SITE CHARACTERIZATION FOR LIL RADIOACTIVE WASTE DISPOSAL IN ROMANIA (LA-UR-01-4599)

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ABSTRACT

Recent studies in radioactive waste management in Romania have focussed mainly on the disposal of low and intermediate level waste from the operation of the new nuclear power plant at Cernavoda. Following extensive geological, hydrological, seismological, physical and chemical investigations, a disposal site at Saligny has been selected. This paper presents description of the site at Saligny as well as the most important results of the site characterisation. These are reflected in the three-dimensional, stratigraphical representation of the loess and clay layers and in representative parameter values for the main layers. Based on these data, the simulation of the background, unsaturated-zone water flow at the Saligny site, calculated by the FEHM code, is in a good agreement with the measured moisture profile.

INTRODUCTION

Following the start-up of the first CANDU unit at Cernavoda nuclear power plant (NPP), the management of the operational radioactive waste generated by the plant became an important consideration of the Romanian nuclear sector. The radioactive waste management programme has initially focused on the operational low and intermediate level waste (LILW) produced by the NPP. The main avenues followed were: the identifying of potential disposal sites and the subsequent selection of the optimum site, detailed characterisation and validation according to national and international basic principles for protection of humans and the environment.

The currently accepted management of LILW involves conditioning of the waste in steel containers or cans with a biological protection provided by concrete, and then disposing of the waste in a near-surface repository.

Concerning site selection, the preliminary investigations covered Dobrogea, the region of Romania situated in the southeastern part of the country. Seismic, lithologic and hydrologic criteria applied to this area established that 37 sites had the required geological characteristics to be potential hosts for a LILW repository [1]. Supplementary geophysical, geochemical and geo-technical investigations reduced this number and finally only two candidate sites, Cernavoda and Saligny, were selected.

For the Cernavoda site, detailed investigations performed in 1992-1996 [2] pointed to the existence of a red clay continuous layer, having a substantial thickness (13-15m) and good qualities in radionuclide retention given by its homogeneity and a high clay fraction [2]. Even though preliminary studies showed that at the nearby Saligny site this clay layer is not continuous and has a lower thickness (0.5-10 m), with the clay containing a high fraction of fine sand and limestone, the shorter distance to the NPP and other administrative advantages mean that this site is the more attractive. For this reason, the Saligny site has, since 1997, undergone a complete characterisation by physical, chemical, hydrological and radionuclide migration investigations, as required for the preliminary safety report.

The main features of Dobrogea, which make them unique in Europe in this respect, are the presence of the loess and the low depth of the water table. The loess is a soil with, in its natural state, a large water sensitivity and weak foundation properties. Moreover, its large porosity and high permeability do not indicate that, at first sight, it would make a good host for waste disposal. However, there are special methods [3] for consolidating this soil that can not only improve the mechanical properties but also decrease the permeability.

The water table is some 40-45 m from the top surface though the main aquifer of this zone is found at 80m. The aquifer is connected directly to the Danube River and the Danube - Black Sea canal, which border the site on the west and south sides respectively. The reason that this site was still selected as a possible host for a disposal site, even with these unfavourable characteristics, is that the loess was deposited on a clay formation containing a quaternary red clay layer and a pre-quaternary clay layer. The water table is between these two clay horizons while the aquifer is under the second clay layer in the Baremmian limestone platform.

Another argument supporting the selection of this site is the arid climate of the area. The small rainfall rate, half that in the rest of the country, is reflected in a low infiltration rate and a low degree of saturation in the loess formation. These natural conditions result in slow water percolation rates through the unsaturated zone.

In order to have a complete and a realistic image of the Saligny site and accurate input data for the safety assessment codes, a borehole network was drawn up and a methodology for site characterisation was established. Thus, in addition to the lithological information, data was gathered on the mineralogy of the geological formations, the physical, chemical and hydrologic properties, the soil-water retention capacity and the radionuclide migration characteristics [4].

These experimental data have been collected in a database, allowing a statistical calculation to be performed in order to obtain the most representative values for the site characteristics, which were then used as actual input data for the disposal safety assessment programme. To restrict the extent of the site characterisation and safety assessment programme, the experimental investigations were correlated with the results obtained by the modelling of underground water flow in the unsaturated zone performed using two different computer programs: SWMS_2D [5] and, more recently, FEHM [6].

EXPERIMENTAL WORK

The drilling network at the Saligny site currently consists of 24 boreholes, with only 16 of these used to determine the site stratigraphy and eight of these to perform a detailed characterisation. The maximum well depth was down to the Baremmian bed (more than 80 m from the site surface). The drill cores were covered with a paraffin layer in order to preserve the in situ moisture content. Undisturbed samples were taken from the drill cores for physical, mineralogical and hydrological analysis.

The methodology used in the site characterisation brought together a variety of recognised methods for determining the main parameters involved in water and contaminant transport in geological formations. The parameters studied included such aspects as the structural properties of soil (density, porosity, permeability), the soil-water retention capacity, the mineralogical composition and the moisture content.

The density and porosity were established by standard methods on undisturbed samples. For each geological layer, the representative values of structural characteristics were calculated as arithmetical means of the experimental data. In addition, since the underground water moves through the open porosity and a knowledge of porous structure helps in a better understanding of the hydrological properties of the geological formations, the porosity distribution was also determined. Dried samples of loess and clay were analysed by the mercury penetration method using a CARLO ERBA porosimeter which covers a pore radius range between 0.002 and 100 μm.

The saturated hydraulic conductivity *(K)* was measured on undisturbed samples using a permeameter with constant level and suction. For each drillilng, an equivalent hydraulic conductivity was established as a weighted average over each hydrologic layer. To be conservative, the representative value of the hydraulic conductivity was calculated as the minimum value of the equivalent hydraulic conductivities over each geological layer.

The soil water retention curves were determined at 15 pressure levels using two complementary devices: with pressure plate and with pressure membrane, which cover the head pressure range 0.1-15 kg/cm^2. Based on the moisture, θ, and pressure head values, the van Genuchten parameters, representing the residual and saturated water content (θ_r and θ_s) as well as the empirical shape parameter curves (α and n) have been calculated by solving the inverse problem. This was formulated as a classical least square problem in which the parameters are estimated by minimising the discrepancy between observed and computed θ values [7].

The loess and clay mineralogy, performed by X ray diffraction and IR spectroscopy, provides the percentage of quartz, feldspars, mica, carbonates and clayey minerals. These results were especially used in refining the site stratigraphy.

Accurate modeling of groundwater flow and transport at the Saligny disposal site requires the integration of geologic model information with computational grids. To achieve such integration a three-dimensional representation of the geology must first be created that adequately represents the area of interest. Geologic contact information was collected for seven geologic units from 15 boreholes drilled and cored near the proposed Saligny disposal site. Detailed records are kept for the elevations of tops and bottoms of geologic units logged from the boreholes. Two-dimensional surfaces are created for all seven geologic units, by taking all the elevation data for the top of a particular unit and using an inverse-distance-weighting algorithm to create an interpolated surface. Once the geologic surfaces are created they are logically ordered based on their natural deposition and assembled using STRATAMODEL™ to construct a three-dimensional model representation of geology.

The process to create computational grids for the Saligny disposal site begins with the selection of the model domain. For the three-dimensional computational grid, the model domain (1 Km^2) is defined as the extent of the disposal site with a buffer zone. The buffer is used to establish appropriate boundary conditions for the groundwater flow and transport models. Once the domain is chosen, a node distribution needs to be defined that will ensure adequate resolution to capture the areas of interest. The LaGriT [8] grid generation tool is used to create a horizontal (x,y) cell resolution of 20 meters and a vertical cell resolution of 1 meter for the three-dimensional model. The geologic surfaces are then used to define which nodes fall within each of the seven different geologic regions, and these nodal definitions are provided as input to FEHM. Extracting a vertical cross-section from the three-dimensional computational grid creates the two-dimensional computational grid. The two-dimensional grid extent runs north south through the FS2 borehole location and has the same vertical resolution as the three-dimensional grid.

The unsaturated-zone flow simulations were run with FEHM, a two- or three-dimensional finite-element/finite-volume code suitable for simulating systems with complex geometries that arise when modeling subsurface flow and transport [6]. In the unsaturated zone, the governing equations for flow are based on the principles of conservation of water and air. Darcy's law is assumed to be valid for the momentum of the air and water phases in the unsaturated zone and for the water phase in the saturated zone. The convection-dispersion equation governs solute transport [6] in these analyses.

RESULTS

Dobrogea is a region covered by a large loess formation deposited in the quaternary era. The lithological observations indicated that it could be divided into two different layers: silty loess and clayey loess. The loess layers were deposited on a red quaternary clay horizon, which was formed in the Pleistocene on a pre-quaternary clay formation. The loess and clay layers lay on the Baremmian limestone platform.

From the lithological point of view, the loess layers are continuous and almost homogenous. The silty loess thickness varies between 15 and 20m, while the clayey loess is around 20m in depth. On the other hand, the red clay layer is not continuous along the entire site and has large variations in width, with the maximum thickness being some 11m. Furthermore, red clay is mixed with very different fractions of sand, whose granulation shows large variations along the horizon. Thus, pure red clay zones alternate with fine red sand in both the horizontal and vertical directions.

However the most heterogeneous geological layer is the pre-quaternary clay, which contains many kinds of clays, lenses of sand and even gravel, as well as limestone and sandstone. This complex conglomerate is continuous and has a thickness of about 40m.

The Baremmian platform consists of different qualities of limestone, more or less fractured.

The first three layers, the silty loess, clayey loess, and red clay, belong to the unsaturated zone. The degree of saturation increases from 0.1 in the first loess horizon to 0.8 in the red clay. The pre-quaternary clay layer, where the degree of saturation varies between 0.8 and 1 could be considered as the first saturated zone since many sand lenses are filled with water. The second saturated zone corresponds to the Baremmian limestone where the main aquifer of the region is found. It is directly connected to the Danube River and to the Danube - Black Sea canal, which bound the site along the west and south sides, respectively. The level of the Danube controls the aquifer flow direction. Therefore, during wet periods when the level of the Danube is higher, the aquifer moves in an eastern direction, while in dry seasons when the level of the river is lower, there is movement in the opposite direction [1]. Since the canal is maintained at a constant level, the amplitude of these “come and go” movements depends only on the variation in the level of the Danube.

The lithological characteristics of the Saligny site had to be refined as a result of statistical analysis of the experimental results obtained from the analysis of the samples from drill cores, especially from the loess formations. A more refined structure was also demonstrated by the results obtained from the modelling of underground water flow in the unsaturated zone. In accordance with the previous lithological observations, the profile of the computed degree of saturation shows a continuously increasing moisture content while the measured moisture profile, presented in Fig.1, shows the existence of two peaks of water content at the side of the loess horizons.

Therefore the statistical analyse of dried densities and porosity values, the main parameters defining the layer structure, showed a two-modal distribution in silty loess layer and three-modal distribution in clayey loess layer. The points grouped around the lower value of porosity correspond to the loess samples having larger water content. As a structure with small porosity reflects the presence of larger clay content, proved by experimental data and illustrated in Fig. 1, the conclusion was that in side of the loess formations there are two thin and more consolidated layers, with better properties in water retention. With this hypothesis, the statistical analyse of data showed, for each new layer, a normal distribution of experimental values and especially a significant decrease of standard deviation i.e. from 5% in the initial silty loess to 2% in the new considered layer.

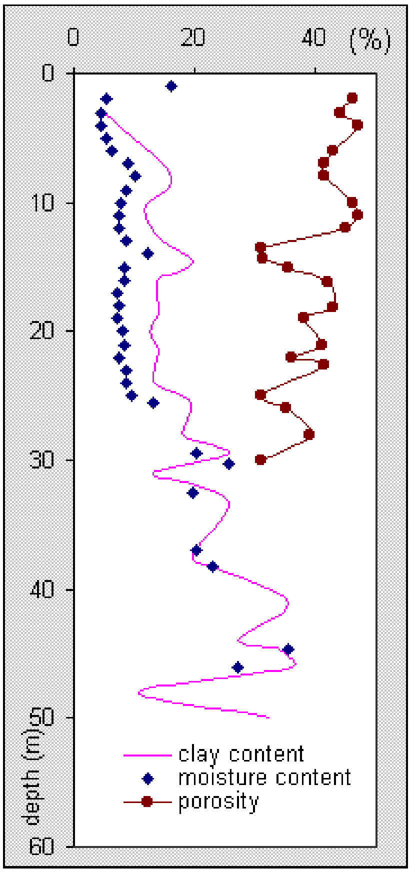

Figure 1. Correlation of the loess structure and composition with the moisture content

The presence of these consolidated loess was also confirmed by porosity distribution diagrams and soil-water retention curves, characteristics directly linked to soil-water interaction.

Even if the diagrams of open porosity distribution in the samples of loess and clay collected from Saligny site showed a large dispersion in porous structure, few characteristics of the porosity distribution in these geological layers could be determined. The large percentage of quartz, mica and feldspars in the loess layers determines the large porosity (40-47%) of these horizons. The pores can generally be grouped into 4 classes defined by most probable radii of 60μm, 9 μm, 0.8μm and 0.3μm. The clay fraction, with particle size less than 2 μm, is responsible for the fine pores smaller than 0,2μm. They are usually grouped around 0.07μm, 0.01μm and 0.003μm. The porosity distribution, quantitatively reflected in the main classes of pores, the mean pore radius (r_P) and the fraction of each pore class in the total porosity (f) is synthesised in Table I for all horizons. Specific pore volume (V_p), total (n) and open porosity (p) complete the characteristic of porous structure of the geological layers.

More than 60% of the open pore volume of the silty loess layer (loess A) is due to the pores ranging between 7 and 15μm, while the fine porosity represents less than 8%. The same groups of pores also determine the structure of the clayey loess, but the larger percentage of clay leads to slight increases in the percentage of fine pores. The porosity distribution in loess A1 and loess B1 is significantly shifted towards the smaller radius values, 0.07μm, confirming the more consolidated structure of these loess layers. The porosity distribution in samples from the red clay layer showed a very large sensitivity to the sand content and to its particle size. The pure red clay has a very fine porous structure, with the pore radius generally ranging between 0.002-0.4 μm, but the presence of the sand brings in classes of pores with mean radii of 0.8 and 9μm.

Table I. Porosity distribution of the geological layers from Saligny site

Horizon	n (%)	P (%)	V_p (cm^3/g)	main pores range (μm)	f (%)	r_p (μm)
Loess A	40÷47	17÷30	0.20÷0.29	7÷100	60÷83	9 and 60
				0.005÷0.5	4÷8	0.8
Loess A1	41÷43	18÷22	0.16÷0.20	0.5÷2	45÷65	0.8
				0.005÷0.5	15÷20	0.07
Loess B	39÷45	13÷23	0.11÷0.19	0.5÷15	26÷52	0.8 and 9
				0.01÷0.5	16÷36	0.3
Loess B1	36÷43	12÷22	0.10÷0.19	0.5÷5	35÷55	1
				0.005÷0.5	15÷45	0.07
Loess B2	39÷45	13÷23	0.17÷0.21	0.5÷15	30÷78	9
				0.01÷0.5	15÷50	0.3
Clay 1	30÷45	10÷23	0.08÷0.19	0.4÷20	50÷77	0.9 and 9
				0.04÷0.02	16÷42	0.01
				0.002÷0.004	20÷56	0.003

The differences in fine porosity fraction are very well reflected in their hydraulic properties. Soil-water retention capacity illustrated in moisture-pressure head curves, w=f(pF), improves as the percentage of fine porosity increases. Figure 2 presents the most representative retention curves for all layers from unsaturated zone. The very similar retention curves for both consolidated loess layers (loess A1 and loess B1) show indeed a better capacity in water retention than the other loess layers. Also, the clayey loess characteristics with regards to water interaction are almost the same for the upper (Loess B) and lower (Loess B2) layers. The same van Ghenuchten parameters express the soil-water retention characteristic, a fact anticipated by the very close values of other parameters as density, porosity or hydraulic conductivity.

From these observations the final stratigraphy of the Saligny site, consisting of five distinct loess layers, two clay layers and the Barremian platform, could be defined and represented in a three-dimensional diagram, Figure 3, using the STRATAMODEL ™ program.

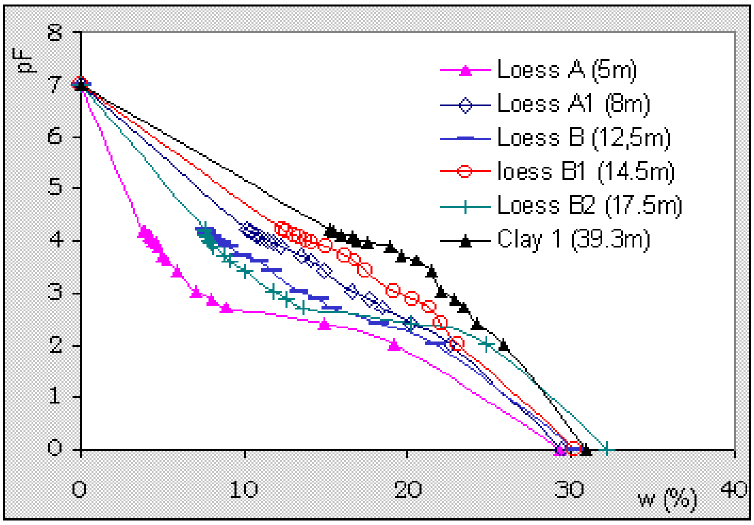

Figure 2. The suction-moisture curves for the geological formations of Saligny site

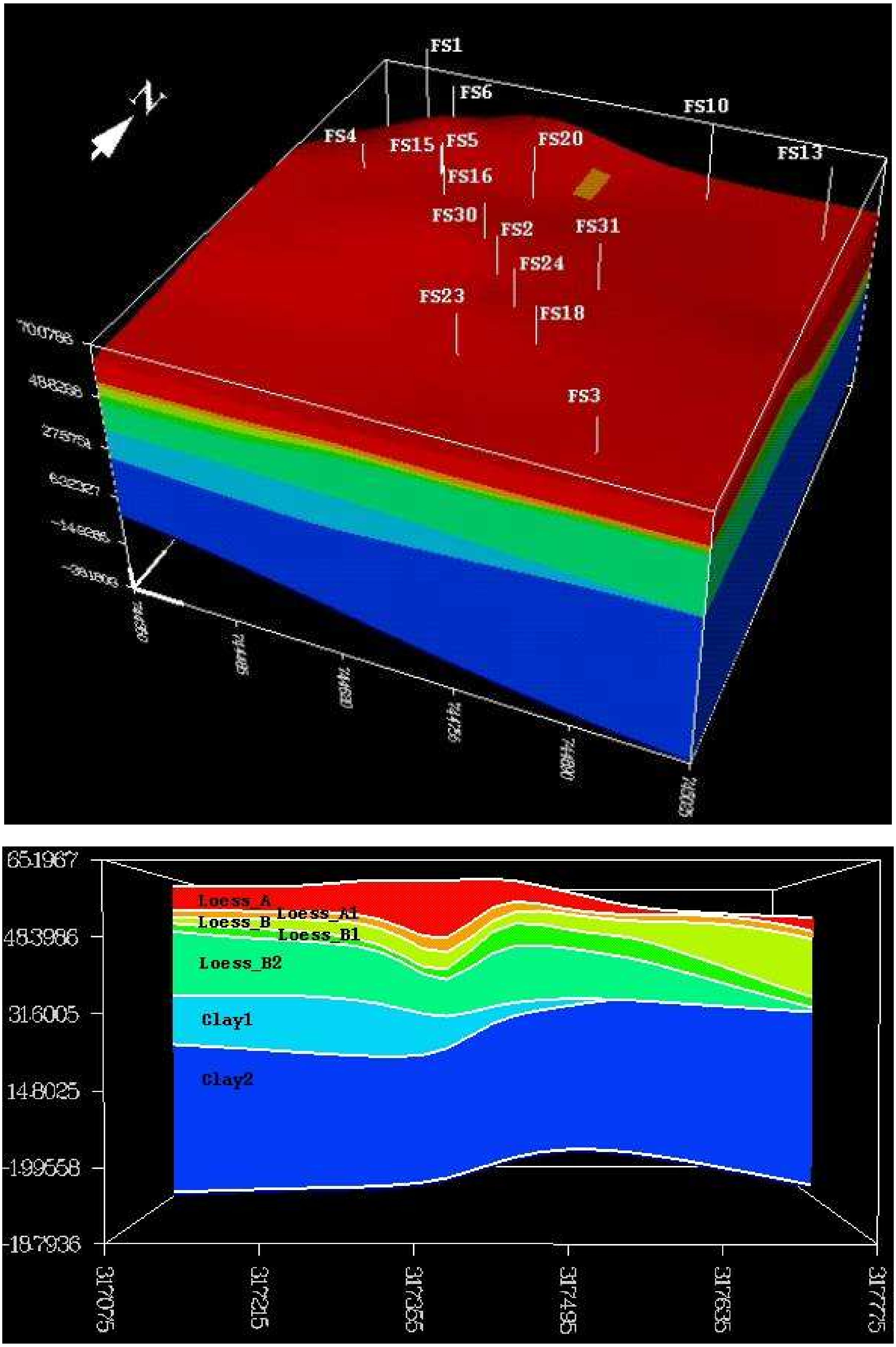

Figure 3. Saligny site stratigraphy (up: three-dimensional representation, down: geological layer variation along N-S direction)

The vertical cross-section along the N-S direction illustrates the geological layer variation at this site and clearly point out the large width variation of the red clay horizon.

The geologic model consists of seven distinct geologic units, presented in Table II. To ensure an accurate reflection of the real world, the geologic model is the basis for defining the geometry of the geologic layers that form the computational grids.

A two-dimensional finite-element grid was generated from the three-dimensional geologic model shown in Figure 3. Steady flow simulations were run with FEHM using the representative values of the main hydrologic characteristics involved in unsatured-zone water transport, as shown in Table II.

Table II. Representative values of the main structural and hydrologic properties of the geological layers from Saligny site

Layer	Geological characteristics	Density (g/cm^3)	Porosity (%)	K_s (cm/s)	Van Genuchten parameters		
					θ_r	α	n
Loess A	Silty loess	1.47	45.01	$8.1.10^{-6}$	0.022	0.173	2.50
Loess A1	Consolidated silty loess	1.53	42.34	$5.7\cdot10^{-5}$	0.005	0.273	1.65
Loess B	Clayey loess	1.54	42.18	$2.1\cdot10^{-5}$	0.005	0.173	2.10
Loess B1	Consolidated clayey loess	1.60	40.05	$1.1\cdot10^{-5}$	0.005	0.273	1.50
Loess B2	Clayey loess	1.55	41.94	$2.1\cdot10^{-5}$	0.005	0.173	2.10
Clay 1	Quaternary red clay	1.58	42.60	$3.2\cdot10^{-7}$	0.019	0.236	1.118
Clay 2	Pre-quaternary clay, with sand and limestone lenses	1.68	38.08	$8.1\cdot10^{-7}$	0.019	0.236	1.118

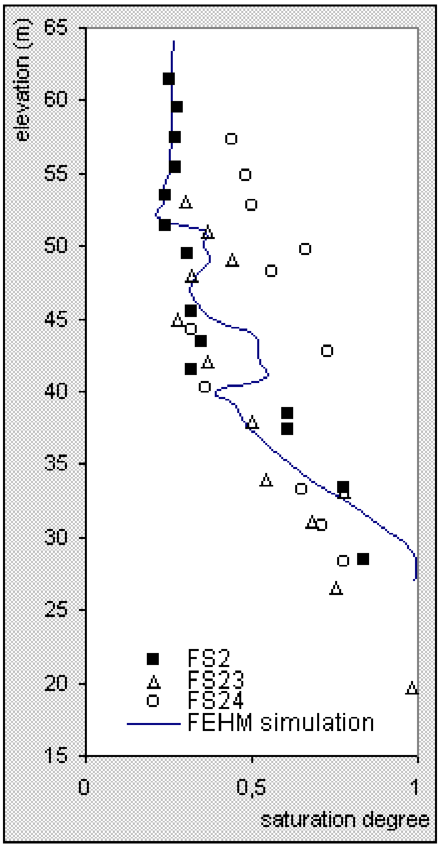

Figure 4. Comparison of FEHM simulation of moisture profile and the experimental data

Figure 4 shows a comparison between the saturation computed with FEHM, using a steady recharge rate of 18.75 mm/yr, and field saturation measured in three boreholes at the site. The computed saturation profile agrees fairly well with data from wells FS2 and FS23. However, the computed saturation profile is considerably lower than saturation data from well FS24, although the shape of the two profiles is consistent. Since well FS24 was drilled following a winter with heavy snowfall, simulations including higher recharge rates or possibly transient recharge rates may be investigated in the future.

The level of confidence in the simulation results can be improved using a measured infiltration rate, since the value used was the result given by solving the inverse problem [9].

Future activities in the site characterisation programme will include further field experiments in order to provide the data needed to complete the description of the site. These experiments will concentrate on the aquifer properties and erosion aspects, the latter being a significant process in this area.

CONCLUSIONS

The low and intermediate level active waste generated by Cernavoda NPP will be stored in a near-surface disposal in the Dobrogea region.

As a result of the research conducted to date on site selection and complete site characterisation, the hydrogeological, physical and mineralogical properties of unsaturated zone are now well defined.

The final site stratigraphy was established as a result of the successive iterations between experimental and modelling activities.

The background water flow in the unsaturated zone of the Saligny site simulated using the FEHM code, with representative values determined statistically, is in very good agreement with the measured moisture profile.

The database containing all parameters determined by laboratory experiments gives the correct input data for the unsaturated zone. Future research in site characterisation will be focused on field experiments in order to determine infiltration rate, aquifer characteristics and soil erosion.

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