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HIGH INTEGRITY CONTAINERS FOR INTERIM STORAGE OF NUCLEAR WASTES USING REACTIVE POWDER CONCRETE

APPLICATION DES BETONS DE POUDRE REACTIVE AUX CONTENEURS DE HAUTE INTEGRITE POUR L'ENTREPOSAGE DES DECHETS RADIOACTIFS

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ABSTRACT: In this study, the use of Reactive Powder Concretes for nuclear waste storage has been investigated. Apart from the compressive strength which is not the determining parameter, microstructural properties of RPC including very low porosity diminish mass transfers. These characteristics are of great interest in the conception of High Integrity Containers for long-term interim storage of nuclear wastes. We present in this paper the results obtained in the characterization of RPC to that respect. Nitrogen permeability, mercury porosimetry of RPC and diffusion of tritiated water are investigated and proved to be greatly improved.

RESUME : Notre étude envisage l'utilisation des Bétons de Poudre Réactive pour l'entreposage des déchets radioactifs. Hormis la résistance en compression qui n'est pas un facteur déterminant, les caractéristiques microstructurales des BPR, notamment une porosité très faible, conduisent à des propriétés de transfert améliorées. Ces propriétés sont d'un grand intérêt dans l'optique de la réalisation d'un conteneur de haute intégrité pour un entreposage de longue durée des déchets radioactifs. Nous présentons ici les premiers résultats obtenus dans cette optique. Ils montrent que les propriétés de perméabilité à l'azote, porosité mercure et diffusion d'eau tritiée sont grandement améliorées.

1- CONTEXT, INTERIM STORAGE CONCEPT AND HIC DESIGN

The French nuclear industry, like any other industry, generates wastes. The harmful effects of these are due to their radioactivity. This can differ greatly depending on its type, intensity and lifetime, properties which are used to define 3 categories of wastes [1] :

- A-type wastes: short-lived wastes, with low to medium activity. These are now stored in surface sites managed by ANDRA (Agence Nationale pour la gestion des Déchets Radioactifs) and represent 90% of produced wastes.
- B-type wastes: low and medium activity wastes containing long half-life radionuclides (generally α). These include cladding and rod ends from nuclear fuel reprocessing.
- C-type wastes: high-activity, long-half-life wastes. These are for example, fission product solutions from reprocessing wich concentrate the essential initial activity of irradiated fuel. These wastes are vitrified and stored in shafts

at the reprocessing site until after their decay heat has peaked.

In the following we will consider only B-type wastes. For these, the law relating to radioactive waste management passed on 30 December 1991 [2] has the following consequence: the deep (and definitive) disposal of these wastes will not be decided before 2006 and therefore this cannot be envisaged before 2015-2020. Thus, (and this is one of the main axes of the law) an intermediate facility suitable for a long period must be developed.

In addition, the CEA, which is also a waste producer, has adopted a waste volume reduction policy. This policy involves "bulk" packaging of the wastes (note that this packaging also makes it easy to recover the wastes for reprocessing).

The long-duration interim storage and waste volume reduction can be envisaged only if long-term safety conditions can be guaranteed. This implies placing the wastes in a container that by itself ensures provision of a containment barrier, the high-integrity container (HIC).

The specifications of such a container are now being defined by the CEA, but they will include some of the following points: durability of the order of 300 to 500 years with respect to factors such as freeze-thaw cycles, water leaching, irradiation, drop-resistance around 5 metres.

On the basis of these factors, it appears important to us to know if reactive powder concrete (RPC) can be possible material for this type of container. We shall thus examine the advantages of this material.

2- STRONG-POINTS OF RPC AS A MATERIAL FOR HIC

Reactive Powder Concrete (RPC) is a class of concretes characterized by a high silica fume content and a very low water to binder ratio (0.10 to 0.17) [3]. A heat treatment, applied to RPC once demolded, with temperatures between 20°C and 400°C enables to enhance hydration and pozzolanic reaction, leading to improved mechanical and microstructural properties [4]. Finally, the presence of steel fibers makes it possible to obtain a high ductility material. Namely, RPC consists of a class of concretes whose compressive strength is ranging between 200MPa and 800MPa [3].

It is to be underlined that RPC is not only a high resistance material, but also *exhibits excellent microstructural and durability properties*. Depending on the adjustment of RPC composition, heat treatment and steel fibers content, it is possible to define a taylor made RPC mix design with the required characteristics, providing they lie in the above mentionned range.

As for the application of RPC as a material for a HIC, let us now examine the properties required by this demanding application.

2.1 Mechanical properties.

It is obvious that the static compressive strength of RPC (200 MPa to 800 MPa) is exceeding by far the service loads to which the container will be submitted. That is why it was decided to focus our study on the optimization of a RPC200 formulation (RPC exhibiting compressive strength in the range of 200 MPa).

Nevertheless, the possibility for the container to fall from a height of 5m without letting its content pass through is far more stringent. RPC200 is characterized by a fracture energy of 30000 J/m^2. As can be seen in Table 1, fracture energy of RPC is more than 200 times as high as that of Ordinary Concrete (OC) or High Performance concrete (HPC). At the same time, it is only one third as much as that of steel.

According to these figures, RPC behavior relative to shocks should be satisfactory.

Table 1 : Fracture energy (J/m^2) of ordinary concrete (OC), high performance concrete (HPC), reactive powder concrete (RPC) and steel.

OC	HPC	RPC	Steel
130 J/m^2	140	30000	100000

2.2 Containement properties.

It comes from the notion of HIC that it should be the only barrier between the wastes and the environment. This implies that the material should have excellent microstructural properties in order to prevent the migration of radioactivity from within the HIC and aggressive agents from outside the container.
The microstructure of RPC has been proved to be very tight [4]. In particular, porosity is very low (less than 4%). The reason for these enhanced microstructural properties is manyfold. First, RPC compactness is very high due to the small size of the constituant (the"coarser aggregate" is a fine sand) and to the optimalized granulometry. Finally, the use of a relevant heat treatment enables to enhance primary hydration and pozzolanic action of silica fume [5].
The microstructural properties of RPC should then be a strong point regarding confinement. In particular, porosity and, gas permeability results will be discussed below.

2.3 Durability.

As a consequence of the service life of HIC, it is necessary for this container to keep satisfying properties on the long run (300 years or more). The material used for HIC should then exhibit excellent durability properties.

It is long known that High Performance Concrete, used for many years in civil engineering, exhibit very high resistance to aggressive agents. This is all the more true for RPC since we have seen that RPC microstructure is greatly enhanced as compared to HPCs. The exposure of RPC to aggressive agents has been thorougly studied [6], in particular, chloride ions diffusion, carbonation resistance, freeeze-thaw scaling, abrasion resistance, are all greatly improved from HPC to RPC. Results obtained from comparative tests are summarized in table 2.

3. RESEARCH PROGRAM, INITIAL RESULTS

Our research program includes four components: microstructural characterisation of the material, study of its transfer properties, its mechanical behaviour and durability. Each of these will be described in giving the first results.

3.1 Material microstructural characterization

The objective of this program component is to obtain a detailed description of the RPC microstructure, as this is obviously related to material durability. The techniques used are mercury intrusion porosimetry, SEM

Table 2 : Compared durability properties of ordinary concrete (OC), high performance concrete (HPC) and RPC.

Property	OC	HPC	RPC
Chloride ions diffusion (10^{-12} m^2/s)	1.1	0.6	0.02
Carbonation depth (mm)	10	2	0
Freeze-thaw scaling (g/cm^2)	>1000	900	7
Abrasion coefficient	4.0	2.8	1.3

observations and a sorption-desorption test based on the BET technique developed by V. Baroghel-Bouny [7, 8].

The first available results are those obtained by mercury intrusion porosimetry. The difference between the pore size distributions for a mortar studied at the CEA [9] and RPC are shown in Figure 1. The total porosity of our RPCs is measured at a level of 3-4% compared to 13% for the mortar. This value is very low and close to the limit of detection of the mercury porosimetry technique. Sorption techniques should provide us more accurate data.

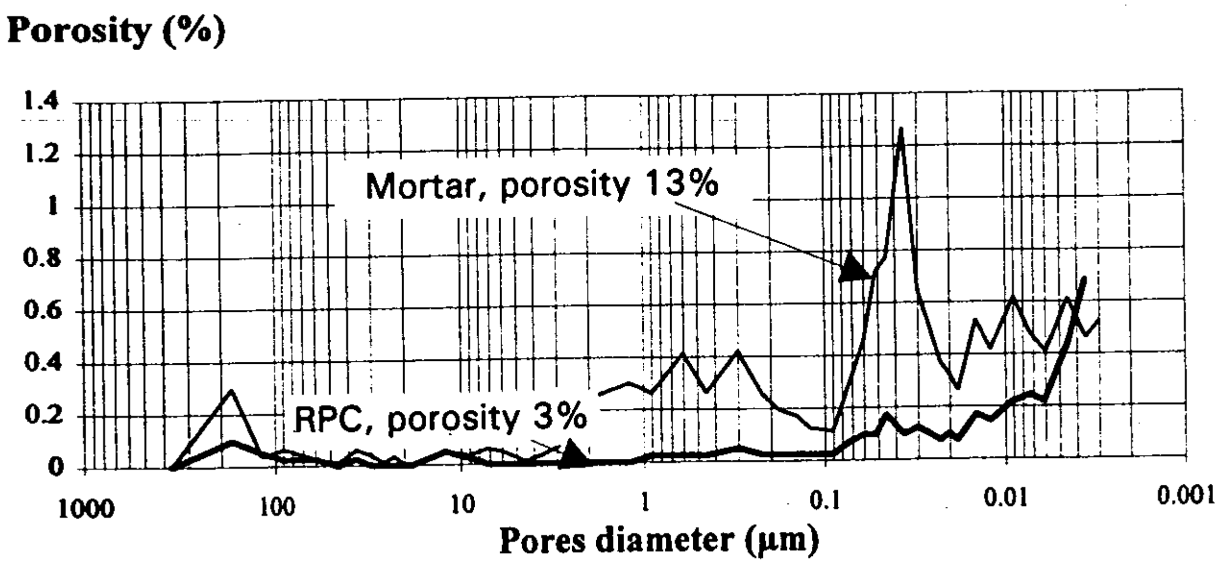

Figure 1: Comparison of the microstructural properties of our RPC and a plain mortar.

3.2 Transfer properties

The transfer properties are, of course, essential factors in radioactive wastes containment. Modelling transfer requires knowing the gas conductivity of the containment material. Similarly, as discussed below, modelling the durability with respect to water leaching needs diffusion coefficient values.

The CEA uses Hassler instruments for measuring gas permeability allowing testing 11-cm diameter, 22-cm high or 4-cm diameter, 8-cm high specimens (Figure 2) [10].

The diffusion measurements are made using tritium (Figure 3) which has the advantage over e.g., Cl^- ions, of a very weak interaction with the matrix [11].

The first permeability results allow us to compare RPC with the mortar cited above. These are given in Table 3 and compared to ANDRA's specification for concretes used for surface storage [12]. We can compare the preliminary results of diffusion with those for different materials tested (Table 4) and the ANDRA specification. It can be seen that RPC transfer properties are, at least, an order of magnitude better than those of HPCs.

3.3 Mechanical behaviour

The problem of RPC behaviour on subjection to shocks is very important for HIC applications. In fact, HIC should resist a 5 m drop, which is far more stringent than surface storage conditions where containers must resist

Table 3 : permeabilities comparison (m^2)

ANDRA specification	RPC after curing at 20°C and 65% RH	mortar after curing at 20°C and 100% RH	mortar after drying at 50°C during 2 months
$< 5.10^{-18}$	10^{-22}	10^{-19}	10^{-17}

Table 4: diffusion coefficients comparison (m^2/s)

ANDRA specification	RPC	HPC	standard concrete
$< 1{,}7\ 10^{-12}$	$\approx 10^{-14}$	$\approx 10^{-13}$	$\approx 10^{-12}$

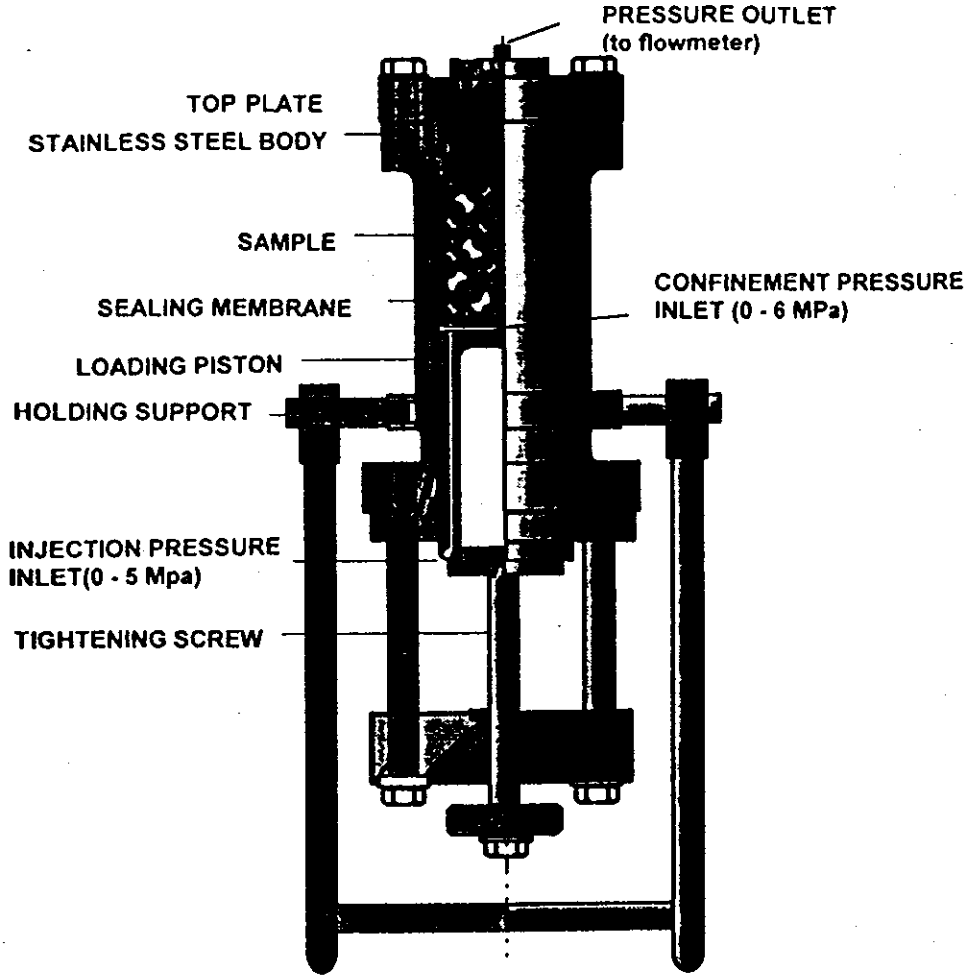

Figure 2 : Schematic view of the apparatus for gas permeability measurement

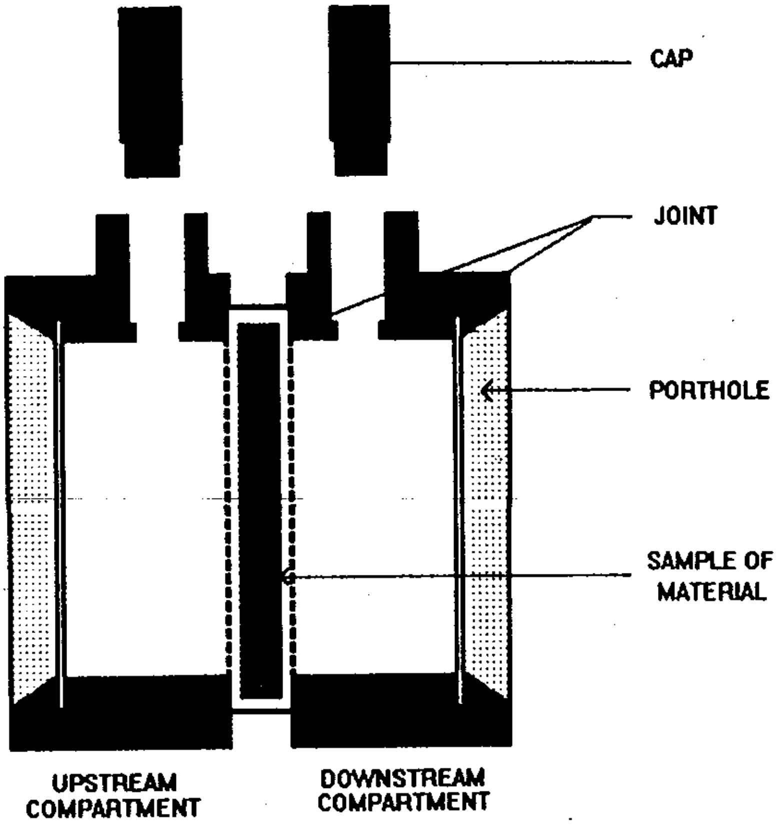

Figure 3 : Diffusion cells

a 1.10 m drop. Ascertaining RPC shock behaviour and its modelling thus become imperative. We therefore have a collaborative research program with the Laboratoire Central des Ponts et Chaussées (LCPC) which has carried out experiments on concrete dynamic behaviour [12]. The results of this work will be published later.

3.4 Durability

To estimate the lifetime of concretes used in storage facilities, the CEA carries out leaching tests where the concrete is degraded by deionised water [13]. Concrete behaviour under these conditions, which are much more severe than those in interim storage or even final disposal, and diffusion coefficient values make it possible to model concrete degradation [14]. RPC leaching studies have just started and will be reported later.

4. CONCLUSIONS

The CEA has adopted a waste reduction policy for the interim storage of their class B radioactive wastes. This has lead to the concept of High Integrity Container (HIC). This new concept is far more stringent than the currently used containers. Namely, reversibility demands the bulk storage of wastes (without inerting them) in the HIC. This means that the HIC will assume alone the role of confinement barrier. This is why, the concept of HIC requires the use of a material exhibiting excellent mechanical, confinement and durability properties. The previous studies concerning RPC showed that it could be a good material to that purpose. That is why it was decided to define a RPC formulation specifically adapted to the demand of the HIC. The evaluation of this formulation regarding mechanical, microstructural and durability properties is currently under study. The first results, hereabove presented, concerning porosity, gas permeability and radioactive ion diffusion are satisfactory.

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REFERENCES

1 - Teillac J. (1988). Les déchets nucléaires. Que Sais-je n°2385

2 - Loi n° 91-1381 du 30 décembre 1991 relative aux recherches sur la gestion des déchets radioactifs. Journal Officiel du mercredi 1er janvier 1992.

3 - Richard P. & Cheyrezy M. (1995). Composition of Reactive Powder Concretes. *Cement and Concrete Research*, vol 25, n 7, pp 1501-1511

4- Cheyrezy M & Maret V & Frouin L (1995). Microstructural analysis of RPC (Reactive Powder Concrete). *Cement and Concrete Research*, vol 25, n 7, pp. 1491-1500

5- Zanni H & Cheyrezy M & Maret V & Philippot S & Nieto P (1996). Investigation of hydration and pozzolanic reaction in reactive powder concrete (RPC) using 29Si NMR. *Cement and Concrete Research*, vol 26, n 1,to be published.

6- Roux N & Andrade C & SanJuan M.A (1996). Experimental study of Reactive Powder Concretes. *Journal of Materials ASCE*, to be published

7- Chaussadent T. & Baroghel-Bouny V. & Gawsevitch J. (1994), Caractérisation texturale des pates de ciment et des bétons durcis, *Journées des Sciences de l'Ingénieur*, Giens.

8- Raoof A. & Guilbaud J.P. & Dupas A. & Nicolas P. (1994). Appareil microgravimétrique de sorption d'eau en continu sous vide d'air. *Journées des Sciences de l'Ingénieur*, Giens.

9- Bourdette B. (1994). Durabilité du mortier; prise en compte des auréoles de transition dans la caractérisation et la modélisation des processus physiques et chimiques d'altération. Thèse de l'INSA de Toulouse.

10- Gallé C. (1994). Relation entre la microstructure et l'aptitude au transfert dynamique des pates de ciment. Note Technique SESD 94.12.

11- Richet C. (1992). Etude de la migration des radioéléments dans les liants hydrauliques - Influence des viellissements des liants sur les mécanismes et la cinétique des transferts. Thèse de l'Université d'Orsay.

12- Toutlemonde F. (1995). Résistance au choc des structures en béton. Thèse de l'ENPC.

13- Adenot F. & Buil M. (1992). Modelling of the corrosion of the cement paste by deionized water. *Cement and Concrete Research*, vol. 22, pp. 489-496.

14- Adenot F. (1992). Durabilité du béton : caractérisation et modélisation des processus physiques et chimiques de dégradation du ciment. Thèse de l'Université d'Orléans.