

# DISPOSAL OF RADIOACTIVE WASTE INTO CLAY LAYERS THE MOST NATURAL OPTION.

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## ABSTRACT

Among the geological formations suitable for the disposal of radioactive waste, the clay formations provide outstanding opportunities : impermeable for water, self-healing, strongly absorbing for ions, widespread in nature.

The self-healing properties of large clay deposits have been demonstrated by their auto-sealing and plastic response to tectonic stress and magmatic intrusion. The discovery of fossil trees preserved after geologic periods of burial in clay is one of the most dramatic illustrations of their entombment ability.

The physicochemical and hydrologic characteristics of the Boom clay are very favorable for the confinement of migrating radionuclides within the layer. Except for the extremely long half-lives ( $^{237}\text{Np}$ ,  $^{129}\text{I}$ , ...) no radionuclide can escape from the clay body.

The effects of heat, metal corrosion, material interaction and biochemical degradation on the natural properties of the clay layer are discussed in some detail and related to the natural properties of the clay formation which have to stay unaltered for geologic periods.

The first Safety Assessment Report, established by NIRAS-ONDRAF in close collaboration with SCK-CEN, has been submitted to a multi-disciplinary task force which is to advise the Belgian Government on the suitability of the Boom clay layer below the Nuclear Research site of Mol as a potential host formation for nuclear waste coming from the electronuclear program.

## INTRODUCTION

Throughout the world, attention was initially exclusively directed towards the use of crystalline and saline formations for the disposal of radioactive waste. For example, in the US essentially basalt, salt or tuff formations were considered being suitable as host rock formations. In Europe and in particular within the EEC, the situation developed differently from the onset of the CEC program in the early seventies. Three types of formations were selected for further investigation : crystalline formations (especially granite), saline formations (mainly salt domes but also bedded salt) and argillaceous formations (clays and shales). An extensive review of the potential merits of each of these formations, and their occurrence throughout Europe, was published as the so-called "European Catalogue" which ranked the three formations at the same level (1).

In this European Catalogue, a first overview was given of the potential role clays might play in the disposal of radioactive waste. Large deposits, suitable for further investigation, were cited and among the EEC countries Belgium and Italy jointly took the lead in developing the clay option (2). Regional investigation studies were undertaken in Italy by ENEA and its associated institutes which, on the one hand, aimed at a better understanding of the role clays submitted to tectonic action can play in the long term isolation capacity of formations from water penetration (3) and, on the other hand, shed some light on the thermal stability of clays, even under extreme temperature gradients.

In Belgium, a site-specific approach was followed which concentrated from the beginning on the Boom-clay layer situated at a depth of 200 to 300 m underneath the Nuclear Research Center at Mol, Belgium. Since the mid-seventies, a comprehensive R&D program (4) has been undertaken on the hydrologic, geotechnical and radiochemical properties of the Boom clay by carrying out investigations on samples taken during exploratory drillings with non-disturbed sampling and geophysical logging. The results were so encouraging that in 1980 the decision was taken to build an underground laboratory within the Boom clay at a depth of 230 m below ground level.

In the meantime, this success story has aroused interest for argillaceous formations all over the world, with new activities going on in France (5), Switzerland (6) and South Africa (7).

Finally, it has to be stressed that clays, as was evidenced by the Swedish program, play a major role in all backfilling materials used to seal the repository in crystalline formations from unwanted water penetrations. To a certain extent this sealing property extends even to salt domes and deep dry mines which are protected from surface water penetration by thick clay covers extending over very wide areas and constituting natural hydraulic barriers.

## LONG TERM ISOLATION CAPACITY OF CLAY

A generic study carried out by ENEA on 19 different sites (8) spread all over the Italian peninsula, has shown that clays are very impermeable to water (permeabilities down

to  $10^{-11}$  m/s) and even to geothermal fluids under a pressure of 40 to 80 bars and temperatures ranging from 150 to 250°C.

Secondary permeability created by tectonic effects is very limited and the general assumption that this permeability of clay formations is significantly reduced by faulting and fracturing phenomena, should therefore properly be brought to its real importance. While clays are deposited in a marine environment under oxic conditions, these are turned into reducing conditions during the first diagenetic process. This is generally proven by the occurrence of pyrites (FeS<sub>2</sub>) which derive from bacterial reduction of sulphate (SO<sub>4</sub>=). Studies at the ORCIATICO site have furthermore shown that clays can withstand magmatic intrusion. A few meters of thermometamorphic changes occurred in pleistocene clay under a temperature gradient of 800°C or higher. Recrystallization phenomena have been observed with formation of pyroxenes, plagioclase and biotite near the hot interface. It is, however, very important to note that, after some time, smectites are formed again in the metamorphic halo.

Finally, a highly interesting discovery was made of a fossil forest at DUNAROBBA which is estimated to be 1.5 M years old and still showing the original ligneous structures of the trees. The hydrologic isolation and/or geochemical barrier within the clay assured the preservation of the organic perishable material, though the clay layer covering the trees did not exceed a thickness of 10 m.

This highly important research work carried out by our Italian colleagues within the CEC program shows that clays are the natural entombment material of biological and mineral substances which can be preserved for geological periods.

#### GEOLOGY AND GEOCHEMISTRY OF THE BOOM CLAY IN BELGIUM (9)

At the site of the underground laboratory, the Boom clay is situated at a depth of about 200 m and has a thickness of roughly 100 m. The layer is part of an alternating sand/clay sedimentary sequence down to 800 m depth. The Boom clay is from the midoligocene period (20 to 30 M years old) and is part of an extensive clay deposit covering NW Europe and extending as far east as Poland and as far north as Denmark. The layer dips at an angle of 1% in the NE direction. The clay deposit is affected by a system of faults resulting from the Rhine tectonic activity. The nearest proven fault is about 7 km from the nuclear site and was still active during the Pliocene period (2 - 5 M years ago).

The very compact clay layer is over-consolidated as a result of the pressure buildup exerted by the sedimentary overburden and displays a band structure. Within each band of roughly 10 m, there is a black horizon rich (up to 5%) in organic matter (humic acids) and a thin carbonaterich layer transformed into "septaria" nodules. The bulk of the clay is

composed of 40-50% illite, 10-20% smectite and 10-30% interstratified smectiteillite. The organic matter content is generally around 1 to 2% (mean average 1.23%). The carbonate content has been redistributed throughout the interstitial liquid showing an alkaline pH (8.5-9.5), which is physicochemically ruled by the bicarbonate system in solution.

The initial sulphate content has been transformed into pyrite (FeS<sub>2</sub>) which occurs in the form of small granules with sizes from mm to cm and could make up about 5% of the total formation.

The groundwater permeability is extremely low and piezometers installed at regular intervals in the clay body show a mean value of  $10^{-12}$  m/s. On the regional scale (2500 km<sup>2</sup>), the permeability is of the order of  $10^{-10}$  m/s.

#### RETENTION PROPERTIES OF BOOM CLAY FOR RADIONUCLIDES (10)(11)

The retention properties of the Boom clay are essentially determined by four physicochemical parameters: Molecular diffusion, Redox potential, Alkalinity, Complexation, and by one hydrologic factor: the Darcy permeability.

- The migration ability of a radionuclide is determined by the diffusion coefficient D (cm<sup>2</sup>/s) and by the retardation factor which encompasses all phenomena by which a nuclide moves slower than a water molecule. Clays display ion exchange properties for Cs and Sr, but hydrolysed multivalent ions are trapped by adsorption and complexation phenomena. Anionic species move with the water. Table I shows some experimental diffusion coefficients.

TABLE I  
Diffusion Coefficients D in Boom Clay at Different Consolidation Pressures

Nuclide	Diffusion Coefficients (cm <sup>2</sup> /s)	
	Consol. 5 bars	Consol. 45 bars
HTO	$3.5 \cdot 10^{-6}$	$3.2 \cdot 10^{-6}$
I	$4 \cdot 10^{-6}$	$1.4 \cdot 10^{-6}$
Cs	$2.8 \cdot 10^{-9}$	$1.6 \cdot 10^{-9}$
Sr	$2 \cdot 10^{-7}$	$5 \cdot 10^{-8}$
Eu	$\leq 10^{-9}$	---
Np	$2.1 \cdot 10^{-10}$	$3.2 \cdot 10^{-11}$
Pu	$3 \cdot 10^{-10}$	---
U	$\leq 10^{-9}$	---
Tc	$2 \cdot 10^{-10}$	---
	( $\times 5 \cdot 10^{-8}$ )	

- The redox potential ( $E_h$ ) in natural deep clay deposits is in general very reducing due to its pyrite ( $FeS_2$ ) content. However, in situ experiments have shown that a positive (oxidizing) potential is measured by a Pt electrode in clays which have previously been contacted with air. By keeping the electrode in the clay and shutting off air inlet, a gradual decrease of the  $E_h$  value is observed. The first weeks this drop is fairly rapid (from + 0.3 V to - 0.25 V vs SHE) but below that level it takes up to 2 years to reach - 0.35 V. This redox potential corresponds to a redox system ruled by the  $FeS_2$  system in water (Fig. 1). Under these strong reducing conditions most multivalent radionuclides are brought to their lower valency states, Pu(III), U(IV), Np(IV)(V), Tc(IV), etc.
- The alkalinity characterized by the pH value of the carbonate/bicarbonate equilibrium (pH 9) also varies indirectly with the presence of oxygen. The reaction of oxygen with a pyrite-containing medium leads to oxidation with acid formation and reaction with carbonate leading to a drop in pH. By shutting off the air, the natural conditions are gradually restored at the expense of the solid  $CaCO_3$  present in the formation and the pH values rise again to the value of the carbonate/bicarbonate buffer (Fig. 2).
- In such conditions, the hydrolysable ionic radionuclide species of the actinide and lanthanide series are

transformed into hydroxides, carbonates and colloidal complexes which are easily adsorbed on the clay particles and the organic matter.

- Finally, the organic matter overrules the complexation effect which might be exerted by the carbonate/bicarbonate species in the interstitial solution. The humic acids are high molecular weight polyelectrolytes on which multivalent ions, particularly actinides and lanthanides, attach themselves very strongly. Since 99% of the humic acids have molecular weights in excess of 40.000 Dalton, their mobility within the pores of clay minerals is practically zero. As long as the humic acids do not move, the attached isotopes cannot migrate. Recent spectroscopic studies have shown that interstitial water contains tiny colloids of humic acids with a diameter of 0.13  $\mu$ m, sometimes aggregated to particles of 0.26  $\mu$ m. However, these colloidal humic acids do not make up more than 1% of the total humic acid concentration.
- The most stringent hydrological parameter is the Darcy permeability. Measurements, first in laboratory, on so-called "undisturbed" samples, showed a permeability coefficient of  $10^{-10}$  m/s. In the meantime, the permeability has been measured in situ by installing a sequence of micropiezometers at different distances from the gallery wall within the clay formation at 230 m depth. The measurements are taking place

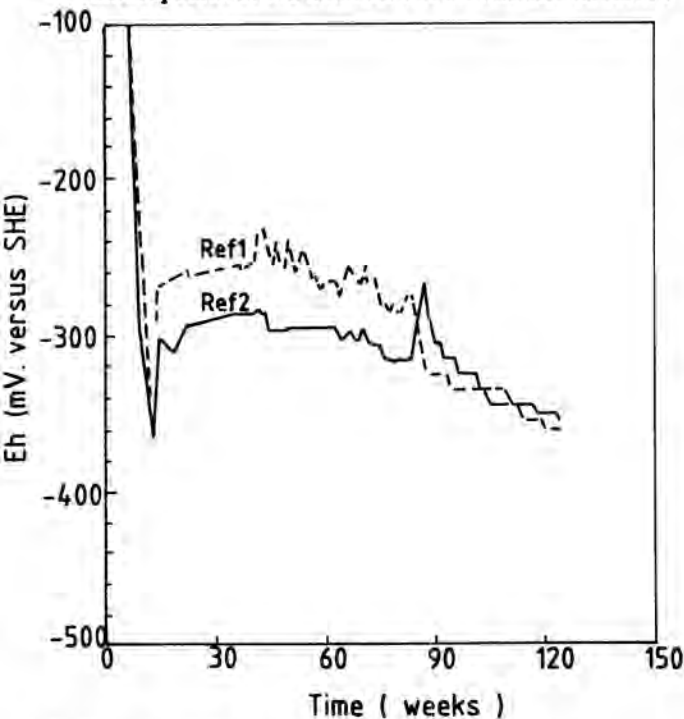


Fig. 1. In Situ Eh Evolution at Room Temperature October 1986 - March 1989.

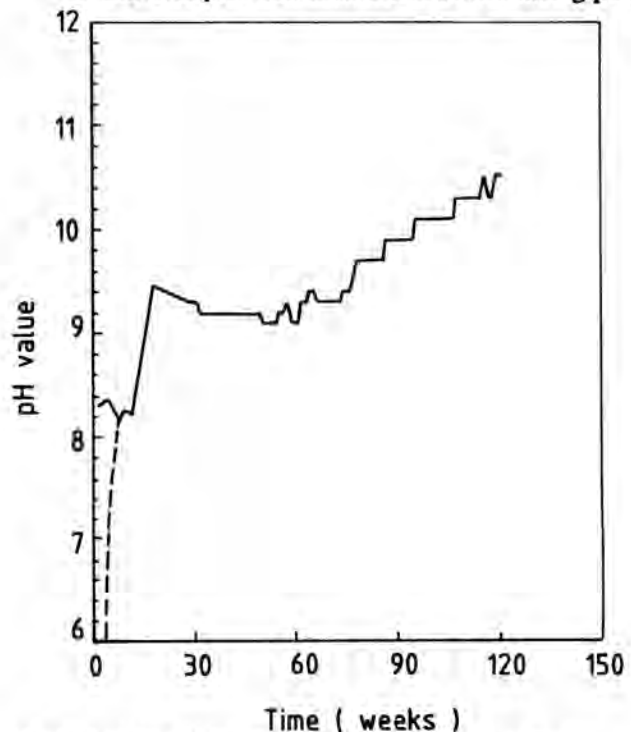


Fig. 2. In Situ pH Evolution at Room Temperature October 1986 - March 1989.

under the artificial hydraulic gradient of 22 bars towards the walls of the gallery and led to astonishingly low permeabilities of  $10^{-12}$  m/s. These measurements have now been going on for several years and the measured values seem unchanged. Macroporosity tests are in preparation.

- The retention properties of the Boom clay formation for radionuclides are the result of the combination of very favorable hydrologic and physicochemical parameters which reduce their mobility to such low values that none of the characteristic actinides and lanthanides of the fuel cycle can leave the clay formation. A few exceptions, however, deserve further attention e.g.  $^{129}\text{I}$ ,  $^{237}\text{Np}$ ,  $^{99}\text{Tc}$ ,  $^{135}\text{Cs}$  and some minor nuclides, because of their extremely long half-lives.
- If the clay layer is kept in its natural condition during excavation and waste loading operations, there is no evidence that radionuclides will migrate to surrounding aquifers and from there to the biosphere.

#### INFLUENCE OF THE WASTE FORMS ON THE NATURAL PROPERTIES OF CLAYS

As outlined in the previous section, hydraulic and mechanic excavation response and a massive intrusion of air into the formation must be avoided during excavation operations by sealing and lining the gallery walls as soon as possible by appropriate means. The remaining questions are related to effects of waste disposal: heating of the formation, radiolysis of interstitial water, chemical interaction with waste forms and structural materials.

#### Heat Transfer

The most drastic change to the natural environment brought about by the disposal of HLW is the in situ heat production. The most vulnerable component in the clay formation being the interstitial water, care must be taken not to modify its content. The Boom clay at the level of the underground laboratory contains 18% water.

The heat transfer coefficient has been measured during in situ pilot scale experiments, centered around the heated corrosion loops. A loop heated at  $170^{\circ}\text{C}$  was kept operational during two years (1986-88) and interpretation of the temperature profiles around the loop yielded a heat transfer coefficient of  $1.72 \text{ W/m}^2\text{C}$  which is, within the experimental error, the same as the value of  $1.69 \text{ W/m}^2\text{C}$  determined at a surface clay quarry near Boom in 1980.

In order not to damage the clay/water equilibrium, it has been decided to limit the temperature increase at the interface backfill/clay or concrete/clay to  $100^{\circ}\text{C}$ . At the interface clay/sand at the roof of the formation, a maximum temperature elevation of  $5^{\circ}\text{C}$  may not be exceeded. Under these limitations the admissible thermal load is set at 25

kW/ha which corresponds to an equivalent of 37 standard COGEMA vitrified HLW containers cooled during 50 years. If the cooling time is decreased, the anticipated disposal area must be extended proportionally to the nominal increase of the heat output per canister.

A secondary effect from the presence of heat sources in the clay layer is the increase of the water flow in the surrounding aquifers. Simulation studies have shown that an increased convective water flow in the sandy aquifer above the clay layer cannot be avoided and might reach 11 m/year or twice the natural groundwater flow at that depth.

#### Chemical Interactions

Incorporation of extraneous compounds in the clay layer might in the long run alter the natural geochemical equilibrium. Among the waste forms borosilicate glass, cement matrices and bitumen matrices are to be expected. Structural and backfill materials are cement, steel and clay based materials.

- Borosilicate glass is very stable in geologic environments and will undergo an extremely slow dissolution process. Exchange will take place between the cations in the interstitial water and the ions in the amorphous glass matrix. In case of clay as backfill material with direct contact, a very slow diffusion-controlled reaction will attack the glass superficially. In case of cement backfill the high alkaline pH might dissolve the glass at a faster pace. Such processes will in any case take thousands of years and not induce significant geochemical changes.
- Cement matrices, be it structural materials or waste forms, will react with the surrounding interstitial liquid and drastically increase the pH to 12 or 13. These fairly high pH values will only be obtained locally at the interface between concrete and clay and may influence the stability of clay minerals all the more since temperature increase is not excluded. However, the reaction zone will be rather small since the concentration gradient of the produced NaOH or KOH will rapidly diffuse into the interstitial liquid. Over a quite long period of time the pH around the concrete will be determined by the  $\text{Ca}(\text{OH})_2/\text{CaCO}_3$  system and range from initially 12 to 9 after exhaustion of the  $\text{Ca}(\text{OH})_2$  supply.

The solubility of most hydrolysable radionuclides will be lowered but Cs and Sr will continue to migrate at the same pace or faster. However, their half-life of about 30 years is very small compared to the theoretical migration period required to reach the clay/sand interface. This implies that the radionuclides will have decayed completely and

have been transformed into the stable species  $^{90}\text{Zr}$  and  $^{136}\text{Ba}$ .

In the pH range of 9 to 12, the migration of the actinides and lanthanides will be reduced due to the formation of hydrolysed and polymerized species but the role of the organic matter as an immobile adsorption complex for these radionuclides might be weakened and needs further study.

The use of cement-type materials with a lower Ca/Si ratio might be considered, but it needs very careful thought, since other parameters e.g. porosity and mechanical strength, are of the same importance to the overall concept. A thorough study of the reaction mechanisms, involved over a very long period of time, is therefore desirable.

Since clays are also proposed as backfill materials, the interaction between concrete and clay is of general significance for crystallinerock as well as for clay-type repositories.

But anyway the extension of the "chemically" disturbed zone must be small compared with the size of the repository formation. In this case the global geochemical equilibria will not be affected. When considering the Boom clay case, a repository of 1.2 km<sup>2</sup> with 12 km of galleries (3.5 m diam.), this mass ratio amounts only to about 0.1%.

#### – Bitumen and organic matrices.

An important fraction of MLW is embedded into a bitumen matrix. These waste forms contain generally insolubilized sludges from the effluent treatment plant and evaporator residues which are rich in  $\text{NaNO}_3$ . When bitumen enters into contact with interstitial pore water, the soluble salts, essentially  $\text{NaNO}_3$ , will be hydrated and slowly eluted from the bitumen matrix to enter the nearby materials and the surrounding clay. Taking into account the highly reducing properties of the medium, it is to be expected that the nitrates will be transformed into nitrites ( $\text{NaNO}_2$ ) and further into more reduced species and even gaseous  $\text{N}_2\text{O}$  by bacterial action. Relatively little is known about these phenomena in totally anaerobic conditions prevailing in deep clay formations.

In aerobic conditions, existing in the beginning of the disposal period, the bitumen matrix will be slowly attacked by bacteria with the formation of  $\text{CO}_2$ , which will dissolve in the groundwater. If nitrates serve as oxygen donor for bacterial reac-

tions, a fraction of the bitumen could be degraded and yield more soluble and perhaps complexing compounds.

Once the oxygen donors have disappeared, anaerobic fermentation could theoretically further degrade the matrix, but this phenomenon is unlikely to occur on macroscopic scale.

The discovery of unaltered wood in the Italian fossil forest of DUNAROBBA indicates that anaerobic decomposition of ligneous material is very limited even over geologic periods of time.

The most preoccupying problem which remains to be investigated is the role  $\text{NaNO}_3$  will play in the potential decomposition of bitumen and to what extent these by-products might accelerate the migration of actinides.

Bituminized waste, free of nitrates and positioned in zones where the temperature is not expected to rise, are probably stable for geologic periods.

Low-level waste with a significant fraction of cellulosic material will decompose and produce gaseous decomposition products which cannot escape from the compact clay. Mineralization prior to conditioning in concrete or bitumen is a prerequisite to any disposal operation in order not to jeopardize the natural intrinsic confinement of the clay layer.

### Gas Formation

During the sedimentary formation period of the clay layer, the fermentation gases could escape from shallow depths into the seawater. The more compressed buried horizons were this way gradually freed from fresh organic matter. Once the petrifiable mass had disappeared, the residual organic substances turned into lignitic fractions and humic acids which remained practically at the place where they were originally deposited.

When digging a gallery into a compact and impermeable clay formation which has been in geochemical equilibrium with its surroundings for geologic periods, the problem arises how to cope with possible gas formation due to interaction between structural components or waste materials and the geologic environment.

This problem recently attracted very much attention (12) and can be summarized as follows :

- any corrodable metal will produce an amount of hydrogen gas proportional to the weight of metal transformed into its oxide;
- any biochemical degradation reaction produces  $\text{CO}_2$  and  $\text{H}_2\text{O}$  under aerobic conditions and  $\text{CH}_4$  and/or  $\text{H}_2$  under anaerobic conditions;

- any radiation (alpha, beta, gamma) inside a water-containing material will induce the formation of radiolysis gases ( $H_2$  and  $O_2$ ) which can recombine or react with the surrounding medium; in presence of organic matter,  $H_2$  and  $CO_2$  are formed.

From the point of view of magnitude, the corrosion process is by far the most important one, followed by biochemical degradation reactions. The production of radiolysis gases is relatively small compared to the first two types of gas production reactions. Experimental corrosion tests in the underground laboratory over 2.5 years have shown that corrosion rates of 5 to 7  $\mu\text{m}/\text{year}$  are observed on C steel. The final products are  $FeOOH$  (Limonite) and  $Fe_2O_3$  (Haematite) at room temperature. Above  $150^\circ\text{C}$   $Fe_3O_4$  (magnetite) should be formed but has not yet been detected. Per mole corroded Fe 3 ion equivalents of H are released into the environment. If the redox potential continues to decrease, the formation of  $H_2$  gas is to be expected. A complete transformation of Fe into a series of corrosion products ( $Fe_2O_3$ ,  $Fe_3O_4$ ,  $FeO$ ,  $FeCO_3$ ,  $FeS_2$ ) can thermodynamically produce 400 to 600  $\text{cm}^3\text{STP}$  per g Fe depending on the type of end product. However, anaerobic corrosion seems to proceed very slowly at about  $1\ \mu\text{m}/\text{year}$  or less.

If an over-pack of 25 cm thickness is provided as a cylinder around the HLW canisters, the corrosion process will take thousands of years to attack the metal and produce potentially 400 to 600  $\text{m}^3\text{H}_2$  gas per t Fe. A compact clay formation can never cope with such a large potential volume of gas without losing its natural integrity. Indeed, the hydrogen gas produced will first fill the voids of the engineered barriers (concrete or clay) and subsequently migrate towards the interstitial liquid. A fraction of it will react with the solid phase but the remainder will stay in the liquid phase till the solubility is exceeded with formation of hydrogen bubbles. The most difficult question is to know whether this potential  $H_2$  volume will be produced or whether the corrosion process will be slowed down or even blocked by the presence of a protecting hydrogen film or a passivating chemical compound. Any decision on the use of a metallic overpack depends on the outcome of these corrosion studies.

Extensive studies have been undertaken at UKAEA Harwell (13) which indicated low gas transfer coefficients in dry concrete (diffusion coefficient of  $10^{-4}\ \text{cm}^2/\text{s}$ ) but that moist concrete is very impermeable. A buildup of hydrogen within the technical structures is expected to occur and might result in the expulsion of water but no guarantee can be given about the real outcome without pilotscale experiments in representative conditions. The corrosion of metal fragments incorporated in MLW packages deserves also major attention, since their corrosion will lead to a gas

production the volume of which must be compatible with the uptake capacity of clay formations.

### Radiolysis

The presence of HLW canisters in underground galleries will, in the absence of an overpack, induce radiolysis in the structural and backfill materials. Whether clay or concrete surrounds the HLW canisters, the gas production by radiolysis will be rather similar, since the water content of both materials is about 20%. A HLW canister equivalent to 1 t Heavy Metal cooled for 50 years will contain  $1.14 \cdot 10^5$  Ci with a mean energy of 0.6 MeV. Taking into account a self-absorption factor of 0.13 and a  $G_{H_2} = 0.1$  value the average  $H_2$  production by radiolysis amounts to  $1.1 \cdot 10^7$  Mole/s. This process is essentially due to the presence of fission products which decay almost completely within 300 years. The accumulated  $H_2$  produced throughout this period amounts to  $3.8\ \text{m}^3\text{STP}$   $H_2$  which is compressed to  $0.17\ \text{m}^3$  at 22 bars. Such a volume is compatible with the uptake of a concrete cylinder of 1 m thickness the void volume of which is of the order of  $1.4\ \text{m}^3$  per current meter cylinder.

A first analysis of the radiolysis caused by betagamma emitters in bitumen shows that the gas production ( $H_2 + CO_2$ ) is very limited. However, the alpha content (0.26 Ci/220 l) produces  $2.4 \cdot 10^{-9}$  Mole/s or  $47\ \text{m}^3/\text{year}$  if the total amount of expected bitumen drums (28000) is disposed off in clay. The production of this gas volume declines only slowly over a long period of time (principally with the half-life of  $Pu^{238}$  (80 years) and accumulates in the gallery. But since these drums are envisaged to be stored in a 3.5 m diam. gallery of 1 km length with 24 drums/section, it is to be expected that the void volume of the backfill material will be sufficient to take up a volume of several thousands  $\text{m}^3\text{H}_2$  at standard temperature and pressure.

Anaerobic fermentation of organic materials, e.g. bitumen and plastic materials embedded into concrete, must be carefully investigated on this potential gas formation in order not to jeopardize the natural confinement of the impermeable clay layer.

Very little is known about anaerobic fermentation, but more and more evidence seems to emerge that degradable organic matter is unwanted in a deep impermeable clay layer. To what extent the organic matter has to be mineralized has not yet been established and will have to be thoroughly studied in the near future, before any decision is taken with regard to the disposal of organic matrices in a deep clay layer.

### CONCLUSIONS

- The self-healing capacity of clay layers submitted to tectonic effects and to hydrologic pressure of overlying aquifers have shown to be effective over geologic periods of time.

- The preservation of fossil trees in shallow clay layers is a geological demonstration that ligneous materials can be preserved from decomposition for millions of years.
- The hydrological characteristics of the Boom clay layer (Belgium) are very favorable for geological disposal since in situ tests have shown that the permeability coefficients are of the order of  $10^{-12}$  m/s.
- The retention properties of the Boom clay rely on the stable carbonate/bicarbonate pH buffering system (pH 8.5 - 9.5) and on the reducing properties of FeS<sub>2</sub> which impose a natural E<sub>h</sub> value of -350 mV. The presence of natural poly-electrolytes plays a fundamental role in the in situ fixation of multivalent ions. When these conditions remain unaltered, the migration of radionuclides is reduced to an extremely slow diffusion phenomenon which prevents the radionuclides from migrating into the surrounding aquifers.
- The most important perturbation of the natural medium is the in situ production of heat which is orders of magnitude above the natural geothermal gradient. By imposing stringent limits, e.g. 25 kW/hA, no major hydrogeologic disturbances are expected to be induced.
- The interaction between waste forms and structural materials on the one hand and the clay body on the other hand, needs a thorough investigation. No deleterious effect on the geochemical equilibria seems to be expected from the disposal of vitrified HLW. The use of large amounts of concrete will markedly influence the near field by drastically increasing the pH. But, on the time horizon of thousands of years, the global impact of this on the clay formation is negligible.
- The incorporation of corrodible metals in the underground structures or in waste forms, might threaten the natural confinement capability of the compact clay layer and care must be taken to limit the metal load to quantities which are below the natural uptake capacity of the layer. Alternatively, corrosion-free metals, e.g. Hastelloy, can be used as overpack material for HLW.
- The disposal of organic materials into impermeable clay layers may not be decided before extensive evidence has been provided of the biochemical stability of the organic substances. The conjunction of soluble nitrates with bitumen matrices might lead to the local destabilization of the electrochemical conditions and must be stud-

ied in situ before any irreversible decision is taken.

- Radiolysis of inorganic waste forms and structural materials does not affect the near field or the geochemical equilibria to a significant extent. However, radiolysis of organic substance might produce gas volumina exceeding the natural uptake capacity and can in principle lead to the formation of complexing agents. Real-scale experiments with the radioactive bitumen drums are required to provide adequate answers.

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