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DURABILITY ISSUES FOR CONCRETE IN A DEEP GEOLOGIC REPOSITORY

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ABSTRACT

Concrete and cement-based materials are being considered in the design for sealing components in emplacement rooms, access tunnels, shafts and ramps and boreholes for deep geologic repositories. Cement-based materials are being considered as a grout material to isolate hydraulically active fractures and to fill interfaces between concrete and rock. Concrete may be used as a working surface floor in emplacement rooms and in access tunnels and as large structural seal components in composite seal systems. In some applications concrete may be used as a tunnel support in the form of shotcrete or rock bolt grout. Because of the potential for their wide use in various applications and locations, concrete and cement-based materials in a repository would experience different conditions at different times. These conditions would be related to the degree of saturation of the surrounding materials, the groundwater composition and movement, the rock type and character, and the extent of its exposure to either air or heat from containers. This paper reviews the potential mechanisms that could affect durability of concretes and grouts used in a repository.

I. CONCRETE USE IN A REPOSITORY

Concrete has been used as a building material for centuries. In its simplest modern form, concrete is a mixture of paste and aggregates. The paste, composed of Portland cement and water, coats the surface of the fine and coarse aggregates and binds them together. Pozzolanic materials (Silica fume, fly ash, etc.) can be used to replace all or portions of the Portland cement to modify properties including curing temperature and permeability. The percentage and types of components, such as Portland cement, silica fume, fly ash, silica flour, as well as coarse and fine aggregate, all influence durability. The physical and chemical behaviour of concrete is complex as it is dependant on many factors including the materials used as the cement, cement size, water content, aggregate size and type, the addition of chemicals, the relative proportions, the amount and quality of mixing, placement, curing, and environment of exposure (air, groundwater, thermal cycling). The final placed concrete product depends on the skill and quality of the production, the skill of the workers placing the concrete and the curing conditions.

Concrete is being considered for use in the engineered barrier systems of deep geologic repositories for used nuclear fuel by countries including: Canada, France, Finland, Japan, Sweden, Switzerland and the United States of America. In Canada, the sealing system designs presented in Atomic Energy of Canada Limited (AECL)'s Environmental Impact Statement (EIS)[1], a second case study (SCS)[2], the Third Case Study (TCS)[3] and the horizontal borehole emplacement concept[4] all call for the use of concrete.

Concrete and cement-based materials used in sealing systems may be emplaced during the construction, operation or closure of the repository. During construction, concrete may be used for tunnel floors, for tunnel support, and for service areas. Cement-based materials may be used in grouting to isolate hydraulically active zones. During operation, concrete floors may be placed in emplacement rooms and concrete seal components may be placed at the entrance to those rooms, and these seals may include cement-based grouting. The ability of the concrete floors without reinforcement to withstand the expected loads of the current reference used-fuel containers has yet to be assessed. The mechanical performance of concrete in a repository could be enhanced through the use of reinforcing materials, such as steel rebar or non-metallic fibre. During closure some concrete could be removed from floors and service areas floors. Concrete could be used to seal access tunnels, ramps and shafts and to cap the accesses to repositories at that time. Potential locations for the use of concrete in a repository are illustrated in Figure 1. The amount of concrete would be large and therefore its durability and impact on repository sealing systems must be considered.

II. REPOSITORY NEAR FIELD ENVIRONMENT AND EVOLUTION

In terms of this discussion, a deep geologic repository will experience two phases of evolution, preclosure and postclosure.

During the preclosure phase, which may last up to 100 years[5]:

- the shaft/ramp, access tunnels and emplacement rooms are excavated and required services are installed;
- emplacement rooms are filled and sealed;
- monitoring is conducted;
- the near-field rock surrounding the excavations desaturates;
- in most excavations (other than sealed rooms) a normal oxygen atmosphere exists;
- thermal transient conditions occur, temperatures are approximately 85°C near used fuel containers with peak temperatures occurring 15-30 years after emplacement of used fuel and the thermal plume moving out from the containers, within 100 years only the rock in the immediate vicinity of the repository would be heated[6]; and
- seal areas may reach various stages of resaturation.

During the postclosure phase:

- all repository excavations are sealed;
- oxygen is depleted (a reducing environment develops);
- after 1,000 years the rock within several 100 m of the repository would experience elevated temperatures, after 10,000 years the thermal plume would reach its greatest extent followed by a slow decline to near ambient temperatures by about 100,000 years[6];
- the repository excavations and near-field rock resaturate and repressurize at varying rates; and
- seals at or near the surface may undergo freeze thaw cycles.

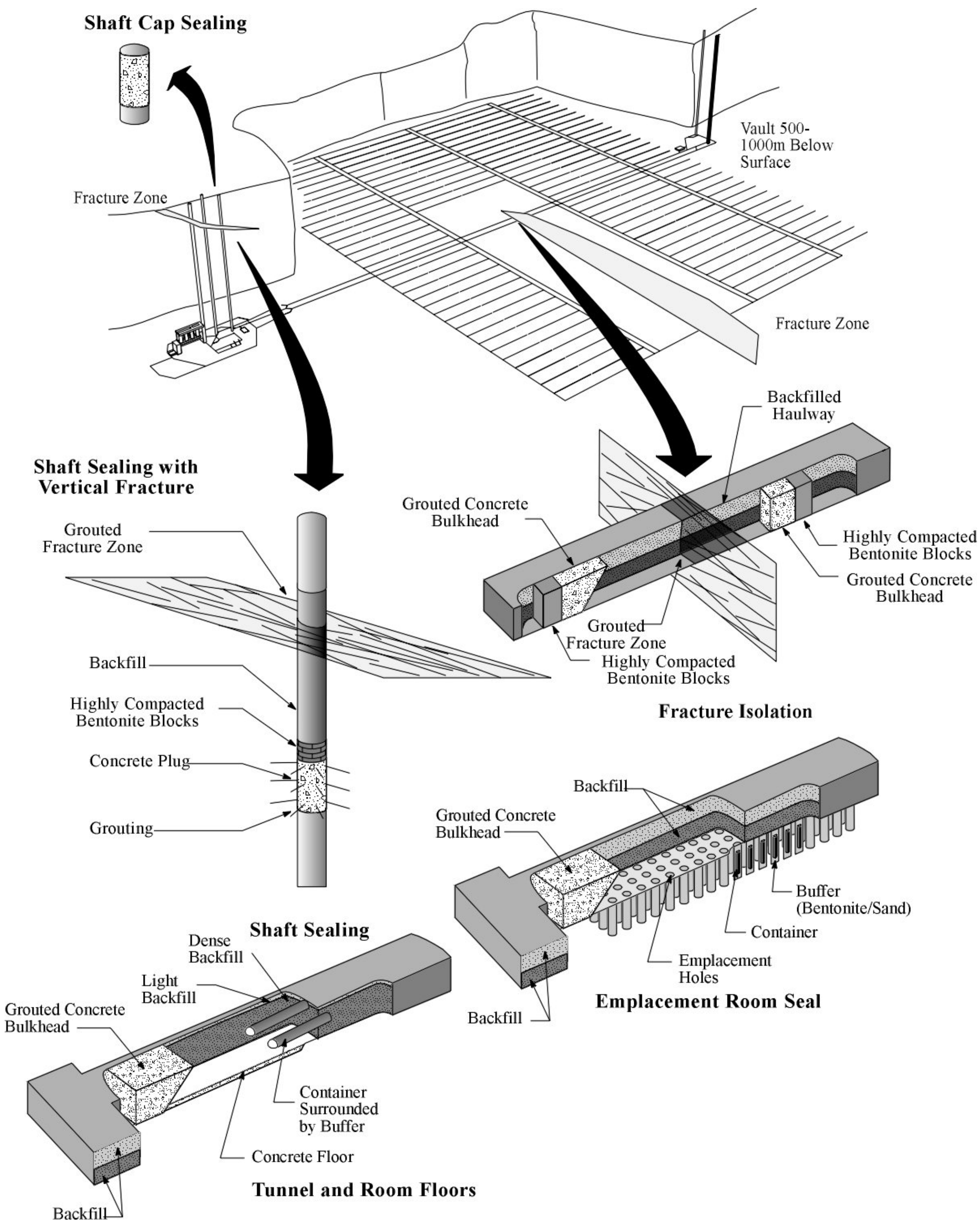

Figure 1: Potential uses of concrete in a deep geologic repository.

Concrete and cement-based materials at any location in the repository will go through a cycle that includes: heating and drying, cooling and saturation, and change from an oxygen-rich atmosphere to a reducing environment[7]. The timing of these changes, the rate at which they occur, and their magnitude will vary depending on the location of the concrete in the repository (e.g., cement-based grout isolating hydraulically active fractures would likely be continuously saturated). Thus, concretes in or near emplacement rooms may reach higher temperatures and drier conditions than other locations as they are nearer the heat-generating containers. This effect will be influenced by local hydraulic conditions, including proximity to water bearing fractures, recharge rate and rock mass permeability. Humidity and contact with groundwater will gradually increase as the local temperature cools. Over a long period concrete will progress from partially to fully-saturated, anaerobic conditions. The time to reach anaerobic conditions will depend on the rate that local materials interact with and consume free oxygen. The groundwater chemistry (dissolved chlorides and sulphates) will have an impact on concrete durability. When considering the durability of the concrete these variable factors must be taken into account. The condition of the concrete at the end of one phase of the repository will be the condition of the concrete at the beginning of the next phase in terms of considering the ultimate durability.

For civil engineering design, durability has in the past been tied to the strength, i.e., the stronger the concrete the more durable it was considered. With aging infrastructure including concrete, it has become apparent that durability is more than just strength and other factors must be considered at the concrete mix design stage. Durability is becoming recognized as the ability of concrete to resist the effects and influences of the environment while performing its desired function[8]. Neville[9] states in order for a concrete to be durable it must be able withstand the processes of deterioration to which it can be expected to be exposed, but cautions that durability does not mean an infinite life, nor does it mean withstanding any action on concrete. In a repository, the concrete must maintain a low hydraulic conductivity both intrinsically and at interfaces with the rock[10]. In fact, the primary consideration for durability is having a concrete with low permeability[11,12]. The low permeability reduces the rate of penetration into the concrete of potentially detrimental chemicals carried by groundwater.

III. DESIGN REQUIREMENTS FOR REPOSITORY SEALING SYSTEMS

In the current Canadian repository design, composite seals that include concrete and swelling-clay based components must function for at least 100,000 years. However, the structural role of the concrete element and cement-based grouts, may be fulfilled when the swelling clay components are fully saturated and exerting swelling pressure in the adjacent materials. At this time, the concrete would cease to be a significant structural component and would be considered a filler material, occupying a volume in a composite seal. In the long term, the volume occupied by the concrete filler must be equal to or larger than the volume occupied by the concrete structural element.

In the preclosure phase, concrete and cement-based materials should contribute to sealing systems that must:

- provide restraint for other sealing system materials,
- remain as an intact mass without volume loss under thermal-mechanical loads,
- remain as an intact mass without strength loss in the repository chemical environments,
- protect other sealing materials from volume change,
- provide access control and safeguards systems seals,

- provide a stable working surface when used as a floor, and
- contribute to minimizing water movement and contaminant transport in the excavations, in the excavation damage zone in the near-field rock mass, or to/from hydraulically active fractures intersecting sealed excavations.

Concrete and cement-based materials with proper formulation, preparation, placement and curing would not experience significant degradation in the preclosure period. Therefore concrete would fulfill preclosure functions. This conclusion can be based on general literature about degradation of concrete as many conditions in a preclosure repository will be similar to those encountered in existing concrete installations.

In the postclosure phase, concrete and cement-based materials should contribute to repository sealing systems that must:

- provide restraint for sealing system fill materials,
- remain as an intact mass under thermal-mechanical loads and in the repository chemical environment until the remaining components of the sealing system have saturated and developed their final swelling pressure,
- retain or increase their as-placed volume as they degrade following saturation and swelling of the clay components of the sealing system,
- isolate hydraulic activity from other repository sealing system components,
- contribute to minimizing water movement and contaminant transport in the excavations, in the damage zone in the near rock mass, or to/from hydraulically active fractures intersecting an excavation, and
- provide security/safeguards seal at surface entry points to a repository.

There is less direct evidence in literature to provide information on the durability of concrete in the postclosure period. The concrete will be affected by its environment and an understanding of the mechanisms that can affect the integrity of concrete is a useful starting point.

IV. MIX DESIGN AND CURING

Mix design and preparation are very important for providing a durable concrete. The basic composition of modern concrete is relatively simple. Concrete can be considered to be basically a mixture of two components, aggregates and paste[13]. The aggregates normally comprise sand and gravel or crushed stone. Selecting the aggregate material for concrete is very important[11]. A mismatch in the elastic moduli or in the coefficients of thermal expansion between the cement paste and aggregate can result in the formation of microcracks in the concrete. An aggregate that reacts chemically with the cement paste can damage concrete (Section V). The paste of cementing materials is typically Portland cement, with or without supplementary sealing materials, plus water and air. Portland cement, the fundamental ingredient in concrete, is a calcium silicate based material made with a combination of calcium, silicon, aluminum, and iron.

Water is required to hydrate the dry cement compounds. The amount of water strongly influences the strength of concrete. High water content dilutes the cement paste, increasing concrete volume and reducing density. It is important to note however that sufficient water must be in the mix for the concrete to properly hydrate. Therefore a balance in the mix design must be developed for the water to cementious materials ratio in order to ensure there is sufficient water to hydrate the materials but not an excessive amount that would weaken or increase porosity in the cured concrete. By achieving this balance in the water to cementious materials ratio, the following advantages can be gained.

- Increased compressive and flexural strength,
- Lower permeability,
- Increased resistance to weathering,
- Better bond between successive layers and to reinforcement,
- Reduced drying shrinkage and cracking, and
- Less volume change from wetting and drying.

When designing concrete for use in a repository, it must be recognized that it will be in contact with clay-based sealing materials. The general concern with concrete-clay interactions is the effect of concrete degradation products and the elevated pH of concrete leachates on reducing the swelling properties of clays. The generation of high pH leachates affects clay minerals, which reduce the swelling potential of clays[14]. To maintain clay swelling ability, the concrete mix must be designed to have a pH less than 11. This can be achieved by lowering the alkalinity of the concrete by substituting pozzolans for Portland cement. AECL's Low-Heat High-Performance Concrete (LHHPC)[15] has a pH in the range of 9 to 10. While this is higher than the pH of a clay system (pH~8) and thus still has the potential for long-term alteration of the clay,[14] it is much closer in pH value to the clay than a regular concrete. Therefore it is considered a more suitable candidate for use in a repository.

One concrete mix design may not be suitable for all repository applications. For example, the concrete in an emplacement room seal would require a different formulation than for a shaft cap seal. The concrete mix design must be prepared carefully with quality control applied to materials. Even if the mix is designed correctly, using poor quality or incorrect materials can defeat the design. The source for materials specified in mix design must be used in the production or substitutions must be tested to ensure they meet design criteria. Following standard practices, and understanding mixing, pumping and consolidation issues can assist in achieving proper placement.

Similarly, curing is important for production of a strong and durable product. Controlled curing is vital to produce quality concrete particularly in the areas of durability, strength, permeability, resistance to abrasion, freeze thaw damage and sulphate attack[16]. In most repository seal applications, a massive pour of concrete is presumed in the designs. Curing is an exothermic reaction and the heat generated from the hydration can lead to cracking, especially in massive structures. Replacement of Portland cement by pozzolanic materials will decrease the curing temperature of the concrete[15]. Proper curing of concrete without cracking or with only minor cracking is needed to produce a durable, low permeability structure. While post-curing grouting is a feasible method of sealing cracks[17] minimizing crack formation by decreasing the thermal flux during curing is more effective. This can be accomplished in large pours through the use of techniques to reduce temperature that include pre-cooled concrete and use of a low-heat mix design. It is important to supply water to concrete as it cures, as hydration ceases if the relative humidity drops below 80%[9,13] and that this can lead to large increases in pore structure[18]. This will increase permeability and decrease durability. Higher ambient temperatures can also increase the risk of drying and increase water demand[9].

At the operational level, procedures that specify mix design, preparation, placement methods, curing requirements and environmental controls should be developed. Qualified personnel should supervise each of these activities.

V. PROCESSES AFFECTING THE DURABILITY OF CONCRETE AND CEMENT-BASED MATERIALS

The evolution of the environment at each location where concrete is used in a repository will be different and therefore will have different mechanisms or rates of degradation of concrete[19]. When considering the potential for durability of a concrete installation, the processes should be treated as a sequence of degradation mechanisms[8]. Potential mechanisms for the degradation of concrete can be loosely grouped into physical and chemical modes.

Chemical degradation mechanisms are as follows.

- Alkali aggregate reactivity is a reaction between active mineral constituents of some aggregates and cement paste. There are two forms: one is alkali silica reactivity (the more common), and the other is alkali carbonate reactivity. These reactions can cause swelling and cracking of the concrete.
- Carbonation results when carbon dioxide in the air penetrates the concrete and reacts with moisture to form carbonic acid[9]. The acid reacts with the hydroxides in the concrete (calcium hydroxide) to form carbonates. This reaction results in a slight shrinkage of the surface paste of the concrete. Rapid drying and carbonation can affect surface durability but this can be minimized by proper curing. If steel reinforcement is present, the cracking caused by carbonation can act to expose the steel, which would then be subject to corrosion.
- Sulphate attack occurs when sulphates in solution react with hydrated compounds in the hardened cement paste. The reactions can induce sufficient pressure from volume expansion to disrupt the cement paste, which will result in the disintegration of the concrete. The consequences of sulphate attack include not only expansion and cracking but also the loss of cohesion in the hydrated cement paste and the attendant loss of adhesion between the paste and aggregate.
- Delayed ettringite formation is a mode of sulphate attack involving sulphate already present in the concrete at the time of placement. The exact mechanism is still debated but it involves the mineral ettringite, which forms from hydrated cement compounds, fills a larger volume than the original materials and thus induces a swelling pressure, which can induce cracks in the concrete.
- Chloride attack involves penetration of the concrete by chloride ions, which can cause corrosion of reinforcing steel. The corrosion product expands up to four times the volume of the uncorroded steel, which can induce cracks in the concrete.
- Leaching requires percolation of water through the concrete that is deficient in those ions that may be leached from the concrete. The process requires a supply of moving water as stagnant water would become saturated in leachable ions and the process would cease. Leaching can remove components of the cement paste, weakening the strength and increasing the permeability of the concrete.

Physical degradation mechanisms are as follows.

- Freeze thaw attack occurs when water enters existing voids, fractures or susceptible aggregate and freezes. The ice occupies a volume 9% greater than the water exerting swelling pressure on the surrounding concrete, which could extend existing cracks and create new ones. This would only occur in concrete installed in repository entry caps in surface environments that include freezing conditions.

- Abrasion is wear caused by equipment movement on floors of repository tunnels or glaciers moving across surface caps. Abrasion can leave areas on a concrete surface more susceptible to chemical or freeze thaw attack.

The significant mechanisms that will degrade concrete inside the repository are chemical. Most available literature discusses chemical attacks with unrestricted flow and continual supply of attacking chemical chemicals. As a repository would be designed to limit water movement, hence the potential rate of movement of environmental contaminants, the availability of chemicals that would degrade concrete attack should be much lower than for surface installations.

VI. DISCUSSION

The designs of the engineered barriers and the potential equipment, facilities and ground support needed to operate a repository would likely include the use of concrete and cement-based materials. In a repository context, the durability of concrete or cement-based grout is the ability of the concrete or grout to satisfy its functional and performance requirements under the initial and evolving environmental conditions at the that location. Concrete durability can be controlled to some extent by the selection of components and the design of the mix. It can also be enhanced by proper placement and curing techniques.

The use of concrete in repository sealing systems is influenced by the functional requirements, the evolving repository environment and the degradation processes that could be active at a potential installation location. The function requirements for concrete will be generally the same for all installations but there may be differences in preclosure and postclosure requirements. The initial environmental conditions for each concrete installation will vary depending on the repository site conditions at a given location and will evolve through the preclosure and postclosure phases until the temperature and groundwater conditions return to a steady state. Temperature will remain elevated for thousands of years with temperatures approaching ambient values (17°C) after 100,000 years[6]. Peak temperatures will be dependant on the location in the repository[20] but can be over 70°C from 100 to 3000 years at the centre of the repository[6]. Chemical degradation processes will operate at all concrete installations, but physical degradation processes will only occur for near-surface installations and for floors of underground excavations before they are filled. It is clear that the conditions effecting a given concrete emplacement are complex and all concrete in a repository must be considered based on local environments[21].

The concrete used in each application may require a different mix design developed to suit the functional requirements and expected environmental conditions. Selection of concrete component materials and mix design must therefore be considered with the same care as in selecting and developing other materials for use in a repository. An example where mix design is important is to reduce the effect of the concrete on the swelling ability of adjacent clay. Leachates from regular Portland cement have been shown to inhibit swelling in bentonite due to high alkalinity. Concretes, such as LHHPC[15] have been developed with a significantly lower alkalinity than regular concrete and thus less concern for impact on the swelling ability of clay[14].

For concrete to be used as part of a sealing system, repository planners must have assurance that the concrete will have sufficient integrity to perform its designed role. Concrete has been shown to last for hundreds and even thousands of years when installed in less than perfect conditions[22,23,24,25]. It may be required to perform over a much longer period of time in a repository. Coons and Alcorn[26] suggest that this durability is possible but keeping permeability low will be of greatest importance[10]. Once clay materials are fully saturated and exerting a swelling pressure, the concrete would not have to provide continuing structural support. It would then act as a stable fill to prevent volume expansion and loss of swelling pressure in the clay.

The weight of evidence in the literature indicates the physical and chemical composition of concrete and its immediate environment have the greatest influence on the durability of concrete[27]. The use of concrete as a restraint for other sealing repository materials can be reasonably justified for the preclosure phase. Only a few millimetres of degradation are expected in 100 years from sulphate and chloride attack[5] and unless there is a mechanism that will remove weakened concrete, there would not be a volume loss.

In the longer term, after closure of the repository, the slow water movement expected would make dissolution and removal of significant volumes of material unlikely. There are some studies[22,23,24,25,26] that suggest that concrete can survive for a long period of time in an environment that has similarities to a repository environment. Most of these studies deal with chemical changes brought about by concrete degradation and do not state that a significant loss of material volume can be expected. The probability of volume loss is deemed to be low, since the paste material would have to be degraded and transported in large quantities to free the aggregate and this is unlikely given the low flow conditions expected in a repository. The aggregate could not be transported from a closed repository even if the paste material could be removed. The effect of the elevated temperature on the degradation processes must be determined.

Full understanding of the durability and longevity of concrete and cement-based grouts repository materials can only be reached from a combination of complementary field, laboratory and natural analog studies[25]. Mehta[12] states that accelerated laboratory tests do not correlate well with the behaviour of concrete structures in practice. In order to develop appropriate concrete materials and mixes and to ensure that they are sufficiently long-lived and durable for use in a repository, long-term research on the *in situ* performance of concretes needs to be started to allow long time periods for experiments. Atkinson and Hearne[28] indicate that if multiple degradation processes are expected to act on a mass of concrete, the effects may be cumulative and accelerate the rate of degradation. Mehta[21] states that degradation processes must be viewed holistically. No concrete will perform perfectly in all conditions, but proper mix design, preparation, placement and curing can help to ensure that the concrete used will meet its functional requirements as part of the repository sealing systems.

VII. SUMMARY

The use of concrete in a repository has been assumed in many conceptual repository designs. Concrete is proposed for components of repository seals to be placed at the entrance to filled emplacement rooms and at other strategic locations throughout the repository and as floors for various rooms, tunnels and service areas. Cement-based materials are being considered as a grout material to isolate hydraulically active fractures and to fill interfaces between concrete and rock. The amount of concrete and cement-based materials involved in a repository would be large. The durability of these materials as they relate to the effect on the repository chemistry and ability to remain as an intact mass in part of a sealing system are important for the performance of repository sealing systems.

In order to produce a durable concrete, it must be designed, prepared and placed with a key design goal of producing a low permeability material. At the operational level, procedures that specify curing, placement methods and environmental controls should be developed. Qualified personnel should supervise these activities. Poor curing techniques can undo a good mix design by causing cracking within the concrete. The concrete mix, preparation, placement and curing specifications should be application specific because the requirements for and environmental conditions affecting each concrete application could differ. While a large body of literature exists on the topic of concrete durability under general conditions and while some of the information is applicable to conditions that may be expected for concrete used in a repository, only a limited amount of work has been done specifically for concrete to be used in an actual repository.

A number of degradation mechanisms that may occur in a repository setting have been identified. These include chemical degradation mechanisms; alkali aggregate reactivity, carbonation, chloride attack, sulphate attack, interaction with other repository materials, leaching and physical degradation mechanisms; abrasion, and freeze thaw cycling. Interaction with other repository materials can also influence the longevity of concrete. The processes need to be viewed holistically when determining the potential durability of a concrete.

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