MODELLING TRITIUM TRANSPORT, MATRIX DIFFUSION AND MULTIPHASE FLOW IN A CONCRETE CELL USED FOR STORING RADIOACTIVE WASTE AT 'EL CABRIL' (SPAIN)

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Abstract *'El Cabril' is the low and intermediate level radioactive waste disposal facility of Spain. Water with high concentration of tritium is collected from a drain, which is situated at the centre of a concrete cell that stores radioactive waste, indicating flow of water within the cell. 2D numerical models have been made in order to develop a qualitative and partial quantitative understanding of the processes that causes this phenomenon. The conceptual model considers evaporation and condensation processes due to temperature gradients, matrix diffusion between a mobile zone (with advection, dispersion and diffusion) and an immobile zone (with only diffusion). Model results suggest that only the tritiated water used to prepare the mortar to refill the containers, which can be in liquid and gas phases, could leave the cell being collected in the drain.*

1 INTRODUCTION

Cementitious materials play an important role in the radioactive waste storage. Not only are they used to store low and intermediate radioactive waste, but also they are the final barrier in nuclear reactors that avoids tritium contact with the atmosphere[i]. Tritium is one of the isotopes of hydrogen, which can take part of the water molecules as tritiated water, both in liquid and gas phases. Often it is related with nuclear industry. It is for this reason that it is important to study the transport of tritium in cementitious materials. In the literature there are several studies related with tritium transport in non-saturated conditions in soils[ii, iii, iv, v]. Also, there are some studies related to the tritium diffusion in cementitious materials[i, vi, vii, viii]. Our work is focused on the concrete used for storing radioactive waste at 'El Cabril', which is the low- and intermediate- radioactive waste facility for Spain, where tritiated water is collected from the concrete cells storing the waste. A previous work studied the processes within the concrete that allow flow of water inside the cells by means of 2D numerical models. Results showed that this phenomenon was caused by capillary rise from the phreatic level, evaporation and condensation within the cell produced by temperature gradients caused by seasonal temperature fluctuations outside[ix]. However, the concentration of tritium of the collected water was not taken into account. In this work, we want to understand why the collected water contains tritium. To do so, we follow the same conceptual model used by Chaparro and Saaltink (2016), taking into account the temperature oscillations outside the cells and the capillary rise from the phreatic level. Moreover, we apply the same double porosity conceptual model used by Chaparro et al. (2015) in tracer test where the same concrete manufactured at 'El Cabril' was studied.

2 NUMERICAL MODEL

2.1 Conceptual model

Figure 1 displays the conceptual model and a scheme of a concrete cell where the low- and intermediate- radioactive waste is stored. The same conceptual model used by Chaparro and Saaltink (2016), is used, which considers that water (without tritium) could ascend from the phreatic level to the wall of the cell due to capillary rise through the unsaturated rock. In summer, the wall of the cell is hotter and the wall of the container is colder because the air gap acts as a thermal insulation. Thus, water can evaporate from the wall of the cell. Vapour diffuses through the air gap due to the temperature gradient between the wall of the cell and the wall of the container. Water condensates at the wall of the container because of its lower temperature. Then the condensed water could be mixed with the tritiated water in the container. Consequently, tritiated water runs off to the drain. In winter, the wall is colder and the container is hotter. Hence, tritiated water evaporates at the container and condenses at the wall. So, again tritiated water runs off to the drain. This occurs in summer and winter because the temperature difference across the air gap is large enough to produce this phenomenon. In addition, we also applied the double porosity conceptual model used by Chaparro et al, (2015), which considers a mobile zone with advection, dispersion and diffusion and an immobile zone with only diffusion. Moreover, the conceptual model considered isotopic fractionation and radioactive decay. The simulations were carried out using the code Code_Bright[xi]. Some changes had to be made in order to be able to simulate isotope fractionation and tritium existing in both liquid water and vapor.

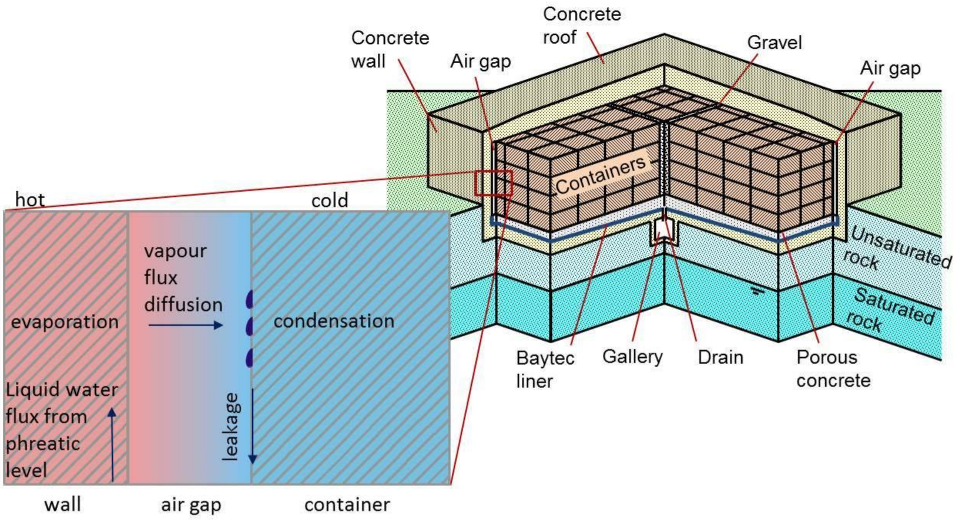

Figure 1: Conceptual model[ix]. Situation in summer: water ascends from the phreatic surface, evaporates at the hot side (wall of the cell), vapour diffuses through the air gap and water condensates at the cold side (wall of container).

2.2 Geometry, mesh and materials

First, we added the tritium transport to the same numerical model (single porosity model) that Chaparro and Saaltink, (2016), which had an axisymmetric geometry, considering the geometry of the cell as a cylinder where the volume/surface ratio was the same as the real cell. However, that geometry was too complex to apply the double porosity conceptual model, in which the porosity is divided into a mobile and immobile part. Then, before applying the double porosity, an 1D numerical model with 105 nodes and 104 one-dimensional elements was made. The model considered the wall of the cell, the air gap and the container where the waste is stored. Finally, we distinguished a mobile zone and an immobile zone in a 2D domain, the second domain representing the immobile zone. The mobile zone was represented by the *x* axis (the same as the 1D model). From each of the nodes of the mobile zone, we added a series of 10 one-dimensional elements in the *y* axis representing the immobile zone. Therefore, the whole mesh had 625 nodes and 624 elements.

2.3 Initial and boundary conditions

The initial temperature of the entire cell was 19 ºC and the initial liquid pressure was -0.9 MPa. Containers had an initial tritium concentration of 1.45×10^{-4} GBq/L. As Chaparro and Saaltink (2016) did, temperature was prescribed and varies with time at the wall of the cell, using the daily average temperature measured by the sensors situated outside the cell. The water table was simulated by prescribing the liquid pressure at 0.1 MPa. A leakage boundary condition was applied to the gap of air between the wall and the container allowing water to leave the cell only when liquid pressure exceeds atmospheric pressure. This represents the leakage to the drain.

3 RESULTS AND DISCUSSION

Figure 2 displays the flow rate. Measured data, results from the axisymmetric model[ix] and the double porosity model are compared. The two periods (summer and winter) can be distinguished. Data are only available for the two last years, which the axisymmetric model reproduces well. However, the calculated flow rate for the double porosity model is lower than the measured one. Results from axisymmetric model show that water from the water table enters the cell; with a maximum of 5 l/d during the summer. Then the wall of the cell is dryer and a larger amount of water can ascend from the water table. Nonetheless, the double porosity model shows that water enters the cell with a maximum of 1 l/d. According to the axisymmetric model, condensed water leaves the cell in two periods with large fluxes in winter (around 10-15 l/d) and small fluxes in summer (less than 5 l/d). The double porosity model shows that water leaves the cell in the same periods but with lesser amounts (around 2 l/d in winter and 1 l/d in summer). These differences could be due to the simpler geometry deminishing the effect of the temperature in the entire cell. Nonetheless, our objective is to develop a qualitative and partially quantitative understanding of the processes that yield the tritiated water in the drain. Hence, we assume this differences not to be important.

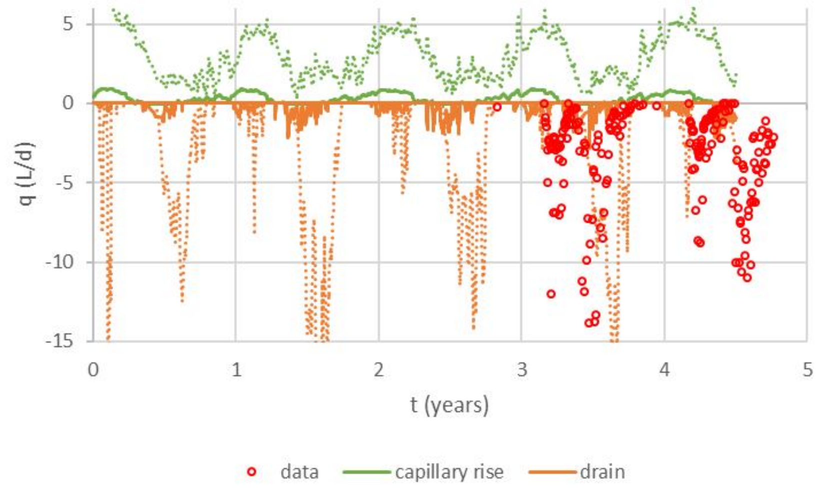

Figure 2: Flow rate of water. Positive values mean that water enters the cell, and negative values mean that water leaves the cell. Continuous lines are the double porosity model, dashed lines are the axisymmetric model and dots are the measured data.

Figure 3 compares the concentration of tritium measured in the collected water at the drain, calculated by the axisymmetric model and the double porosity model. Measured data corresponds to an average of the measured data in winter and summer respectively. The measured tritium concentrations are between 900 and 2500 Bq/l, being larger in winter because, then, the evaporation is produced in the containers. However, both model results show larger tritium concentrations in comparison to the measured data. The axisymmetric model gives concentrations between 15000 and 40000 Bq/l, and the double porosity model calculates tritium concentrations between 7000 and 30000 Bq/l. Both models show larger concentrations in winter as measured data. Applying the double porosity conceptual model, a maximum reduction of tritium concentration by a factor of 2 is obtained. This reduction of tritium concentration in the collected water in the drain could be due to matrix diffusion, but also due to the fact that the flow rate is lower. However, we think that the double porosity conceptual model is more realistic, because the concrete cell also contains iron (reinforced concrete), metals (drums) and solid waste, which is considered as immobile zone, where the transport mechanism is diffusion. The main point is the fact that both model overestimated the tritium concentration by an order of magnitude. The reason of that is because both models consider as initial tritium concentration the concentration that is in the whole cell. However, ENRESA stores tritium in the containers but also they use tritiated water for preparing the mortar used the refill the containers once all the drums are inside. Results obtained suggest that only the tritiated water used in the mortar, which can be in liquid or gas phases, could leave the cell through the drain. Moreover, only the containers near the wall can be affected by the evaporation and condensation processes, because inside the cell the temperature gradients are not high enough for causing this phenomenon.

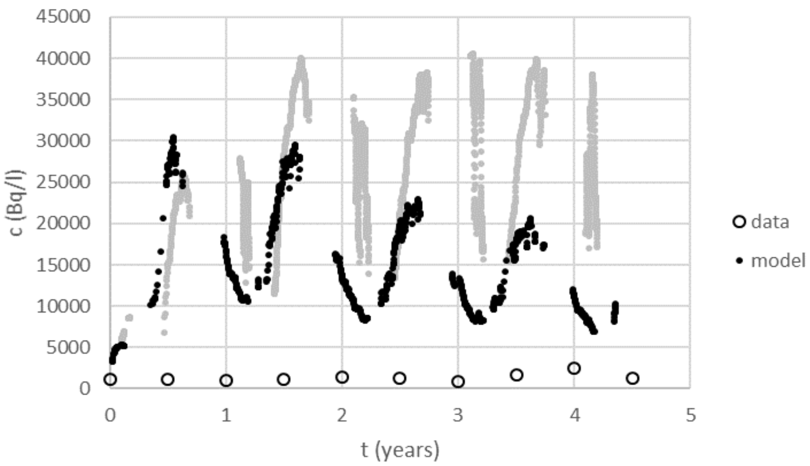

Figure 3: Concentration of tritium of the collected water. Filled black dots correspond to double porosity model, filled grey dots correspond to the axisymmetric model and empty dots are the measured data.

4 CONCLUSIONS

- Numerical models taking into account multiphase flow, matrix diffusion and tritium transport has been developed with Code_Bright in order to understand why tritiated water is collected at the drain of a concrete cell where radioactive waste is stored.
- Two numerical models have been compared, one with an axisymmetric geometry and single porosity, and another one with a simpler 2D geometry but with double porosity.
- Results suggest that only the tritiated water used to prepare the mortar used to refill the containers, which can be in liquid and gas phases, could leave the cell.

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