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# Corrosion des barres d'armature due à la carbonatation dans les bétons armés de charpente pour les dépôts de déchets de faible activité près de la surface - mécanisme de défaillance critique

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by

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# CORROSION DES BARRES D'ARMATURE DUE À LA CARBONATATION DANS LES BÉTONS ARMÉS DE CHARPENTE POUR LES DÉPÔTS DE DÉCHETS DE FAIBLE ACTIVITÉ PRÈS DE LA SURFACE — MÉCANISME DE DÉFAILLANCE CRITIQUE

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## RÉSUMÉ

Le toit en béton d'un dépôt de déchets radioactifs près de la surface constitue la principale protection contre les infiltrations d'eau et les intrusions. On examine ici le mécanisme de rupture éventuelle du toit suivant : le dioxyde de carbone produit par la biodégradation de matières organiques dans le dépôt amorce la corrosion des barres d'armature en acier encastrées dans le toit en béton. Étant donné que la surface inférieure du toit est en grande partie sous tension, elle a tendance à se fissurer. La voie de migration du dioxyde de carbone passe par les fissures du béton entre le fond du plafond et les barres d'armature. La corrosion des barres d'armature due au carbone peut produire un effritement du béton, une corrosion plus prononcée des barres et finalement une défaillance structurelle. On porte une attention particulière à ce mécanisme de défaillance car il a été généralement négligé dans les évaluations du comportement des dépôts.

La documentation pertinente à ce type de défaillance fait l'objet d'un examen. Les conditions requises pour la corrosion des barres d'armature sont la présence de dioxyde de carbone et d'oxygène dans l'atmosphère du dépôt, une humidité relative élevée et des fissures débouchantes dans le béton. On prévoit de fortes concentrations de dioxyde de carbone et une humidité relative élevée dans le dépôt. On y prévoit d'autre part une très faible concentration d'oxygène, et cela devrait freiner la vitesse de corrosion des barres. Il est probable que des fissures se forment à des endroits où les contraintes de traction sont fortes. Le colmatage des fissures pourrait présenter un facteur atténuant, mais, d'après notre analyse, on ne peut guère s'y fier. Afin d'éviter le plus possible que ce mécanisme de défaillance ne se manifeste dans la construction souterraine anti-intrusion (IRUS), un dépôt près de la surface proposé au Canada, le dioxyde de carbone contenu dans l'atmosphère gazeux du dépôt sera absorbé par du béton poreux réactif placé entre les déchets et le toit.

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## ABSTRACT

The concrete roof of a near-surface radioactive waste repository is the principle protection against water infiltration and intrusion. The following potential roof failure mechanism is examined: carbon dioxide generated by the biodegradation of organic materials in the repository initiates corrosion of reinforcing steel embedded in the concrete roof. Because the bottom surface of the roof is mostly under tension, it is susceptible to cracking. The migration path for carbon dioxide is through cracks in the concrete between the bottom of the roof and the reinforcing bars. Carbonate corrosion of the reinforcing bars may result in concrete spalling, more extensive rebar corrosion and ultimately structural failure. Attention is brought to this failure mechanism because it has generally been overlooked in repository performance assessments.

Literature relevant to the above failure is reviewed. Prerequisites for rebar corrosion are the presence of carbon dioxide and oxygen in the repository gas, high relative humidity and through-cracks in the concrete. High carbon dioxide concentrations and relative humidity are expected in the repository. The oxygen concentration in the repository is expected to be very low, and that is expected to minimize rebar corrosion rates. Cracks are likely to form in locations with high tensile stresses. Healing of the cracks could be a mitigating factor, but based on our analysis, it can not be relied on. To minimize the potential of this failure mechanism occurring with the Intrusion Resistant Underground Structure (IRUS), Canada's proposed near-surface repository, carbon dioxide from the repository gas will be absorbed by reactive, porous concrete placed between the waste and the roof.

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# 1. INTRODUCTION

Reinforced concrete is the principle construction material for most radioactive waste repositories sited in the vadose zone. A concrete roof acts as a barrier to both intrusion and infiltration. The natural degradation of this barrier material will ultimately lead to a loss of integrity and the infiltration of water into the repository.

The assessment of degradation processes of reinforced concrete has been the subject of significant R&D, with the objectives being to design durable structures and estimate the rate of degradation. Radioactive wastes placed in near-surface repositories require 300 to 500 years of isolation from the environment, during which time radionuclides such as $^{137}Cs$, $^{90}Sr$, and $^3H$, with half lives $\leq$ 30a, decay to insignificant concentration levels.

The carbon steel reinforcing bars can be one of the weak links in the long-term stability of the concrete structure. As long as the steel is encapsulated in the concrete, it remains in its passive state and hence the corrosion rate is very low. However, the ingress of corrosive agents, by diffusion through the concrete or through cracks, will result in accelerated corrosion of the reinforcing bars and ultimately the failure of the structure. The chemicals most responsible for accelerated corrosion are chlorides and carbon dioxide. Carbon dioxide is generated in the repository by the biodegradation of organic waste material. Carbon dioxide reacts with the alkaline (mostly calcium hydroxide) components of concrete, shifting the pH of the concrete pore water from alkaline, towards neutral, where carbon steel is then in the active corrosion range.

Requirements for the retrievability of waste from near-surface repositories vary with jurisdiction. Where retrievability is not a requirement, the space between the waste packages is generally backfilled with porous concrete. The concrete backfill is reactive with carbon dioxide and thus the repository pore gas is not expected to contain carbon dioxide. Where retrievability is a requirement, usually only small quantities of materials are present that are capable of absorbing the carbon dioxide. In these repositories, a high concentration of carbon dioxide can be present in the repository gas, and hence structural failure initiated by carbonate-induced rebar corrosion is a possibility.

In this paper we examine the potential mechanisms that can lead to an early failure of the repository structure, review the literature and its relevance to the expected environment, and suggest an approach for the removal of carbon dioxide generated in the repository to reduce the potential for rebar failure.

# 2. POSTULATED MECHANISM FOR REBAR FAILURE

The prerequisites for rebar corrosion are the migration of chloride and/or carbonate to the rebar, the migration of oxygen to the rebar, and the presence of moisture on the rebar surface. Papadakis et al. [1] examined the parameters that determine the rate of carbonation of concrete. They found that the rate of carbonation is highly dependent on the relative humidity (RH) of the environment in which the concrete is placed. Carbon dioxide can migrate to the concrete through the gas phase, or as carbonate through the liquid phase. In

dry environments, most of the pores in the concrete are filled with air. Since the rate of the gas phase diffusion is about four orders of magnitude higher than the liquid phase, the carbonation reaction proceeds fastest in low RH environments. Experimental results [1] indicate that the carbonation rates are asymptotic to zero at 100% relative humidity. The RH of repositories placed in humid climates is expected to be very close or equal, to 100%. In such an environment the migration rate of the carbonate front to the rebar is negligibly slow, and the repository concrete structure is not expected to absorb a significant portion of the carbon dioxide generated in the vault. In arid climates carbonation of the concrete is of no concern for rebar corrosion, since the rebar is expected to be dry, and hence corrosion rates will be negligible.

Another, but more important route for carbon dioxide penetration to the rebar is by gas-phase diffusion through fractures in the concrete, when the fractures are dry or only partially filled with water. Concrete fractures occur in regions where the structure is under tension. When the concrete structure is subjected to design loads, mostly micro cracks are formed. Through- cracks, that lead from the surface, through the concrete cover, to the rebar, can also develop in the structure. These through-cracks can be conduits for carbon dioxide and oxygen diffusion from the repository or the outside soil pore gas to the rebar. There are two factors that can mitigate rebar corrosion: the expected low oxygen concentration in repository pore gas, and the potential for the healing of cracks in concrete. Examination of both of these factors follows.

The oxygen concentration in soil pore gas, outside the repository, is not much lower than in air. This gas can penetrate the repository mostly by atmospheric pumping, driven by atmospheric pressure changes. While most of this will take place through the bottom of the repository, such as the drain line leading to the sump, some may occur at joints in the concrete. In the repository, oxygen is scavenged by steel corrosion and the aerobic degradation of organic matter. However, the presence of oxygen inside the repository, in regions where air ingress takes place, can not be ruled out.

The healing of fractured concrete has been widely observed, and has been the subject of several investigations. Guppy [2] noted that only a limited amount of experimental and archival work is available on crack healing. The mechanism of crack healing is not well understood, but the most likely process responsible is calcite formation under wet or high humidity conditions. Investigators found no evidence of amorphous material, such as calcium silicate hydride (CSH) gel formation, in the cracks. The following components are required for crack healing:

- flooded or high humidity conditions;
- a supply of carbonate or carbon dioxide; and
- a supply of portlandite.

Portlandite ($Ca(OH)_2$) in cement is produced by the hydration of Ordinary Portland Cement (OPC). In the newer, blended cements generally used for repository construction, a portion of the OPC is replaced by Blast Furnace Slag (BFS) and/or Pulverised Fly Ash (PFA), resulting in reduced portlandite production that is partly due to the reduced OPC content, and partly due to the pozzolanic reaction between portlandite and BFS/PFA. We were unable to locate literature on the crack healing properties of these advanced cement compositions.

The carbon dioxide concentration in air is approximately 0.03 vol.-%. Most of the experience in crack healing is with air containing ambient carbon dioxide concentrations, or water in equilibrium with ambient air. The carbon dioxide concentration in the repository gas is expected to be at least several percent, which is two to three orders of magnitude higher than ambient.

For crack healing to take place, calcite must form within the crack. The carbon dioxide (or carbonate) supply to the crack controls the reaction rate. Under those circumstances, there is ample supply of portlandite in the concrete pore water adjacent to the crack. When there is a large concentration of carbonate ion in the pore water, and limited portlandite supply, calcite formation in the cement matrix is favoured rather than in the crack. The carbonation front advances into the concrete structure. This does not lead to crack healing. Brodersen et al. [3] assessed crack healing in flooded conditions, and advanced a similar argument to explain why cracks only heal for a short time after their formation. Thus the concrete used for repository construction and the repository environment are different in at least two aspects from those associated with in current field experience, and both differences disfavour crack healing. There is a need for research in crack healing, where both the repository-concrete and repository-environment are simulated.

## 3.    LOCATION AND CONSEQUENCES OF REBAR FAILURE

The physical integrity of the repository concrete roof is essential for prevention of both infiltration and intrusion. The function of the walls is less critical, as their primary role is to provide stable support to the roof. Only extensive deterioration of the walls will compromise the integrity of the roof. Tensile stresses are expected to be highest in the roof, since it has to carry its own weight, the weight of the water shedding layers and the soil cover. Thus premature roof failure has been the focus of our analysis for the Intrusion Resistant Underground Structure (IRUS) planned for the Chalk River site. While we expect to have significant quantities of chloride ions present in the repository pore water, chloride initiated corrosion of the rebar in the roof is not considered to be an issue. The repository roof will be supported by the walls, but it will be poured over repository contents covered by sand and porous concrete. With time the contents of the repository are expected to subside, creating an empty space between the waste and the roof, that will form a diffusion barrier for the salts leached from the waste. Our concern with carbonate induced rebar corrosion is due to the following factors:

- The formation of cracks in the roof and the ingress of carbon dioxide into the cracks can start soon after the repository roof is poured.

- Once corrosion is initiated, the deterioration of the structure is a progressive process. As corrosion proceeds, the expansion of the corrosion film can apply pressure on the concrete surrounding it, resulting in the widening of the fracture, and the eventual spalling of the concrete cover [4]. This process then exposes more rebar surface area to the corrosive environment.

## 4.    PREVENTATIVE MEASURES

While the projected low oxygen concentrations in the repository provide some assurance of low corrosion rates of rebar exposed to a high carbonate environment, an additional preventative design feature was deemed desirable. A layer of porous concrete with a high surface area will be placed between the waste and the repository roof. The portlandite and other basic components of the porous concrete will react with, and hence remove, the carbon dioxide generated in the waste, and thus minimize the carbon dioxide concentration at the inner surface of the roof. The composition of the porous concrete has not been finalized yet, but it will have sufficient capacity to remove carbon dioxide generated during the first 500 years following repository closure.

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