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Preliminary modelling of radionuclide migration in the argillaceous sediments of the Sumer Formation (Northwestern Bulgaria)

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Abstract. In Bulgaria, from the preliminary analyses performed for site selection of deep geological disposal of high-level waste (HLW) and spent fuel (SF), it was concluded that the most promising host rocks are the argillaceous sediments of the Sumer Formation (Lower Cretaceous), situated in the Western Fore-Balkan Mts. The present paper aims to compare the transport of three major radionuclides from a hypothetical radioactive waste disposal facility, which incorporates an engineering barrier of bentonite into the argillaceous (marl) medium. The simulations were performed by using HYDRUS-1D computer programme. The results are used for a preliminary estimation of argillaceous sediments as a host rock for geological disposal of HLW.

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Keywords: HLW geological disposal facility, marls, numerical modelling.

INTRODUCTION

In Bulgaria, the present national strategy for RAW management (Council of Ministers, 2015) has not made a definitive decision on final disposal of high-level waste (HLW) and spent fuel (SF), but deep geological disposal is considered to be the most ethical, sustainable and safe approach to the management of these wastes.

In the preliminary analyses performed for site selection of deep geological disposal of HLW and SF, it was concluded that the argillaceous sedimentary formations in Northern Bulgaria are perspective in this regard. According to the safety requirements for the geological environment and the degree of knowledge, the most promising host rocks are the argillaceous sediments of the Sumer Formation (Lower Cretaceous), situated in the Western Fore-Balkan Mts (Karastanev *et al*., 2011). Besides, the latter rocks have previously been assessed as having good encapsulating properties during the exploration of gas fields in this region (Monahov and Monov, 1969).

In accordance with the IAEA documents, the HLW final disposal has to apply a multibarrier approach ("defence in depth") in order to ensure the safety of the storage facility during operation and the post-closure period (*e.g.*, IAEA, 2006, IAEA, 2011a, b). Thus, being part of the approach, the main role in host rock selection for geological disposal requires the so-called safety assessment analysis, which includes model studies for an eventual migration of radionuclides from the repository (*e.g.*, Mallants *et al*., 2001, 2011; IAEA, 2006, 2013). An initial step in such type of investigations is the elaboration of a hydraulic model of the argillaceous rocks of Sumer Formation and determination of the infiltration rate (flux) throughout the potential disposal zone of the Sumer Formation for thousands of years (Tsvetkova *et al*., 2019). To date, preliminary investigations concerning some aspects of the natural conditions of the site have been made, but a mass transport model, integrating the source (*i.e.,* the disposed HLW), the engineer barrier and the argillaceous (marl) rocks with their specific characteristics, is not yet available. The aim of the study is to compare the migration of three major radionuclides from a hypothetical radioactive waste disposal facility, which incorporates an engineering barrier of bentonite into argillaceous medium.

MATERIALS AND METHODS

Geosphere and repository schematization

Geological setting

The accommodating environment of the potential storage system is composed of the marls of the Sumer Formation, representing a spatially sustained and significantly homogeneous geological environment with a total thickness of about 1,200 m (Fig. 1). It consists of dense clay marls with rare layers of sandstones, which were found in the upper part of the profile. In depth, the sandstones become even rarer, with small thicknesses, and laterally restricted. The marls have a very dense structure, composed of silt particles with clay-carbonate solder and variable carbonate content (Karastanev *et al*., 2011). The effective porosity is between 6% and 8% (Georgieva, 2016). In the uppermost part of the profile, up to ten metres, the marls are weathered, but in depth they are dense and unaltered. With the exception of the uppermost part (the weathered zone), hydraulically the marls of the Sumer Formation represent an unsaturated medium.

Assumed repository concept

For the purpose of the current analysis, it is assumed that in the hypothetical repository situated at a depth interval of 335–350 m will be disposed 800 t heavy metal in stainless containers. These containers with the respective HLW will be placed in a volume of 360 m^3, which can be represented approximately as a cylindrical object situated horizontally with a circular cross-section area of 6 m^2 and a length of 60 m. Similarly to several scientific and technical publications (*e.g.*, Westsik *et al*., 1983; IAEA, 2003; Sellin and Leupin, 2013; Kaufhold and Dohrmann, 2016), a bentonite engineering barrier around the canisters is introduced. The adopted herein bentonite barrier is situated around the general cylindrical body with HLW and has a thickness of 6 m all around it. This barrier, together with the storage canisters, can also be represented as a cylindrical body with a volume of 12,297 m^3.

According to Oversby (1986), one hundred years after the disposal of HLW from Pressurized Water Reactor type (as Kozloduy NPP), some of the main radionuclides are: ^{137}Cs with activity of 10,300 Ci/MTHM (curie per metric ton of heavy metal); ^{90}Sr with activity of 6,710 Ci/MTHM; ^{241}Am with activity of 3,750 Ci/MTHM. These isotopes are selected to be the subject of simulations in the present study. As a scenario for the applied modelling procedure, for the repository plus the bentonite barrier, an effective medium porosity of 10% is arbitrarily used, which after 100 years of operation of the storage will be filled with dissolved radionuclides. This assumption is actually part of the so-called conservative approach that takes into account the most unfavourable value in the case of porosity. Based on the above data and parameters of the storage facility, the following

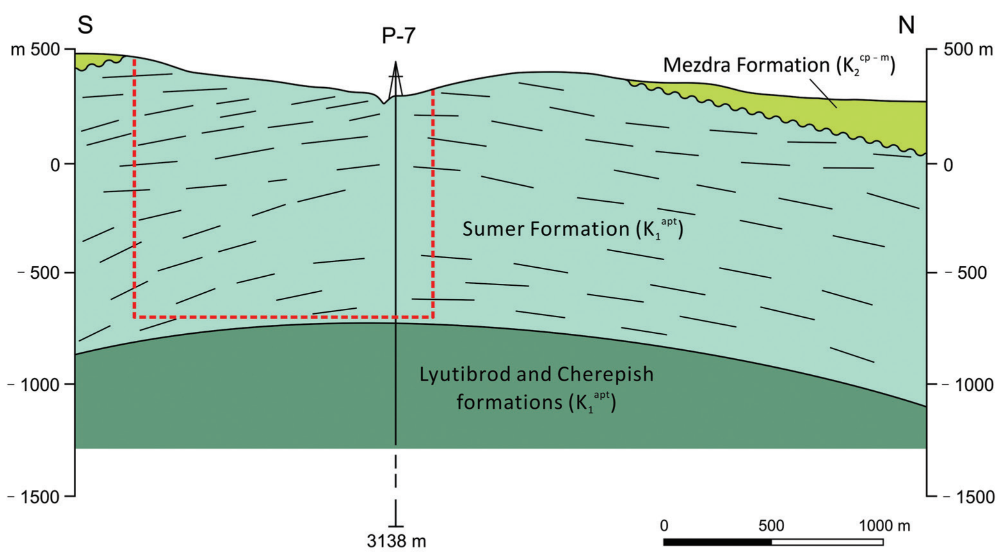

Fig. 1. Potential zone (red dotted line) for optional geological repository of radioactive waste in the Sumer Formation.

concentrations are obtained for the three radionuclides (Table 1).

Formulation of the hydraulic (water flow) model

The hydraulic model of the Sumer Formation at the specific site was already a subject to previous investigation (Tsvetkova *et al.*, 2019). In brief, the simulations showed that, at this low porosity (6–8%) and large thickness of the marl massif (about 1200 m), the main parameter that controls the water transport is the permeability of the porous medium. The water flow, after establishing a "quasi-equilibrium state", takes place at a speed of 1.73×10^{-6} m/d (2.0×10^{-11} m/s). This equilibrium state occurs after 940 years from the beginning of the simulations for the whole simulation domain of 400 m.

Formulation of the mass transport model

Based on the geological settings of the investigated site, as well as the position of the storage facility, one-dimensional convective-dispersive model of the water flow describing the transport of mutually independent solutes being subject to decay processes has been used. In this model, the retardation properties of the engineered layers and argillaceous sediments are described by the so-called distribution coefficient K_d. Such a model has been applied in similar studies, both in Bulgaria (Stoyanov, 2012, 2019; Kotsev *et al.*, 2018) and elsewhere (*e.g.*, Robinson and Bussod, 2000; Mallants *et al.*, 2001, 2011; Šimůnek *et al.*, 2006; Merk, 2012).

Elements of the Mass Transport Field (Model)

In order to perform a complete assessment of the radionuclide migration in the argillaceous sediments of the Sumer Formation, the following elements were considered in one-dimension vertical model with particular characteristics: from 0 m to 335 m below the surface – argillaceous sediments (Sumer Formation); from 335 m to 350 m – repository with engineered barrier; and from 350 m to 400 m – argillaceous sediments. In addition, mass transport parameters of the selected three key radionuclides were also taken into account (see Table 2). The constructed model was implemented in computer code HYDRUS-1D (Šimůnek *et al.*, 2008) numerically solving the partial differential equations for the convective–dispersive equations.

RESULTS

In order to illustrate the results obtained, in the model study for all three radionuclides, "observation nodes" were selected at three specific points in the simulation profile: *1*) exactly at the bottom of

Table 1
Sample radionuclide inventory and concentrations after 100 years from the disposal of HLW

Radionuclide	Activity after 100 years after disposal for MTHM	Total weight of wastes	Total activity	Concentration in the solute phase
	Bq	MTHM	Bq	Bq/m^3
^{137}Cs	3.8110×10^{14}	803664	3.0626×10^{17}	2.4908×10^{12}
^{90}Sr	2.4827×10^{14}	803664	1.9953×10^{17}	1.6226×10^{12}
^{241}Am	1.3875×10^{14}	803664	1.1151×10^{17}	9.0683×10^{11}

Table 2
Basic parameters characterizing the behaviour and transport of the considered radionuclides

Radionuclide	Decay constant, λ	Distribution coefficient K_d for the bentonite barrier*	Distribution coefficient K_d for marls**
	1/d	m^3/kg	m^3/kg
^{137}Cs	6.29×10^{-5}	0.33	0.15
^{90}Sr	6.55×10^{-5}	0.39	0.16
^{241}Am	4.39×10^{-5}	30.00	8.00

*The values given are for clays from Thibault *et al.* (1990).
**The values for the distribution coefficient are determined on the basis of specialized literature data (EPA, 1999, 2004) and are consistent with the mineral and chemical composition of the marls.

Table 3
Maximum C_{max} and minimum C_{min} concentrations in the solute phase of the investigated radionuclides for the whole period of simulation at the three observation nodes' depths

Observation node (distance from the surface)	Radionuclide		
	^{137}Cs	^{90}Sr	^{241}Am
	$C_{max} - C_{min}$ [Bq/m^3]	$C_{max} - C_{min}$ [Bq/m^3]	$C_{max} - C_{min}$ [Bq/m^3]
1 (350 m)	$2.49\times10^{12} - 0$	$1.62\times10^{12} - 0$	$9.07\times10^{11} - 0$
2 (351 m)	$6.12\times10^{8} - 0$	$4.55\times10^{8} - 0$	$2.40\times10^{8} - 0$
3 (355 m)	0 – 0	0 – 0	0 – 0

the storage facility; *2*) one metre below the storage facility (in the marl domain); and *3*) five metres below the facility with the presumption that the main pathway of radionuclide transport is downwards. So as to compare the results, the maximum and minimum concentrations (C_{max} and C_{min}) of the three radionuclides at the three points were also represented (Table 3). The simulations show that, at the bottom of the storage facility, the maximum concentrations, coinciding with the initial concentrations, decrease to "zero" for the three radionuclides for the period of investigation, as the period of simulation is 10,000 years for ^{137}Cs and ^{90}Sr; and 100,000 for ^{241}Am, respectively. The time of zero concentration for Cs, Sr and Am, respectively, is after 1,980 years, 1,854 years and 28,049 years. It should be noted that, for this particular study, the "zero concentration" was assumed as less than 1 Bq/m^3.

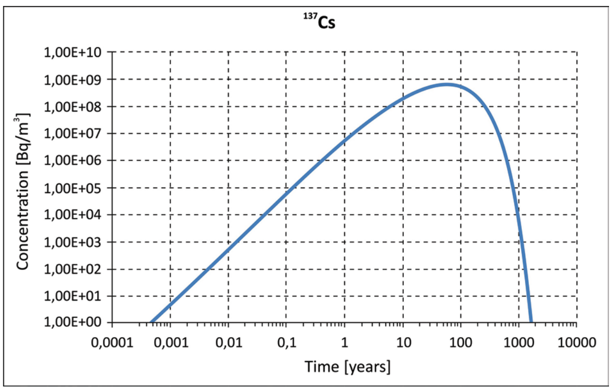

Fig. 2. Cesium concentration in solute phase vs time at 1 m below the bottom of the storage facility.

At one metre below the facility, the peak values of concentrations are between three and four times of magnitude lower than the initial ones (Table 3). For Cs, the peak value of 6.12×10^{8} Bq/m^3 (almost four orders of magnitude lower) was obtained after 66 years and "zero concentration" was obtained after 1,644 years (Fig. 2). For Sr, the peak value of 4.55×10^{8} Bq/m^3 (about three and a half orders of magnitude lower) was obtained after 60 years and "zero concentration" was obtained after 1,542 years (Fig. 3). For Am, the peak value of 2.40×10^{8} Bq/m^3 (almost four orders of magnitude lower) was obtained after 929 years and "zero concentration" was obtained after 23,069 years (Fig. 4).

At 5 m below the storage facility, the concentrations are "zero" during the whole period of simula-

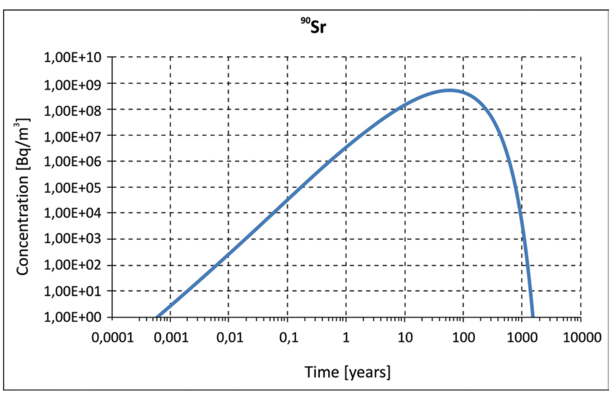

Fig. 3. Strontium concentration in solute phase vs time at 1 m below the bottom of the storage facility.

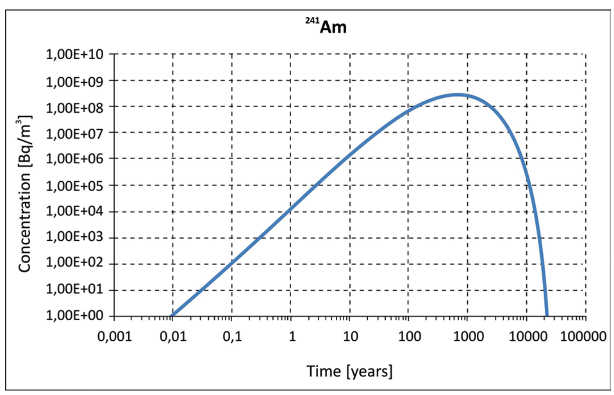

Fig. 4. Americium concentration in solute phase vs time at 1 m below the bottom of the storage facility.

tions, *i.e.*, 10,000 years for Cs and Sr and 100,000 years for Am (Table 3). In addition, the relative errors in the solute mass balance of the entire flow domain for the three simulations are very reasonable: 2.98% for ^{137}Cs; 3.09% for ^{90}Sr; and 2.89% for ^{241}Am. Hence, the performed mass transport model is mathematically verified. Therefore, marls are a promising domain as a deep geological disposal medium, but stating that this study is a preliminary attempt to characterize the Sumer Formation. It should note that the results for the concentration of radionuclides are based on officially published data and simulations of numerical modeling.

CONCLUSION

The comparison of the transport of three main radionuclides at 1 m below the repository shows that they will decrease their concentration activity as subject to the decay processes and retardation properties of the storage facility and marl medium with about three orders of magnitude. In addition, at 5 m under the storage facility, the concentrations are zero for the whole period of simulations. Thus, an eventual repository for HLW hosted in the argillaceous sediments of the Sumer Formation will function for thousands of years in a safe manner from the viewpoint of the investigated radionuclides (or others with similar parameters).

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