500 YEAR CONCRETE FOR A RADIOACTIVE WASTE REPOSITORY

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ABSTRACT

The IRUS repository planned at Chalk River for the belowground disposal of low level radioactive waste relies on the durability of concrete for the required 500 year service life. A research program for the IRUS repository to design a durable concrete and also to predict its longevity under the repository environment is in progress. The methodology involves the identification of major degradation agents, and the assessment of the rate of diffusion of corrosive ions and/or the rate of advancement of the reaction front into the concrete. Accelerated test methods are being used on laboratory specimens in conjunction with extrapolation procedures to predict long-term durability from short-term data. The inherent limitations are also examined.

INTRODUCTION

A program is underway at Chalk River Nuclear Laboratories to construct and commission a prototype belowground repository labelled IRUS (Intrusion Resistant Underground Structure), for the disposal of low-level radioactive waste. The disposal concept meant to endure for a service life of 500 years or more relies on the following engineered barriers (1).

Physical - Waste form
- Disposal containers
- Concrete vault and roof

Chemical - Buffer material
- Buffer backfill

Figure 1 shows a cross section of the IRUS disposal facility after closure. The facility will be located in a free-draining sand dune, with the foundation of the repository placed a meter above the maximum recorded groundwater table. A plan of the facility during the operational phase is shown in Fig. 2. During the operation, the repository will be covered by a temporary weathershield building and served by a gantry crane. The bottom of the facility is designed to be permeable to allow any water that might enter the facility to drain freely rather than to remain in contact with the waste. Walls of the repository will be made of 0.6 meter thick reinforced concrete and the roof will have a minimum thickness of one meter. As additional barriers to water infiltration, the reinforced concrete roof will be provided with a polyethylene sheet, 0.3 meter gravel layer, 0.6 meter of fine consolidated sand and one meter of top soil. The additional cover materials will place the repository concrete below the frost level. The ground will be graded to encourage surface water to run off to the sides and away from the repository.

The curved shape of the repository walls provides the following advantages:

- creates compressive stresses in the walls from external loads, thus minimizes the potential shrinkage cracks
- reduces the need for reinforcement bars, thereby reducing the corrosion of reinforcement bars, a potential cause of concrete failure
- reduces construction costs by efficient use of materials.

CONCRETE DURABILITY

Since the vault and roof are major components of the engineered barriers, the durability of concrete is an important aspect for the integrity of the facility.

To endure an engineered service life of 500 years, the repository concrete must undergo a very slow rate of deterioration. The durability will be influenced by the following three major aspects:

- service environment to which the concrete is exposed
- quality of the various ingredients selected for the formulation and their proportions in the mix design
- internal stresses from normal service loading and abnormal events such as earthquake or soil liquefaction.

Chemical Environment

The chemical environment inside the repository will be different from that of the outside (2). The composition of the water inside the repository is controlled by the following:

- leaching of the waste forms
- carbon dioxide generated by the biological processes
- organic compounds such as fatty acids generated by thebiological degradation processes

- interaction of constituents of water with buffer and backfill materials.

To assess the durability and service life of concrete, it is necessary to consider the various degradation mechanisms, especially those that are present in the service environment, both now and during the long post-closure period.

Degradation Mechanisms

The following are some of the degradation mechanisms of concrete:

a) Internal Causes

- alkali-aggregate reaction
- differential thermal expansion of cement and aggregates
- internal stresses from external loading, effects of creep and shrinkage
- gel crystallization

b) External Environment

- chemical deterioration by chloride attack, sulphate attack, carbonic acid corrosion by CO_2, acidic corrosion by organic acids
- freeze-thaw scaling
- leaching and dissolving of lime
- actions of micro-organisms.

Of the internal reactions, gel crystallization will not result in greatly inferior properties while continued hydration and increase in strength has beneficial effects (8). The internal stresses from external loading and the shrinkage and creep effects can be kept within the allowable limits by proper structural design and construction methods. More important, and therefore included in the study, is the reaction between cement and concrete aggregates and their compatibility in thermal expansion. The corrosion can be severe if the materials are not well selected.

The chemicals present in groundwater, and the chemicals leached from the disposed waste into the groundwater, can react with the cement paste of the repository concrete *and* cause deterioration. Sulphate and chloride attack can cause severe degradation of concrete. Also, leaching of the free calcium by the groundwater can reduce the structural strength (see Research Program on Concrete Durability).Under the Canadian climatic condition, the frost action is an important cause of concrete deterioration. When concrete is cooled below zero degree centigrade, water in small pores migrates to larger pores where it freezes. This finally results in a situation similar to ice lensing in soils, and the resulting high internal pressure causes cracking and spalling of concrete (10). The freezing and thawing cycles relevant to IRUS are those during the construction and operation only, since the repository *will* be protected from freezing by the soil cover during the post-closure period. Actions by micro-organisms produce acid, which attacks the concrete dissolving calcium hydroxide. The amount of corrosion to some extent will depend on the availability of oxygen because, in most cases, the micro-organisms are of the aerobic type (4).

Figure 3 shows a possible failure tree for the repository concrete.

Quality of Materials and Concrete Formulation

A denser concrete will have less air voids in it and hence will be less permeable. Reduced permeability will increase the resistance of concrete to aggressive chemicals (by reduced ion diffusion) and lower the leaching of calcium hydroxide by groundwater. The addition of supplementary materials such as blast furnace slag, fly ash and silica fume to the cement has been reported to reduce the permeability of concrete (11).

It is important that all materials selected for the repository concrete should be durable and compatible with each other. The coarse and fine aggregates will be tested for alkali-aggregate reactivity and other code requirements. Any performance additives and admixtures used in the concrete mixture will also be tested for their long- term durability and compatibility with the other ingredients.

Structural Design and Construction Techniques

The structural analysis will consider all externally imposed loads and internally induced loads such as thermal stresses and shrinkage of concrete. The design procedure will proportion the member sizes to keep the internal stresses within the allowable limits. Proper construction techniques and quality assurance procedures are as important as design requirements and will have to be properly applied.

FAILURE CRITERIA

It is difficult to establish exactly what degree of corrosion will constitute failure of the repository concrete. One way to establish the failure criteria is to look at the structural and safety requirements of the concrete and violation of any of the major requirements can be construed as a failure. The concrete must retain its strength to support the imposed loads and should keep its physical integrity to minimize water infiltration and to discourage any potential inadvertent intrusion by humans into the repository. Infiltration of some water into the IRUS vault will not constitute a failure of the disposal concept since the floor is made of permeable and adsorbing material that allows drainage and restricts the migration of mobile radionuclides. However, in order to evaluate durability, it is necessary to define failure of the repository concrete. Potential failure conditions are:

Reduction of structural strength

Since strength is required for the structural integrity of the member, a reduction in concrete strength of over 50% can be considered as failure. Leaching of 33%of the free calcium hydroxide by the groundwater would reduce the strength by 50% (4).

Penetration of ions to the mid- point of structural element

Once the reaction front (the chemical reaction of aggressive agents with the cement paste) has gone to the middle of the structural element, the affected concrete would have suffered severe corrosion and hence the structural and physical integrity is greatly compromised.

Penetration of enough chloride ions to diffuse into the concrete to react with the reinforcing bars and start the corrosion process

In most cases, the permeation of corrosive agents from the solid-liquid interface of the concrete controls the rate of deterioration. Hence, the failure criteria must consider the diffusion rate of the corrosive agents through the host media (the environment around the repository concrete), and the quality of concrete on the rate of advancement of the reaction front. The relationship between the induced stresses in concrete and generation of corrosion products around the reinforcing steel is not known. However, if we can assume a percentage of the corrosion of reinforcement that would cause cracking or failure of concrete, for example 25%, then the time required can be evaluated.

RESEARCH PROGRAM ON CONCRETE DURABILITY

Concrete durability depends on the quality of the concrete and also on the service environment, and hence is site-dependent. AECL has identified the various potential aggressive elements for the IRUS repository concrete at Chalk River. The Institute for Research in Construction, National Research Council of Canada (NRCC), with international expertise in concrete research, has established a research program at AECL's request to evaluate the durability of different qualities of concrete under this service environment. The research program was presented to a team of international experts in concrete research, for peer review, in a one-day working session in October 1987. The presentation was well received by the experts and their helpful suggestions for improvement are incorporated in the program.

Research Program Objectives

1) To design a durable concrete for IRUS and to establish short- term corrosion rates under extreme conditions.

The intended duration for this research work is 18 months.

2) To continue the study of the deterioration of concrete, and to predict the useful engineered lifetime. Also to generate experimental data for generic studies so that a durable concrete can be designed for other repository service conditions. The intended duration of this program is 5 to 6 years.

Methodology

If the rate of deterioration of a quality concrete under various aggressive elements is known, then it should be possible to design and to estimate the longevity of that concrete in a particular environment. To do that for periods of over 500 years, extensive data must be obtained to establish the rate of degradation of concrete under various exposure conditions. A thorough analysis of this data has to be done before a full methodology can be worked out (3). Although there is a high degree of uncertainty as to what deleterious conditions, and what range of environments the concrete will be exposed to in the next 500 years, the main corrosive agents and some worst case scenarios can be identified. Based on these, rates of deterioration and hence an assessment on the longevity of concrete can be estimated. Physical modelling, based on mathematical descriptions of physical phenomena, is inappropriate for this study due to the complexity of the system. It is felt that mathematical expressions based on statistical approach can best satisfy the requirements of long- term durability predictions (9). Studies based on empirical models and mathematical expressions by Ming Zhu et al. (12), using the computer code CHEMTRN, shows that calcium hydroxide in a repository concrete will be depleted to a depth of 9 cm in 1000 years.

POSSIBLE CORROSIVE AGENTS

Of the deleterious agents examined, the following are expected to influence the durability of the IRUS repository concrete:

Sulphate ions

Reaction of solutions containing sulphate ions with the cement paste can lead to disintegration of the concrete structure by expansion and cracking. Experiments are underway to determine the average sulphate content of the waste and diffusion rate through the saturated sand backfill. For the durability studies, 6 different concentrations ranging from 1.5 to 60 g/L will be used to cover all expected conditions.

Chloride ion

Chloride ion may be detrimental to concrete in several ways (4,5,6). The worst effect is the corrosion of the reinforcement resulting in cracking of the concrete. We consider the corrosion of reinforcement as one of the major degradation agents. Further study is required to establish the relationship between the thickness of concrete covering the reinforcement, the rate of corrosion of reinforcement, and the induced internal stresses which will cause cracking of the concrete. This study is conducted under a separate research program. The groundwater at present contains less than 300 mg/L of chloride ions. However, for the durability studies, six concentrations from 300 mg/L to 30,000 mg/L will be used to cover a possibility of increased chloride concentrations from road salts.

Reactions with the Aqueous Environment

When concrete is exposed to flowing water, the calcium hydroxide, which is relatively soluble, leaches from the cement paste. The strength of concrete is lowered approximately 1.5% for every 1% loss of total calcium content (4). Even though the repository is located a meter above the water table, it is possible to have the structure in contact with infiltrating surface water or groundwater during abnormally high water levels. To study the leaching process, concrete specimens will be exposed to distilled water with gentle stirring to mimic the flow of groundwater under the repository.

Carbon Dioxide

The reaction between dissolved carbon dioxide and cement can either result in enhanced dissolution of calcium as calcium bicarbonate, or precipitation of calcium carbonate, depending on the pH of the aqueous phase. During the post-closure period, it is possible that the repository will contain high concentrations of carbon dioxide from the degradation of the cellulose wastes. This repository condition will be stimulated by concrete specimens placed in the aggressive media, through which carbon dioxide is bubbled. The rate of reaction of carbon dioxide with the concrete will be measured.

Synergistic Effects

It has been reported in the literature that the presence of chloride ions can increase the rate of reaction of sulphate ion in concrete (7). Experiments will be set up to measure the rate of corrosion under the combined reactions of sulphates, chlorides and carbonates.

MATERIAL TEST PLAN

The material testing will be done in the following 5 stages:

1) Finalize mix design for 20 different concretes and cement pastes. Figure 4 shows the concrete specimen details.

2) Measure concrete and cement paste physical and chemical properties in hardened state before exposure to aggressive media.

3) Conduct experiments to determine the rate of corrosion of concrete specimen exposed to selected aggressive elements containing single or multiple ionic species at different concentrations for a period of 1 to 2 years to obtain the basic experimental data. Extend the duration of these experiments for a number of years to obtain longer term data.

4) Determine the rate of penetration of the reaction front using the following methodology:
   - determine porosity-depth profiles when the amount of reaction is significant
   - determine depth profile of aggressive ion concentration by wet-chemical and microprobe analysis.

5) Evaluate data collected for concrete and paste systems, correlate accelerated data and develop models, and finally validate models against long- term data.

ACCELERATED TESTING

It is important that, for any accelerated testing to be meaningful, the deterioration mechanisms taking place under accelerated conditions should be the same as those occurring under normal conditions. In the planned laboratory testing, one of the accelerating agents beside increased concentration is elevated temperature. The temperature selected must be low enough to avoid any detrimental phase changes in the hydrated products in concrete. Acceleration factors will be obtained by comparing the long- term normal exposure data with the short- term accelerated data.

METHODS OF EXTRAPOLATION

As time passes, changes in concrete may occur due to internal reactions and due to reactions with the aqueous environment. The most important internal reaction is between cement and concrete aggregates. Of the reactions with the environment, sulphate attack, calcium leaching, and chloride attack are most important. The relationship between location of the reaction front in the concrete specimen, and time, will be established for several types of concrete using these aggressive elements. From these, the rate of corrosion of concrete will be evaluated for the short-term basic experimental data. Long- term corrosion rate predictions, even though difficult, can be made by extrapolating the basic data. The procedure is illustrated in Fig. 5, 6. Confidence limits will improve as longer term data from the experiments become available.

CONCLUSION

The durability of concrete is an important aspect for a vault-type radioactive waste repository such as IRUS. The objective of the research project is to identify the degradation mechanisms for the IRUS repository concrete and to establish short-term and long-term corrosion rates. It is also our objective to predict the useful engineered service life for a quality concrete under various repository service conditions. Even though the IRUS repository service conditions are rather favorable, various aggressive agents were selected for the study, taking into account the long post- closure period. To predict long- term durability of concrete from short- term data, accelerated testing and methods of extrapolation will be used. Preliminary results are expected to be available by November 1989.

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