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AN OVERVIEW OF TECHNICAL REQUIREMENTS ON DURABLE CONCRETE PRODUCTION FOR NEAR SURFACE DISPOSAL FACILITIES FOR RADIOCTIVE WASTES

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ABSTRACT

Radioactive waste can be generated by a wide range of activities varying from activities in hospitals to nuclear power plants, to mines and mineral processing facilities. General public have devoted nowadays considerable attention to the subject of radioactive waste management due to heightened awareness of environmental protection. The preferred strategy for the management of all radioactive waste is to contain it and to isolate it from the accessible biosphere. The Federal Government of Brazil has announced the construction for the year of 2014 and operation for the year of 2016 of a near surface disposal facility for low and intermediate level radioactive waste. The objective of this paper is to provide an overview of technical requirements related to production of durable concrete to be used in near surface disposal facilities for radioactive waste concrete structures. These requirements have been considered by researchers dealing with ongoing designing effort of the Brazilian near surface disposal facility.

1. INTRODUCTION

The worldwide technological advance of nuclear power generation improves reliability of nuclear energy utilization. A nuclear energy system encompasses the complete spectrum of the nuclear fuel cycle, i.e. from mining to final end states for all wastes. Therefore, nuclear energy systems must be assessed holistically, from all possible angles of sustainable development. The conception of sustainable development focuses equally on four conditions: (i) improving mankind quality of life and well-being; (ii) meeting the needs of both present and future generations (intra- and intergenerational equity); (iii) justice and equity in terms of recognition, process, procedure and outcome; and (iv) the need for mankind to live within ecosystem limits [1]. As regards sustainability, when evaluated in light of impact on climate, land use, waste disposal, fuel availability, safety (occupational, environmental and personal), internalized environmental costs, and technology transfer, nuclear power is an energy option that is itself sustainable. The proper nuclear waste management is not more a world challenge in the technical aspects. In Brazil, the biggest radioactive waste producers are the two operating nuclear power plants. The waste generated by Angra 1 and Angra 2 is currently being stored inside them. The waste produced by the uranium mining and milling industrial complex, although significant in volume, is kept at the site, in a dam specially built for this purpose. Comissão Nacional de Energia Nuclear (CNEN) – National Nuclear Energy Commission – is responsible for regulation and final disposal of radioactive waste. The Federal Government of Brazil intends to start building a near surface disposal facility for low and intermediate level radioactive waste in 2014 and to begin the operation by the year 2016. This facility will be operated for about 60 years, and it will be released after 300 years after the operational time. The proper functioning of the disposal facility relies on concrete durability.

Durability is not an attribute of concrete in general, but it is rather the reached result from a set of characteristics of a specific concrete subjected to specific environment [2]. During its service life it can be exposed to a wide range of external and internal stresses. Some of these environmental stresses are caused by the aggressive chemicals in groundwater, and soils, marine exposure, and also freeze and thawing cycles. Material related stresses can, in turn, be result from alkali-silica (or alkali-carbonate) reactions, of excessive shrinkage, related to micro-cracking and high permeability. It is accepted that the principal causes for the deterioration of concrete structures are the corrosion of reinforcing steel, exposure to cycles of freezing and thawing, alkali-silica reaction, and attack by sulphate. It is also known that water is the main vehicle for the diffusion of aggressive ions into concrete.

For concrete structures, a long service life is considered a synonymous of durability. According to ACI Committee 201, durability of Portland cement concrete is defined as its ability to resist weathering action, chemical attack, abrasion, or any other process of deterioration; that is, durable concrete will retain its original form, quality, and serviceability when exposed to its environment [3]. It can be stated that when it is properly mixed, placed, solidified, and cured, the concrete is essentially waterproof and should have a long service life under most conditions. One can infer that concrete deterioration is mainly caused by the use of inappropriate materials or poor construction practices. This paper presents an overview of technical requirements related to production of durable concrete to be used in structures of near surface disposal facilities for radioactive wastes. Some basic concepts of radioactive waste disposal systems and chemical deterioration mechanisms on concrete are revised. These requirements have been considered by researchers dealing with ongoing designing effort of the Brazilian near surface disposal facility.

2. RADIOACTIVE WASTE DISPOSAL

According to the CNEN-NE-6.05 Standard, waste disposal is the emplacement of waste in an appropriate facility recommended by CNEN without the intention of retrieval [4]. Waste disposal system is intended to isolate radioactive waste and to control releases of radionuclides into the environment [5]. The overall objective of the design of a disposal facility is to provide design features that ensure the facility can be built and waste received, handled and disposed of without undue risk to human health and the environment, both during facility operation and after facility closure. Near surface disposal is currently practised or envisaged for LILW, essentially short lived radioisotopes (half-live is not in excess of 30 years), with long lived radioisotopes (half-life is in excess of 30 years) being limited to very low concentrations [6]. The two categories of near surface disposal facilities are:

- <u>Above ground disposal facilities or those built within about 10 m of the surface</u> – The disposal units or vaults are typically concrete lined and can be sub-divided into cells by internal concrete partitions to ensure that the operational area used for disposal at any given time is not too large. Each cell is filled in turn. There is usually some form of drainage system to channel any infiltrating water away from the waste.

- Deeper cavern facilities built at depths of up to 100 m – The primary distinguishing sub-surface disposal concept is that the distance below the ground surface is usually adequate to essentially eliminate concerns of intrusion by plants, animals and humans. Typically these caverns may be classified as specially excavated cavities and disused mines. Where the cavities are specially excavated then a variety of shapes and volumes have been employed which have been influenced by the planned disposal methods, the waste type and quantity and, of course, the geometry of the host formation. The repository can include tunnels, vaults, vertical or horizontal caverns or some combination of these.

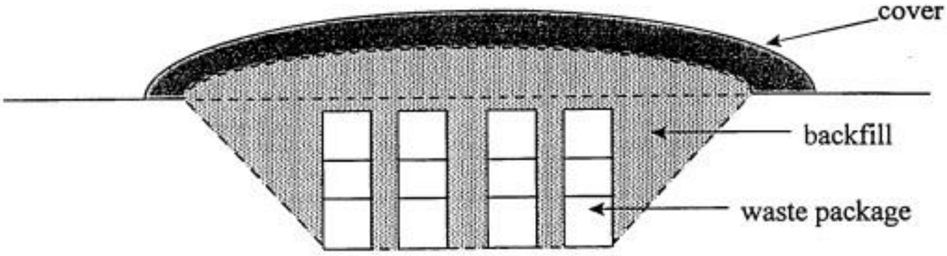
The generic concepts for near surface disposal can be described as follows [7]:

- Covered trench – This is the oldest and simplest of the disposal concepts that consists of placing waste into excavated trenches and covering the filled trenches with soil. Disposal sites using this concept frequently have retrofitted engineered barriers (Figure 1 a).
- Closed vault – This consists of a concrete vault into which is placed packaged and/or treated waste. The voidage may be backfilled and the structure closed with concrete slabs, which may be sealed by, for example, asphalt. The whole structure is then protected by an earthen cap (Figure 1 b).
- Domed vault – This concept infiltration is controlled by placing waste in a dry permeable layer and covering the waste with an impermeable concrete roof that is subsequently protected by an earthen cap (Figure 1 c).
- Open vault – In this concept, a low permeability cap is placed over the filled vault without emplacement of a concrete slab. Waste is however pre-treated to minimize voidage. The cap is designed to accommodate some settlement (Figure 1 d).

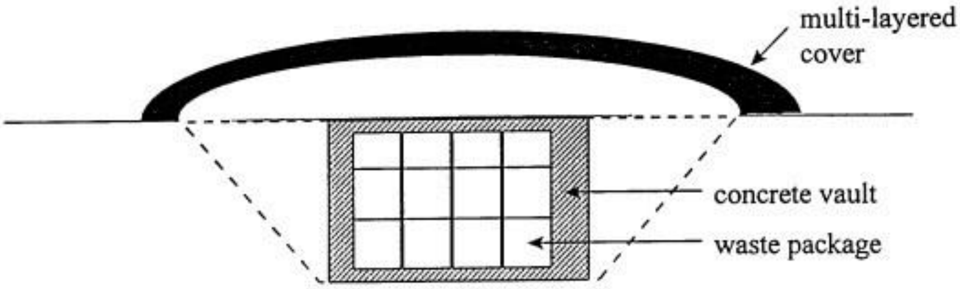
Near-surface disposal in unlined trenches or pits is a disposal option practiced in many countries. Near-surface disposal with engineered barriers systems incorporate a number of engineered barriers that can be used as physical and/or chemical obstructions to prevent or delay nuclide migration from the repository, and the barriers represent an important component of the disposal facility safety from the operational phase, through the period of institutional control, and ultimately to the possible free release of the site [7].

There are three phases associated with the service life of a near surface disposal facility [8]:

- Pre-operational – It includes the necessary siting and design studies and the period of construction of the repository.
- Operational – It includes the period of operations at the repository and the closure of the repository.
- Post-closure – It includes any activities following closure of the repository (for example, periods of active or passive controls).



(a)



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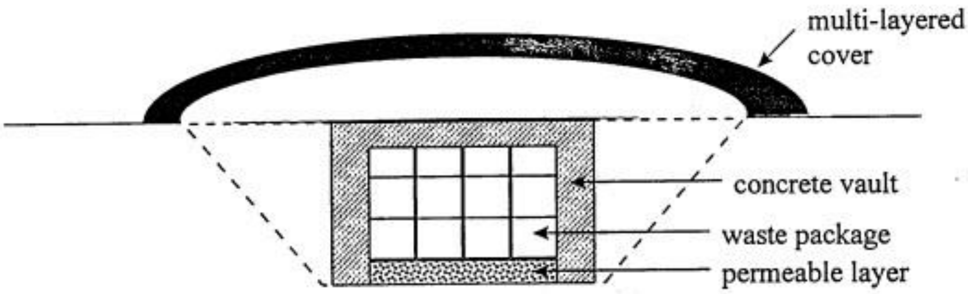

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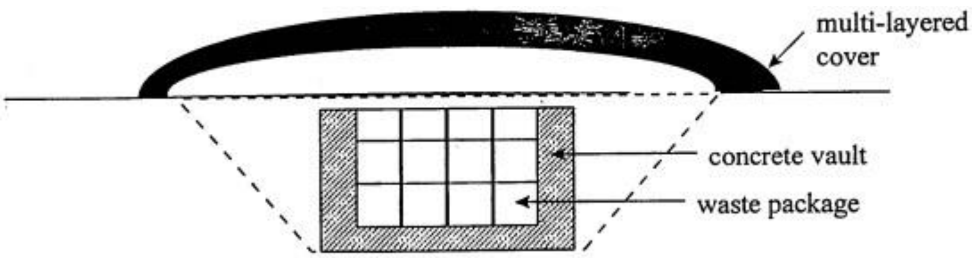

(d)

Figure 1: Examples of near surface disposal facilities concept: (a) covered trench; (b) closed vault; (c) domed vault; (d) open vault [7].

3. EXAMPLES OF NEAR SURFACE RADIOACTIVE WASTE DISPOSAL FACILITIES WITH ENGINEERED BARRIERS

The repository projects of Canada and Spain are presented as examples of near surface radioactive waste disposal facilities with engineered barriers.

3.1. Canada

In Canada, the facility for the disposal of low and intermediate level radioactive waste is currently in design phase. It is called Intrusion Resistant Underground Structure (IRUS), and it will be built in the Chalk River Nuclear Laboratories (CRNL), a Canadian nuclear research institute, located near Chalk River, about 180 km northwest of Ottawa, in the province of Ontario. The disposal facility will be operated by the Atomic Energy of Canada Limited (AECL). The IRUS includes a concrete domed vault with a permeable bottom that will be built above the water table in a sand formation. The permeable floor is designed to minimize the contact of water with the waste. Since the waste will contain small concentrations of very long-lived radionuclides, engineers have planned for the eventual infiltration of water as the concrete deteriorates over the long term: any water is channeled to readily drain through the floor, which is formed of two mixed layers of sand, clay, and natural zeolite. The adsorptive properties of the layers will limit the release of radionuclides with the draining water [9].

The dimensions of the concrete vault are 30 meters wide, 20 meters long, 8 meters high. The concrete vault roof is a 1-meter-thick reinforced-concrete slab designed to resist infiltration of water as well as to deter inadvertent intruders. The roof slab will be overlaid by a multi-layer-earthen cover engineered to limit infiltration through the roof, and to isolate the roof and the module structure from freeze-thaw cycles [10]. Concrete durability is a concern in the IRUS design. The focus is to have a durable concrete for the required service life of 500 years for the IRUS facility. In their research program on concrete durability it is estimated the long term performance of a wide variety of concrete types and qualities under the disposal conditions. Four different water-cement ratios were used for the manufacture of concrete specimens to study the influence of this important parameter on the performance of concrete. Five different concrete systems with different combinations of the following cemetitious materials were selected for study: Canadian Ordinary Portland Cement (ASTM type I), Canadian Sulphate Resisting Portland Cement (ASTM type IS), blast furnace slag, fly ash, and silica fume [7].

3.2. Spain

In Spain, the disposal of low and intermediate level radioactive waste is in a facility called El Cabril, at Sierra Albarrana, about 100 km northwest of Seville, in the province of Córdoba. Its operation began in 1992, after the license has been granted by the Ministry of Industry and Energy. The disposal facility is operated by Empresa Nacional de Residuos Radiactivos SA (ENRESA) – Spanish National Radioactive Waste Management Company. The disposal system is fundamentally based on the incorporation of natural and engineered barriers, isolating safely the disposed wastes by the time necessary for them to be converted into harmless substances [11]. The waste disposal facility consists of a concrete closed vault.

In order to have a compact concrete to minimize progress of aggressive intrusion or carbonation progression, the designed characteristic compressive strength was raised from 30

MPa to 35 MPa. A minimum amount of 400 kg of cement per cubic meter was specified to have a reserve of alkalinity. The Spanish Portland Cement I 45 A SR MR (ASTM type IS) was used and the water-cementitious materials ratio (w/c ratio) was limited to 0.42 to get a low permeability. El Cabril was designed for a service life of 300 years. El Cabril presents the following favorable conditions regarding durability of concrete structures [7]:

- It is far away from marine environment (absence of chlorides in air or solid deposits).
- It is in a rural area without air pollution.
- The site underground water and soil, as well as dam water, used in concrete preparation do not present significant quantities of chloride, sulphate or other aggressive materials.
- Climate is mild with no special frost hazards.
- The disposal system is located above water table level.

4. CHEMICAL DETERIORATION MECHANISMS ON CONCRETE

4.1. Sulphate Attack

In a well-hydrated Portland cement paste present in concrete, the solid phase is primarily composed of relatively insoluble hydrates of calcium such as calcium silicate hydrates gel (C-S-H), calcium hydroxide or portlandite (CH), and aluminum substituted calcium silicate hydrate (C-A-S-H). These hydrates exist in a state of stable equilibrium with a high-pH pore fluid of 12.5 to 13.5; pH is kept high due to the large concentrations of Na^+, K^+, and OH^- ions.

Solid salts do not attack concrete, but when they are in the form of a solution, they can directly react with the hardened cement paste. Sodium, potassium, magnesium and calcium sulphates are the most common in soils. Whenever these sulphates come into contact with groundwater, they form a sulphate solution [12]. Sulphates react with various phases of hydrated cement paste leading to expansion, cracking, and spalling. The sulphate attack is generally attributed to the formation of expansive ettringite and gypsum, which are known to precipitate by through-solution mechanism. Investigations, however, suggested that the sulphate attack is partially due to the cement paste losing its stiffness when exposed to sulphate rich environment [13].

Calcium, sodium, magnesium, and ammonium sulphates are, in increasing order of hazard, harmful to concrete as they can cause expansion, loss of strength, and eventually transform the material into a mushy mass. Calcium sulphate reacts with calcium aluminate hydrates, thus forming expansive ettringite. Sodium sulphate reacts with calcium hydroxide and forms expansive gypsum, which, in the presence of aluminates, may produce ettringite.

Magnesium sulphate reacts with all cement compounds, including C-S-H, forming brucite and gypsum, which, at a later age, can give ettringite. The concrete deterioration by ammonium sulphate covers the most aggressive corrosion on concrete: only expansive gypsum is formed and no protective layer like brucite is created. There occurs not only expansion due to the formation of gypsum and ettringite, but also intensive dissolution of cement hydrates [14]. In order to have a sulphate resisting reinforced concrete structure, a sulphate resisting Portland cement shall be used; its superior performance as sulphate resistant is because of the dilution of the C_3A phase due to a reduction in the quantity of cement.

4.2. Carbonation

Carbon dioxide (CO_2) present in the atmosphere reacts in the presence of moisture with the hydrated cement minerals (i.e. the agent usually being the carbonic acid, since CO_2 gas is not reactive). The extent of carbonation depends on the permeability of the concrete and on the concentration of carbon dioxide in the atmosphere. The penetration of carbon dioxide beyond the exposed surface of concrete is extremely slow. The alkaline conditions of hydrated cement paste are neutralized by carbonation. This neutralization, by reducing the pH from over 12 to about 8.3, affects the protection of reinforcing steel bars from corrosion. Thus, if the entire concrete cover to steel were carbonated, corrosion of steel would occur in the presence of moisture and oxygen [12].

4.3. Acid Attack

Concrete is susceptible to attack by sulfuric acid produced from either sewage or sulfur dioxide present in the atmosphere of industrial cities. This attack is due to the high alkalinity of Portland cement concrete, which can be attacked by other acids as well. Sulfuric acid is particularly corrosive due to the sulphate ion participating in sulphate attack, in addition to the dissolution caused by the hydrogen ion [15]. The deterioration of concrete by acids is mainly the result of reaction between these chemicals and calcium hydroxide of the hydrated Portland cement. In most cases, the chemical reaction results in the formation of water soluble calcium compounds which are then leached away by aqueous solutions.

5. CONCRETE DURABILITY REQUIREMENTS

According to ACI 318 [16], Portland cement concrete to be exposed to sulphate-containing solutions or soils shall conform to requirements stated in Table 1.

For mild acid exposure, the Portland cement concrete mixture should be designed to keeping the water-cementitious material ratio, by weight, lower than 0.40 and using a Portland Cement ASTM type II; for severe acid exposure an acid-resistant coating shall be used. To limit carbonation to a very slight penetration level, the water-cementitious material ratio, by weight, of 0.40 or lower shall be used. In addition, good curing process should be ensured, and the steel bars should be, at least, 50 mm below the surface [2].

In Brazil there are three types of Portland cement suitable for production of concrete that will be exposed to aggressive chemical environment: Brazilian Portland Cement CP-II, CP-III, and CP-IV.

- Brazilian Portland Cement CP-II (ASTM type I (SM)(MS)): It is used where moderate sulphate resistance and low heat of hydration is important.
- Brazilian Portland Cement CP-III (ASTM type IS): It is used where sulphate resistance and low heat of hydration is important. This cement has a very low C3A composition which accounts for its high sulphate resistance. Its blast furnace slag content is from 35% up to 70%.

- Brazilian Portland Cement CP-IV (ASTM type IP): It is used where acid and sulphate resistance is necessary. Its heat of hydration is low. Its pozzolanic constituent content is from 15% up to 50%.

Table 1: Requirements for concrete exposed to sulphate containing solution [16]

Characteristic	Parameter			
Sulphate exposure	Negligible	Moderate	Severe	Very severe
Water soluble sulphate (SO_4) in soil, (% wt)	0.00-0.10	0.10-0.20	0.20-2.00	Over 2.00
Sulphate (SO_4) in water (ppm)	0-150	150-1500	1500-10,000	Over 10,000
ASTM Portland cement type*	–	II, IP(MS), IS(MS), P(MS), I(PM)(MS), I(SM)(MS)	V	V plus pozzolan
Maximum water-cementitious materials ratio, by weight, for normal weight aggregate concrete	–	0.50	0.45	0.45
Minimum f_c for normal and light weight aggregate concrete (MPa)		28	31	31

*MS = Moderate sulphate resistance; PM = Pozzolan modified Portland; SM = Slag modified Portland.

6. CONSTRUCTION TECHNIQUE

The quality of concrete, specifically one with low permeability , is the best protection against some chemical attack, and it is accomplished by producing a concrete with low water-cementitious materials ratio by weight. Moreover, the concerns must be on the proper dosage, mixing, transporting, placing, curing process, and the control procedures for the concrete production. In this context the precast concrete construction arises as a very good option. Precast concrete is a construction product made by casting concrete in a reusable mold, which is then cured in a controlled environment of a precast industry plant, being transported to the construction site and lifted into place. The use of precast concrete systems offers potential advantages over “site casting concrete”, such as a greater control of the quality of materials and products. Besides, the forms used in a precast industry plant may be reused many times before they have to be replaced, reducing the cost of formwork per unit. The concept of sustainability in building and construction could be fully reached using precast concrete systems. Sustainable building design and construction is the practice of creating buildings using efficiently processes and resource, in an environmentally responsible way throughout the life cycle of a building.

7. CONCLUSIONS

Brazil faces nuclear energy as an essential component in an overall energy strategy. Suitable management of radioactive waste is essential for the future of mankind. Therefore, building a surface disposal facility for low and intermediate-level radioactive waste in Brazil is of capital importance.

The concrete structure of a repository should last about 400 years and in order to enhance durability of reinforced concrete structure against chemical aggressive environment, the following parameters should be considered:

- Concrete will not be seriously damaged by sulphate attack if it is used a highly sulphate resisting cement.
- Water-cementitious materials ratio by weight shall be of 0.40 or lower.
- Precast concrete construction shall be used in order to accomplish proper dosage, mixing, transporting, placing, curing process, and control procedures for the concrete production.

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