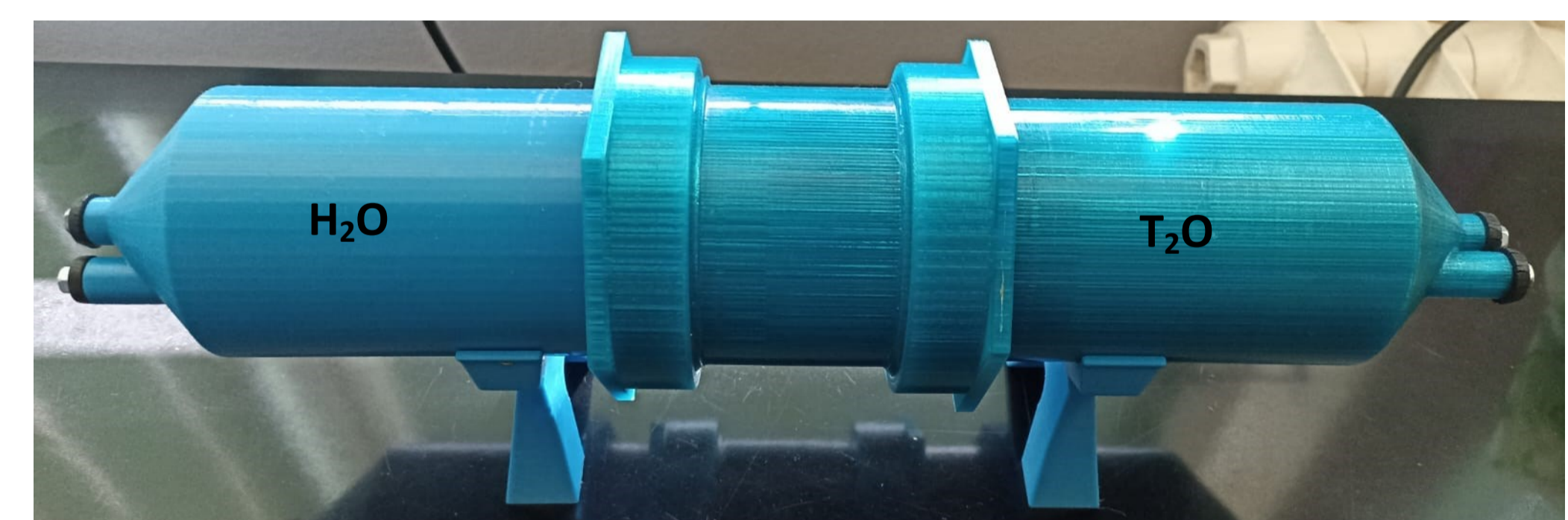
Schematic:



To avoid sealing along the cell, it was decided that the sample would be placed in the central third, with water and tritiated water stored in separate reservoirs. To facilitate the control of a sample of poorly permeable material, two meshes and two sheets of paper filter were placed at the interface with water and tritiated water.

Two openings for pouring water and steaming air are at the same height - the central one is used for steaming air, while the side one is for adding fluid - ensuring that the air can be expelled as easily as possible. A cell placed horizontally would be supported by movable bases - legs.

A cell containing a sample of buffer material and tritiated water



V. RESULTS

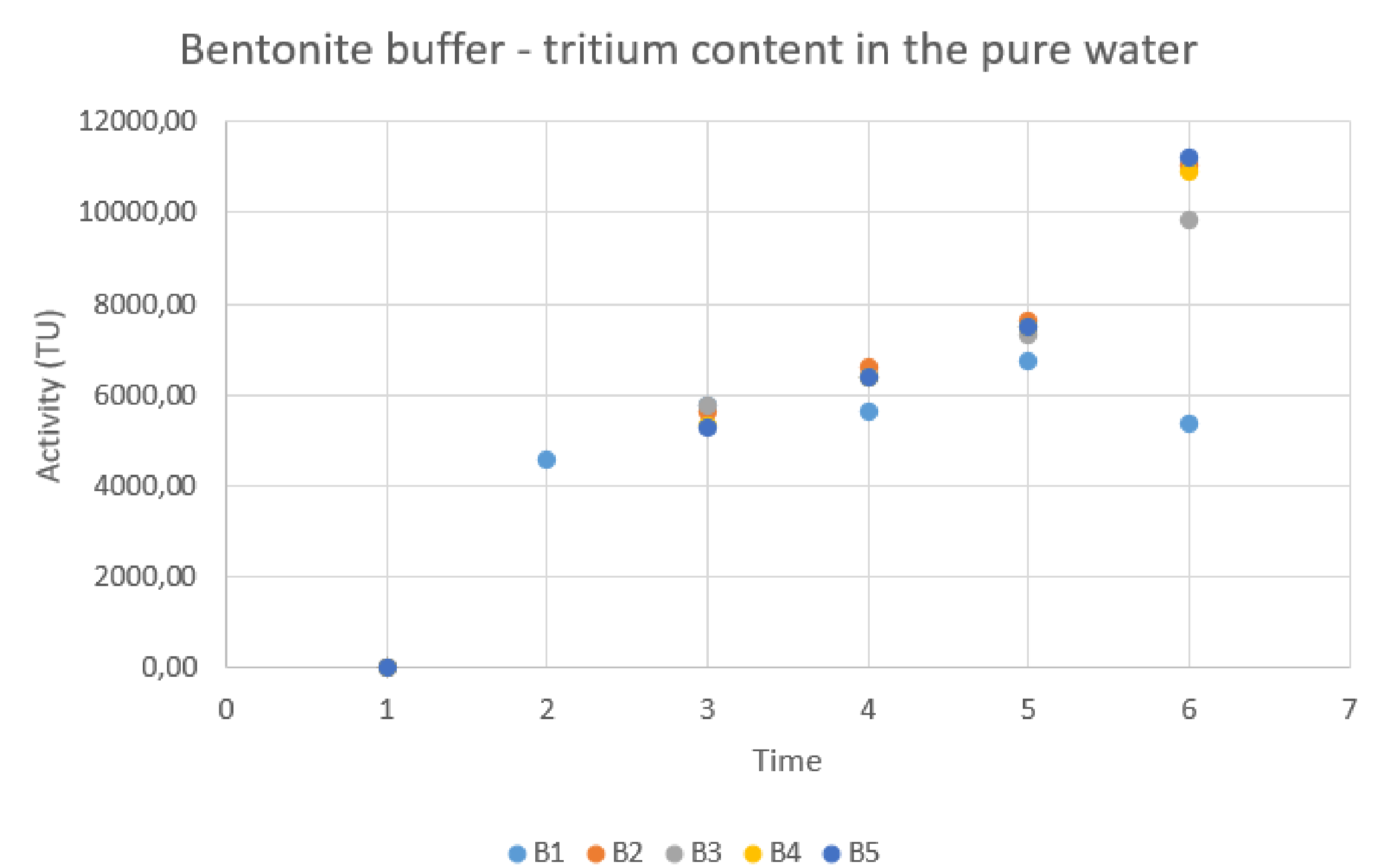
At the beginning of the experiment, the following agreements were made:

- Concentration measurements will be conducted every 2 weeks on all cells (5 with bentonite and 5 with sand-bentonite mixture);
- Measurements will be carried out simultaneously on all cells to obtain statistically processable data.

Date	B1 (TU)	B2 (TU)	B3 (TU)	B4 (TU)	B5 (TU)
16.12.2020.	0,00	0,00	0,00	0,00	0,00
19.2.2021.	4563,23				
5.3.2021.	5787,15	5644,00	5753,85	5327,40	5289,55
19.3.2021.	5628,994	6588,617	6379,675	6397,488	6395,653
1.4.2021.	6726,781	7605,716	7333,705	7483,466	7478,52
25.5.2021.	5349,041	11022,88	9835,905	10907,06	11231,86

Table - The results of tritium concentration measurements - bentonite buffer

Diagram - The results of tritium concentration measurements - bentonite buffer (tritium units)



The obtained results are as expected, given that it's a radionuclide with extremely small molecule. Based on these results, further research will be planned, including the selection of new materials (sealing materials) and methods (water with other radionuclides).