TRANSFER PROCESS STUDIES THROUGH A TOARCIAN ARGILITE

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1. INTRODUCTION

Concerning long lived and high level radioactive wastes, the French Wastes Management Research Act (30th December 1991) has set three ways to be studied : separation and/or transmutation, conditioning processes and volumes reduction, and feasibility of geological disposal. ANDRA, in charge of this last point; has the responsibility to create and develop Underground Laboratories, to elaborate and propose the concept of the future disposal installations with safety demonstration, to be submitted to the French safety governmental authorities (Direction de la Sûreté des Installations Nucléaires). In order to asses these safety demonstrations, to build up its judgement, DSIN will ask technical advice, from Institut de Protection et de Sûreté Nucléaire (IPSN).

To be able to meet these requirements, IPSN is developing, in the framework of its Research and Development safety programs, in situ research concerning the confining properties of geological formations. In order to perform experiments in representative conditions concerning mainly geotechnical and hydrogeological properties of very low permeability rocks, appropriate underground sites have been selected in the past in granite and shale formations. IPSN activities at the Tournemire site are the most important activities regarding specifically research about characterisation of transfers through an argilite formation. This programme is developed in the framework of a cost sharing research contract between IPSN and EC.

2. OBJECTIVES OF THE IPSN TOURNEMIRE TUNNEL PROJECT

2.1 CHARACTERISTICS OF THE TOURNEMIRE SITE

By the end of 1990, IPSN initiated in situ research concerning argillaceous formations, considering the relative lack of knowledge about the behaviour of deep impervious clays. IPSN is studying a geological representative formation constituted by indurated claystones of Toarcian. This site has been selected because of its geological simplicity and also because a former railway tunnel gives access to the centre of the toarcian formation [1]. This is an hundred years old railway tunnel, 1885 meters long, in the close vicinity of the village of TOURNEMIRE (Aveyron), in the South of France, crossing a 200 m thick toarcian clay formation. The overlying limestones are 250 meters thick, so the geotechnical and hydrogeological conditions can be considered as representative of those of a deep repository.

The geological formations of the site are constituted by sub-tabular sedimentary layers of the lower and middle Jurassic Period. The stratigraphic formations involved in the Tournemire environment are as follows : Hettangian and Carixian (limestone, dolomites and interbedded marls), Domerian and Toarcian(245 m of marls and argilites), Aalenian, Bajocian and Bathonian (250 m of massive limestone and dolomites).

The prevailing lower limestone formations of Hettangian and Carixian contain the general lower regional aquifer. At the place of the tunnel, this aquifer is in charge below the impervious domerian and toarcian clays. The limestone layers from Aalenian up to upper Bathonian are the reservoir for the upper regional aquifer : at Tournemire, this aquifer lies in the lower part of the Aalenian and should feed the toarcian clay just below. Taking into account the hydraulic heads of the two aquifers, the fluid transfer through the toarcian argilites, which could behave hydraulically as a semi-permeable medium, is supposed to be vertical downwards.

2.2 WORK PERFORMED

The research program concerns hydrogeological properties in order to determine the characteristics of fluid transfers through the clay formation and it intends to perform modelling of these fluids transfers.

The Tournemire programme includes general geological and hydrogeological investigations, drilling operations for detailed geological studies (information about the depth, lithology, stratigraphy and hydrology), including structural studies relative to paleohydrogeological understanding, development of in situ tests including permeability tests at different level (mainly pulse tests), and boreholes equipment for interstitial water pressure measurement in low permeability rocks, sampling of material for laboratory studies on cores including petrophysical conventional identifications, diffusive characteristics, development of isotopic studies of the interstitial fluid and solid phase (matrix and fractures).

During a first phase, six boreholes have been drilled from the tunnel to the bottom and to the top of the Toarcian formation [1]. From the results of this phase, the programme has been developed thanks to long time hydraulic testings into boreholes specifically conceived for rocks of very low permeability, detailed fundamental studies relative to the possibilities for fluid transfer : diffusive transfer, different fluid potentials and associated fluxes measurements (chemical, suction...), and a natural isotopes measurement programme on the whole formation surrounding the tunnel aiming to characterise transfers both at the geological time scale and at the time scale corresponding to the perturbation due to the presence of the tunnel itself.

As these developments suppose a good understanding of the structural characteristics of the rock, and the achievement of a very high quality sampling phase of the formation, the on going work is based on a coring campaign of height boreholes around the tunnel [2].

3. MAIN RESULTS

3.1 PETROPHYSIC ROCK CHARACTERISTICS

The argilites appear to be very thinly bedded rocks mainly composed of clay minerals (40-50%) such as mica, kaolinite, illite and interstratified clay mineral close to illite, with about 10 to 40% of carbonates (dolomite, siderite and mainly calcite), 10 to 15% of quartz, and a few amount of chlorite, and pyrite.

The water content is very low : 1 to 3%. The free total access porosity measured with mercury is about 3 to 4%. The determination of the pore volume and the distribution size of these pores thanks to the B.E.T. method has shown that the dominant pore size family was centred on the value of 0.0028 µm corresponding to a total porous volume between 0.0071 and 0.0198 cm^3/g. The mean specific surface is about 10 to 25 m^2/g.

Hydraulic conductivity from laboratory tests with water is between 10^{-13} m/s and 10^{-14} m/s. The very low permeability, and the very small dimensions of the porous space lead to conclude that the fluid transfers in this kind of rock are not governed by convective processes, but by processes such as diffusion or others which have to be recognised and quantified.

Mass transfer studies of fresh tritiated water through argilite sample allowed both the characterisation of diffusive transfers, and those (permeation) under an hydraulic head (15 bars). Diffusion coefficient value is estimated around 3.5 10^{-12} m^2/s (with a total porosity between 8.5 to 10.2 %), and permeation coefficient mean value K_p is about 2.5. 10^{-14} m/s, meaning an average permeability coefficient K_H of 9.5. 10^{-15} m/s. These results seem to confirm that water or humidity transfers are essentially diffusive.

3.2 IN SITU HYDRAULIC CHARACTERISTICS

Many in situ permeability tests at different levels have been performed (mainly pulse tests) as well as geophysical loggings. The in situ permeability of the toarcian and domerian clay formations is very low, between 10^{-11} and 10^{-13} m/s [1]. The permeability of the calcareous carixian and aalenian formations is substantially higher : the Carixian has permeability of about 10^{-8} m/s and the Aalenian of about 10^{-6} m/s.

The aquifer heads, in the carixian and in the aalenian limestone correspond with the values that can be deduced from the hydrogeological regional and local system. But it has not been possible up to now to determine the actual head in the argilite formations because of the very long period to reach the steady state after the perturbation induced by drillings resulting from the very low permeability of this kind of rock.

3.3 CHEMICAL AND ISOTOPIC STUDIES FROM INTERSTITIAL PORE WATER

Isotopic and chemical contents of the interstitial fluids from the Tournemire argilites reveal an high heterogeneity of the fluids into the formation [3]. Due to the very low water content (1 to 3% wt) of this rock and to its high degree of induration. it has been impossible to use the squeezing technique for the extraction of total interstitial water. Thus, attempts to evaluate water chemistry have been made by leaching techniques, but interstitial water was exclusively recovered by distillation under vacuum to measure its stable isotope contents (^{18}O et ^{2}H).

Two fluid families, with different chemical facies have been identified (from absolute ethanol leaching) : (i) one $CaSO_4$-Mg type of fluid, representative of both aalenian limestone and high $CaCO_3$ content levels from the upper toarcian, and (ii) fluids with cationic composition dominated by Na coming from the argilites. These changes in the chemical facies probably result from cationic exchange between fluid and the solid matrix.

Four types of pore water can be distinguished from their stable isotope contents (^{18}O et ^{2}H) : (i) meteoric water which is isotopically similar to that of the overlying aalenian karstic aquifer, (ii) meteoric water whose isotopes content ise modified, probably due to exchange with host rock, (iii) water enriched in heavy isotopes possibly due to an evaporation process close to the tunnel and, (iv) water rich in heavy isotopes which could contain a part of residual connate water. The water types (i), (ii) and (iv) show a "randomly" distribution with depth which could probably be controlled by variations in the accessibility to rock porosity. This point has to be clarified, but the direct influence of both water content and $CaCO_3$ of the rock on these two stable isotopes contents has been clearly identified.

3.4 ISOTOPIC STUDIES FROM CALCITE FILLING IN FRACTURES

In addition to the study on pore water, isotopic measurements were made on the secondary calcite that filled the fractures intersected by the boreholes [1] and [2]. Variations of the stable isotopes content (^{13}C and ^{18}O) of these calcites versus depth appear more « regular » and homogeneous than those observed with pore fluid but the measured values imply that several different types of water have circulated in the fractures. An hypothesis to explain the distributions of ^{13}C and Iron contents with depth in fracture minerals would involve both upward and downward circulations, but it has to be confirmed.

All the secondary calcites are ^{14}C free. Their $^{234}U/^{238}U$ ratio are close to one, but they mostly reflect the contribution of a detrital fraction and cannot be used to estimate the age of the calcite. However, the very high $^{230}Th/^{234}U$ ratio measured from the bottom of the profile, probably indicates a secondary sink of ^{234}U. This suggests that water circulation should have occurred in the fractures at the lower part of the toarcian formation, 600 000 years ago at least.

At this stage of the study, it is impossible to establish any link between interstitial fluids and the « paleofluids » identified from the fractures. It could be possible that interstitial fluids reflect a very long and close interaction with the rock itself; and that paleofluids from fractures could represent fluids with very limited isotopic and chemical interactions with the matrix.

4. MAIN CONCLUSIONS

Some time scale indications can be deduced from the measurements on stable and radioactive isotopes both from the liquid and solid phases of the clay :

- after about one century, a distance of 20 à 30 m around the tunnel seems affected by rock desaturation as it can be seen from heavy isotopes values in the interstitial water;
- the existence of fluid which could include connate water indicates very long residence time into the formation. But these old waters seem to remain in coexistence (at some decimetres of distance) with quasi natural meteoric water;
- ^{14}C and U/Th values from fracture minerals lead to think that the clay formation has not been affected by important circulation for more than several thousand years, but it is possible that solutions percolated through a certain number of fractures less than 600 000 years ago.

On the basis of the available data (aquifers heads and permeabilities of the argilites), fluid circulation simulations through the clay were performed [1]:

- results confirm that taking into account only convective transfer, the transient period to reach the steady state after a significant perturbation is very long (several years), and the value of the interstitial water head at different levels in the Toarcian is close to the value of the topographic level at this point. Combined with the very low permeability of the rock, this means that the flow discharge rate by drainage that can be anticipated, specially at the tunnel level, is very low (below 10 cm^3/h) and very difficult to estimate from in situ tests;
- taking into account only transfers thanks to pure diffusion, calculations indicate that the formation could be leached by meteoric water in less than 2 Ma using an effective molecular diffusion coefficient of deuterium close to the one of water into water, or could not be leached after 130 Ma, using a coefficient corrected from porosity and tortuosity. This proves that such simulation would need a better knowledge of the diffusion coefficient of water through clays.

As a result of the coring campaign corresponding to a circular plan (40 m of diameter) perpendicular to the tunnel axis, the argilite formation in this zone appears to be characterised by the existence of two areas structurally different : a west zone relatively highly fractured, and an east zone without any fracture [2]. In some cases, one can observe fault zones, not only infilled with calcite, but mainly constituted of highly disturbed clay material of less good properties (physical and mechanical). The presence and the importance of such structures into argilite formation should be of great interest regarding fluid transfer possibilities.

5. REFERENCES

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