RADWASTE DISPOSAL: MEDIUM PERMEABILITY SHALE AS A GEOLOGICAL BARRIER

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ABSTRACT

One of the major criteria of site selection for low radwaste disposal is hydrogeological. A common practice is to locate sites on a geological barrier, whose minimum thickness depends on national regulations. However, it is generally considered that the minimum permeability of the geological barrier must be less than 10^{-9} m/s with a thickness of the impervious material of at least 20 m.

In general, shale has a permeability approaching 10^{-7} m/s. Therefore, by strict application of the above permeability criteria, they would be unsuitable for use as a natural barrier for a waste repository.

Any contaminant movement can be avoided if between an underlying aquifer and the bottom of the waste disposal an ascending water flow exists. Since it is hard to prove a continuous and perennial ascending water flow below the radwaste repository, one could imagine creating an artificially controlled ascending water flow. This is the case where a deeper shale formation is overlain by an open-jointed surface shale layer containing a shallow aquifer. Their use as a natural barrier to a radwaste repository can be accepted by drawing down the water table. This has the effect of creating an upward flow below the repository.

Using mathematical modeling, different conditions were analyzed such as the drawdown of the phreatic aquifer, water level differences between shallow and underlying aquifers, repository width, permeability of the shale mass and the existence of thin sandstone beds inside the shale body. Modeling showed that for this particular layout migration of pollutants can only occur by diffusion. Contaminant migration time can be evaluated and compared with migration time calculated from analytical equations and the migration time through a thick clay layer.

A promising site has been field investigated. Results confirm the suitability of the shale as a geological barrier.

INTRODUCTION

In order to be able to establish a convincing model of the water flow under a shallow subsurface or surface low-level radioactive waste repository, a set of important hydrogeological criteria must be fulfilled.

One of those criteria, addressing the risk mitigation of the radionuclides migration, is the presence of a water conductivity contrast between a permeable formation on which the repository is built and a lower much tighter layer considered as a geological barrier. The minimum thickness of this impervious unit depends upon the lithogical characteristics of the rock. A thickness of at least 20 m is considered as sufficient if the permeability of the rock is less than 10^{-9} m/s.

In Belgium, only two tertiary argillaceous layers, the Boom Clay Formation and the Yperian Clays, which are in the western and northern parts of the country and are close enough to the ground surface and covered by sandy sediments, can be taken into consideration as geological barriers. Strict compliance with the above criterion, applied to the Palaeozoic shale formations outcropping at the eastern and southern parts of the country and having a permeability approaching 10^{-7} m/s, would lead to the conclusion that such a type of rock is unsuitable for use as a natural barrier. Nevertheless, mathematical modelling allows us to demonstrate that, under certain favourable conditions, even shale formations can be considered as an alternative and effective barrier against migration of radionuclides, providing generally weathered and fissured surface shale covers a deeper, non-fissured or slightly fissured and more impervious shale formation.

In the first part of this paper, we will describe the theoretical and generic approach allowing the evaluation of the hydrogeological conditions which must be fulfilled to consider shale formations as potentially favourable to host a surface repository of low level and short-lived radioactive waste. In the second part of this paper, we will provide the results of field reconnaissance on a specific site where Devonian shale formations are outcropping in southern Belgium and describe to what extend the site complies with the theoretical conditions.

THEORETICAL AND GENERIC APPROACH

Necessity of an Ascending Hydraulic Gradient

The dispersion of the radionuclides outside the immediate vicinity of the repository can be avoided, if an ascending hydraulic gradient exists between the underlying aquifer on which the repository is built and the bottom of the repository foundation. This ascending flow can then be collected and controlled by a drainage system established under or in the repository foundation. Since a natural, continuous and perennial ascending water flow below the repository can never be guaranteed, the artificial creation of such a water flow can be envisaged.

Mathematical modelling demonstrates that drawing down the water table of the phreatic aquifer contained in the upper weathered and fissured shale will create an upward advective flow below the repository. Diverse hydrological conditions and geological settings have been analysed, such as repository width, drawdown of the phreatic water table, piezometric heads difference between phreatic and underlying water tables, and hydraulic conductivity variations of the shale due, for instance, to the existence of thin sandstone beds inside the shale body. Moreover, mathematical modelling shows that, for particular configurations, radionuclides migration can only occur by diffusion. Migration time obtained from this mathematical modelling then can be compared with that calculated analytically for shale formations (in the present case, radionuclides transport by molecular diffusion), as well as with the migration time calculated analytically for a thick clay layer.

Advective and Dispersive Transports

The transport of dissolved solids at the same velocity as groundwater is called advective transport. Movements of contaminants in groundwater also occur by dispersive transport. Dispersion is the mechanism of mixing and spreading caused by variations in velocity within porous or fissured media. Molecular diffusion is the mechanism of mixing and spreading the contaminants within the considered medium.

To distinguish whether diffusion is prevailing on advection, the number of Peclet P_e has been used. This non-dimensional number is defined as (de Marsily 1981; [1]) :

$$P_e = \frac{U \cdot \sqrt{h}}{\omega_c \cdot d_O}$$

where :

- U = Darcy velocity, $[LT^{-1}]$
- h = specific permeability, $[L^2]$
- ω_c = cinematic porosity, [-],
- d_o = molecular diffusion coefficient , $[L^2T^{-1}]$.

Contaminant transport is :

- only controlled by diffusion if $P_e < 2$,
- controlled by a combination of diffusion and dispersion if $2 \leq P_e < 9$,
- related to dispersion only if $P_e > 9$.

Modelling

Principle

The code used in the mathematical modelling was the AQUA3D groundwater and contaminant transport model [2], developed by Vatnaskil Consulting Engineers (Iceland). This model was developed to solve three dimensional groundwater flows and transport equations using the Galerkin finite element method.
This modelling aims at defining the transport regime in the phreatic aquifer underneath the repository. The drainage system of the repository located at the bottom of its foundation collects the water of the phreatic aquifer.

The groundwater flow in the shale (3D model) was modelled taking into account the presence of the repository. The model enables the flow velocities below the repository to be established, from which the Peclet number is calculated. This number gives an indication of the transport mechanisms underneath the repository.

Model Description

This model simulates an open-jointed shale formation overlying a slightly fissured shale formation. The overall structure of the subsurface consists mainly of, at least, 200 m thickness of slightly fissured shale ($k = 10^{-8}$ m/s), covered by a 17 m thick layer of open-jointed, fissured shale ($k = 10^{-6}$ m/s). The repository itself is 65 m wide, and its foundation is situated 5 m below the water level of the upper aquifer (Figure 1).

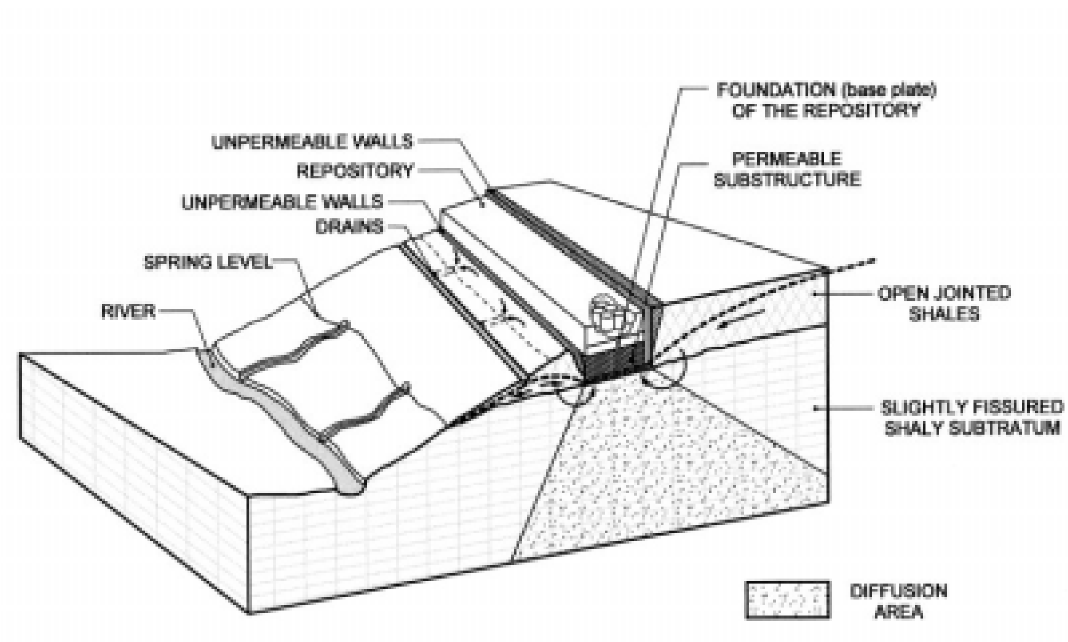

Figure 1 : Cross section through the repository and underlying shale formations

The boundary conditions were defined as follows :

- vertical boundaries are no flow boundaries,
- top boundary is a fixed value boundary (these fixed values correspond to the piezometric heads, which have been deduced from the horizontal model),
- bottom boundary is a fixed value boundary (the imposed values correspond to the piezometric head of the confined aquifer underlying the slightly fissured shale; this piezometric head is always located within the slightly fissured shale units).

The drainage at the base of the deposit has been integrated into the model by setting a piezometric head (fixed value) which is lower than the piezometric level calculated by the 3D-model.

Results

This model has been used to simulate several possible configurations, established by modifying one of the following parameters:

- the depth of the drainage system under the repository,
- the piezometric head of the underlying confined aquifer,

- the permeability k of the shales,
- the width of the deposit,
- the thickness of the slightly fissured shale unit,
- the existence of permeability heterogeneities (sandstone beds) within the shale.

Within these configurations, the most common mode of transport directly beneath the waste repository was diffusion; whereas, at the borders of the repository, diffusion is combined with ascending convection (fig.2). This means that transport by descending convection, which is unfavourable for safety reasons, did not appear as the main transport mode. Moreover, when diffusion occurred underneath the waste repository, it was present in the zone that extends up to the base of the slightly fissured shale which improves the safety of the system.

Modification of the parameters mentioned above did not influence the resulting transport mode to any great extent. In general, only the proportion of the diffusion area relative to the diffusion-convection area changed.

The only exception encountered was in the case of the presence of a bed with high permeability (sandstone beds) within the slightly fissured shale unit, where increasing the permeability led to the appearance of zones characterised by descending convection.

In fact, the presence of a more permeable layer, within the slightly fissured shales, caused the replacement of diffusion zones by descending convection zones. This situation no longer guaranteed the safety of the repository.

Shale specific condition - perennial aquifer

In the long term, the safety of waste disposal on shale, based on the presence of an upward flow beneath the repository, requires that the phreatic aquifer in the upper fissured shale is perennial. This condition has been evaluated by means of statistics.

The basic assumption is that the ascendant groundwater flow will persist through time as long as the groundwater budget (storage) is positive. The groundwater storage at the end of a hydrologic year has been established using the groundwater balance. In addition, the probability of having at least one complete drying out of the phreatic aquifer in a period of 300 years has been calculated using the following equation,

$$P_2 = 1 - \left(1 - P_1\right)^{\frac{N}{n}}$$

where.:

- P_1 = probability of a drying out within 1 hydrological year,
- P_2 = probability of a drying out within N years,
- N = 300,
- n = number of drying out (reserve = 0) within N years = 1.

These probability calculations are based on the assumptions that all variables *are normal distributed* and that the effective drainage porosity (ω) and aquifer thickness (*e*) are constant over the whole catchment area, where the groundwater reserve is $R = S \cdot e \cdot \omega$, with *S* being the surface of the catchment area.

Using Darcy’s law, the flow *Q* (in m^3/s) through any vertical cross section of the aquifer is :

$$Q = T \cdot i \cdot L$$

where :

- *T* = coefficient of transmissivity of the aquifer,
  (T = k·e; with *k* = hydraulic conductivity [LT^{-1}] and *e* = aquifer thickness [L])
- *i* = hydraulic gradient, [-]
- *L* = width of the vertical cross section through which the flow occurs [L].

The perenniality of the aquifer itself is proportional to the groundwater reserve (*R*), inversely proportional to the drainage yield (*Q*), and depends on the periods being without any infiltration within the period of 300 years.

The probability of having at less one drying out of the phreatic aquifer within 300 years, in function of the effective drainage, porosity and hydraulic conductivity of the open-jointed shale, is given in figure 2. This diagram shows the combined effect of porosity and hydraulic conductivity on this probability. These theoretical results have been confirmed by field data from the hydrological years 1994/1995 and 1995/1996, known as exceptionally dry periods in Belgium.

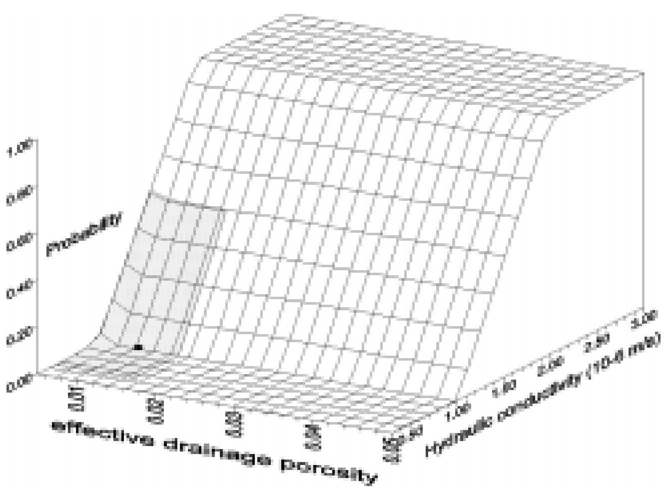

Figure 2 : Probability of having a drying out of the phreatic aquifer within 300 years

Comparison of Migration Times for Shale and Clay

The migration time is the time needed to transfer a thousandth of the initial concentration of radionuclides C_0 from the repository to the base of the impervious, slightly fissured shale barrier. The migration time was calculated by using a finite element approach and by solving the transport equation for different boundary conditions.

The results were compared with those obtained for sites located on a 100 m thick tertiary clay barrier overlying an exploited aquifer, assuming two unfavourable conditions :

- a hydraulic gradient i = 1, meaning an intense water withdrawal in the underlying aquifer, and
- a retardation coefficient R = 1, meaning that there is no retardation at all in the rock mass.

The results of the comparison are given below :

Type of host formation	Migration time $C = C_0/1000$	Type of solution
Repository located on a 100 m thick clay layer	2500 years	analytical solution
Repository located on fissured and permeable shale overlying slightly fissured and impervious shale	11700 years	numerical solution (mathematical model)
Repository located on shale (only diffusion)	3400 years	analytical solution

The longer migration time obtained by mathematical modelling is related to the fact that, in the upper part of the shale formation, the diffusion is limited to the lateral extend of the shale where calculations shown no ascending hydraulic gradient.

Assuming a retardation coefficient equal to 100, which is a valid assumption for heavy metals, the concentration $C_0/1000$ in the underlying aquifer is never reached (for shale as well as for clay barriers).

RESULTS OF FIELD RECONNAISSANCE ON A SPECIFIC SITE LOCATED ON DEVONIAN SHALE IN SOUTHERN BELGIUM

Site Criteria and Scope of the Field Reconnaissance

From a broad geological perspective, a suitable site must satisfy a number of criteria in order to be acceptable for the surface storage of low-radioactive waste. These conditions are :

- an absence of flooding danger,
- a geotechnical stability (the rock substratum of the site must, in any case, be able to support the installation of the waste repository without causing displacements or settlements which would disrupt the underlying drainage system or its structure),
- an acceptable degree of seismicity, for the same reasons as above (major earthquakes would of course also be capable of damaging the structure of the repository),
- an absence of mineral resources in the rock substratum (the presence of such resources could, in the course of time, encourage human intrusion and endanger the isolation capabilities of the repository),

- the hydrogeology of the site must be of such a nature that one can perform detailed characterisation and convincing modelling within the framework of a safety evaluation (it is the analysis of this last condition which forms the primary objective of the field reconnaissance).

The scope and the nature of the field reconnaissance were limited to that part which could be implemented within the very short period allotted (three weeks in the field).

Despite this, geo-electrical profiles, nine borings (including four core borings), a series of 'Lugeon' permeability tests and pump tests, as well as bore hole measurements (several of which were very extensive) were performed. Flow rates of the watercourses that drain the site were also measured.

Given that the geological maps of the site were recently updated, all existing data could be subjected to a critical and thorough analysis. Since the most relevant information was available, the geological and structural context could be optimally determined.

The materials gathered during field reconnaissance and the desk studies performed afterwards allowed to get a proper understanding of the hydrogeological, geological and structural framework, and to reach an unambiguous judgement on the suitability of the site.

General Description of the Site and its Surroundings

The site, which is located in a forest region close to medium range boroughs (4000 inhabitants) about one hundred kilometres south-west of Brussels, is a military base belonging to the Ministry of National Defence. This base, with an approximate surface area of over 220 hectares, was built in the early 1960's to store ammunition. The base has not been used since 1997.

Besides administrative and maintenance zones, which have no geological interest, the main zone of the site consists of 136 concrete ammunition bunkers, spaced approximately 50 m apart, along the site's internal road network.

Geological and Structural Framework

The described region is located on the southern edge of the Synclinorium of Dinant and forms the front zone of the Anticlinorium of the Ardenne

The site extends on both sides from a ridge at the elevation of 210 m, in the middle of a large depression, 3 km wide, known as the Depression of the Famenne. This depression resulted from periglacial phenomena and has an east-westerly orientation. In the north, the site runs down to a stream at an elevation of 160 m.

Towards the south, a pronounced ridge, which very clearly marks the landscape, limits the site. This ridge (160-260 m), whose substratum primarily consists of limestone from the Mid-Devonian period, divides the clastic sediments of the Ardenne Massive in the south from those of the Depression of the Famenne. Towards the north, the surface is limited by a pronounced projection in the relief, which forms the divide between the Depression of the Famenne and the Condroz. This pronounced projection essentially is an undulating plateau between 240 and 305 m in elevation.

Compliance with the Siting Criteria

Danger of Flooding

Because the site lies on the sides of a hill, there is absolutely no danger of flooding.

Geotechnical Stability

Subject to a geotechnical study, which must confirm the current data, the site is geotechnically stable. The slopes are indeed minor (less than 6.5%) and the structure of the underground consists of slightly heterogeneous shale. A fault runs through the site but entails only a slight disturbance and leads to a shale-on-shale contact. The risks of displacements or differential settlements are thus very limited or even non-existent.

Seismicity of the Zone

The detailed geostructural analysis reveals that there are no indications of recent or even quaternary seismic activity. The zone's seismic history is unremarkable. Only a moderate fault step to the north of the site needs more extensive study.

Natural Resources

The are no exploitable minerals or building stones in the underground of the site.

Hydrogeology

Reminder of Long-term Safety Principles

The long-term safety of a surface repository for low-radioactive waste is based on a multi-barrier system designed to prevent radionuclides from escaping into the environment or to limit this escape to a sufficiently low level. The containment system consists of three successive barriers :

- the first barrier is composed of the inert material, the matrix of the waste and the packaging in which the waste is contained;
- the second barrier is the construction works in which the waste is enclosed ;
- the third barrier is the geological and hydrogeological structures in which the repository is establish.

In order to select a potentially favourable zone for setting up a repository, one must define the geological and hydrogeological structures that can play the role of third barrier.

Water plays an essential role in the migration of radionuclides. Thus, it is absolutely necessary that one acquire a good knowledge of the hydrogeology of the zone on which the repository will be set up. It must be possible to characterise and model this hydrogeology with a precise localisation of the springs.

The simplest structure consists of a permeable horizon, which overlies an impermeable one, with a pronounced permeability contrast between both horizons and a limited number of springs. These springs make it possible to control the water which, eventually, will seep from the repository. The difference between permeable and impermeable rocks is arbitrarily set at 10^{-9} m/S (G. de Marsily – *Hydrogéologie qualitative*, page 70).

Shale can be considered under certain conditions as impermeable rock. In the zones where they crop out at the surface in Belgium, one indeed often sees structures that display clearly observable permeability contrasts. The quite permeable superficial horizon corresponds to the weathered shale and the deeper impermeable horizon corresponds to the unweathered and less permeable shale rock. Although the permeability of unweathered shales is generally greater than 10^{-9} m/s because of the presence of thin fissures, such geological structures can be considered as impervious provided that an ascending hydraulic gradient exists in the shale mass.

Field Observations

The field reconnaissance revealed that a pronounced permeability contrast existed between the weathered and the unweathered shale (Figure 3). This contrast was clearly expressed in ten or so springs, within or in the immediate vicinity of the site, all of which lay between +185 m and +195 m in elevation.

This permeability contrast was characterised by heavily weathered surface shales with a thickness of 8 to 12 m, whose permeability ranged from $3.4{\cdot}10^{-7}$ m/s $\cdot \leq k \leq 2.4\ 10^{-6}$ m/s . This surface shale layer laid above slightly weathered shale, which was 25 to 35 m thick and displayed a permeability of $k \cong 3{\cdot}10^{-8}$ m/s. This shale formation, in turn, laid on top of the deeper, unweathered shale which displayed a low permeability ($k \cong 1{\cdot}10^{-9}$ m/s).

In these shale formations, no lithological and structural heterogeneities, which could influence the groundwater flow, were observed in the boreholes, with the following exceptions :

- Some boreholes had locally shown more calcareous and permeable facies or levels where the rock was broken in the deeper shale.

- The geophysical logging (tomography) revealed several rare heterogeneities in the southern part of the site.

- Several surface outcrops showed more silty and sandy facies. The phreatic aquifer beneath the site was completely drained by two well-defined brooks at the north and at the west of the site. The run-off flow rates, which were measured during the field reconnaissance, indicated that all the water was being collected by springs which came together at these brooks. No flow was observed between the springs. Because the period in which the measurements took place was short (a few weeks) compared with the hydrological cycles (years), one must be cautious in drawing conclusions. However, the springs appeared to be clearly localised. In contrast to this, a small part of the site in the east displayed diffuse springs.

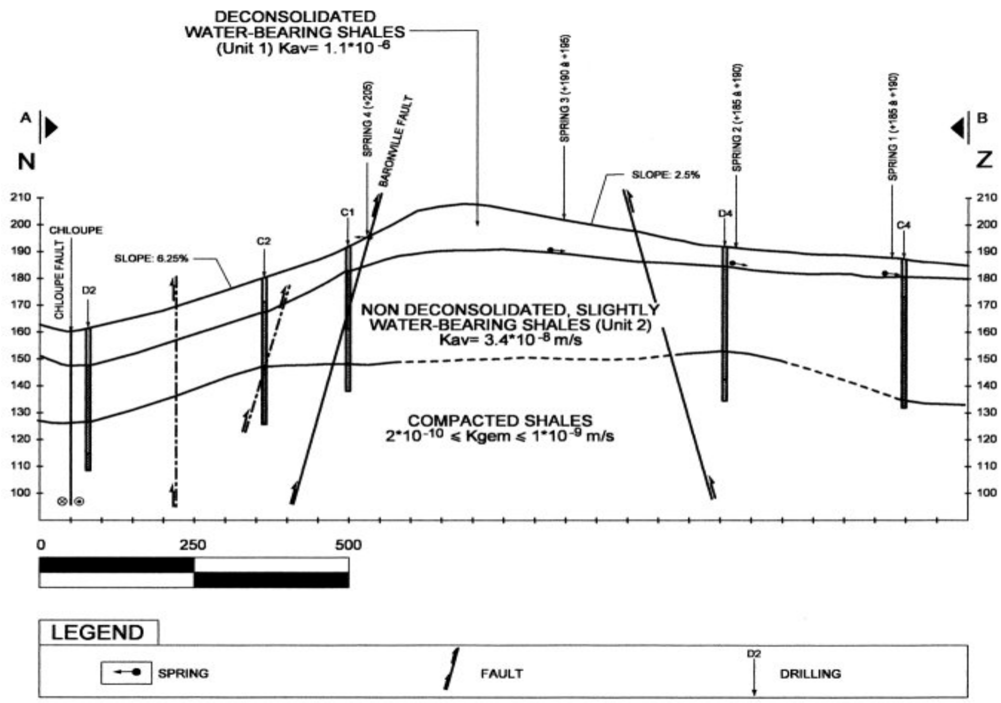

Figure 3 : Cross section through the site

The first significant and potential water-bearing limestone horizon was an upper Devonian one, which would be encountered at a depth of several hundred meters. With the current knowledge of the site, it is not possible to insure that water can be effectively transported through that horizon, nor to determine its piezometric head or its artesian character.

With the aim of creating a continuous ascendant hydraulic gradient, the water table of the phreatic aquifer contained in the surface weathered shale could be drawn down by a gravity drainage system installed beneath the foundations of the repository. For such a drainage system to perform well, the phreatic aquifer of the weathered shale must be perennial over the required time period (300 years). On the basis of the available climatological and hydrological measurements, and after having processed them according to the method presented in the first part of this paper, it appears that this perennial character would be insured (figure 4). The numerical model performed on the groundwater flow confirmed this result as well.

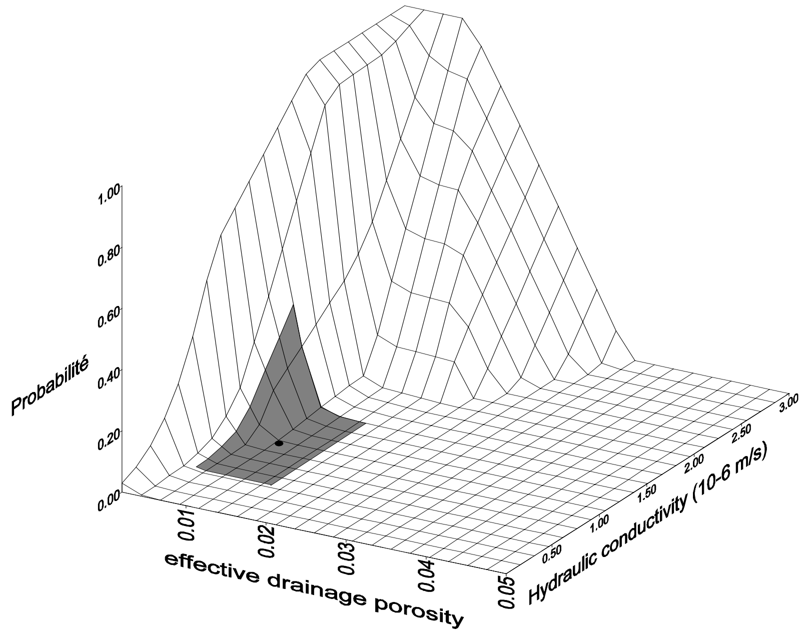

Figure 4 : probability of drying out of the phreatic aquifer within a period of 300 years

CONCLUSIONS

The theoretical approach of a permeable open jointed shale formation overlying a less permeable slightly fissured shale formation, outlined by the existence of springs has been confirmed by an extended hydrogeological field reconnaissance at a specific site with shale substratum. Permeability contrasts between the shale formations and perenniality of the phreatic aquifer have been established on field.

Mathematical modelling and analytical equations, applied to the data gathered on the site, demonstrated that, with the drawing down of the phreatic aquifer, the time for transport of contaminants through a 200 m thick shale layer may be similar to the time for transport through a clay layer. Therefore, it may be possible to use shale as a natural barrier for a low-level radioactive waste repository.

Limiting conditions are the existence of thin continuous sandstone beds in the shale body and the perenniality of the shallow phreatic aquifer.

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