MODELLING THE DURABILITY OF CONCRETE FOR NUCLEAR WASTE DISPOSAL FACILITIES

Olli-Pekka J. Kari (1), Jari A. Puttonen (1)

(1) Helsinki University of Technology, Department of Structural Engineering and Building Technology, Finland

Abstract

The multiple engineered barriers ensuring the safety of low- and intermediate-level waste repositories are required in the Finnish disposal concept to be serviceable for at least 500 years after the facilities are sealed. The engineered barriers mainly consist of concrete structures and this requires the design work and justification to be based on knowledge of the fundamental degradation mechanisms of reinforced concrete under such conditions.

The fundamental questions to be clarified are the effect of the interaction between different mechanisms on the ageing of reinforced concrete and the possible differences between the results of conventional methods and those achieved with a mathematical model that takes into account the interaction of the relevant degradation mechanisms. The mechanisms studied were: the aerial carbonation of concrete; chloride penetration; the corrosion of concrete caused by the sulphate and magnesium, and the leaching of cement paste.

The results achieved by implementing the model into a finite element program indicate that the interaction of deterioration mechanisms is an important factor and should be considered when estimating the durability of reinforced concrete for over 500 years. It was confirmed that the long-term deterioration of reinforced concrete may not be estimated with sufficient accuracy by conventional single-phenomenon models.

1. INTRODUCTION

1.1 Background

The low- and intermediate-level wastes that accumulate during the operation of nuclear plants will be disposed of in an underground repository in the bedrock in the Finnish disposal concept. The safety of the repository is ensured by multiple engineered barriers which mainly consist of concrete structures. It is required that the engineered barriers must be serviceable at least 500 years after the repository has been sealed. However, there is a lack of direct experience with reinforced concrete structures with a service life even close to that demanded, as reinforced concrete has only been used as a material for the construction of buildings for a little less than 150 years. Therefore, the design work of structures of this kind and justification of their service life have to be based on the knowledge of the fundamental degradation
mechanisms of reinforced concrete under such conditions. The need for a mathematical model for the assessment of both the mechanisms and their interaction is obvious.

Traditionally, the mathematical examination of the durability of reinforced concrete has been performed by studying each individual degradation mechanism one at a time. There is, however, doubt as to whether the conventional methods have the capability to describe the deterioration of the structure accurately enough, as the interaction of the various deterioration mechanisms is not considered. This type of deterioration may be even more harmful than the single degradation mechanisms indicated. Thus, the fundamental questions to be clarified are the effect of the interaction between different mechanisms on the ageing of reinforced concrete, and the possible differences between the results achieved with conventional methods and the mathematical model which takes into account the interaction of the mechanisms.

The model developed in this study can be applied not only to the engineered barriers but all the reinforced concrete structures, which makes it an important tool in the ageing management.

1.2 Objective

The primary object of the study is to develop a generalised numerical model for the estimation of the degradation of concrete in final disposal conditions. The solution of the model is to be based on the finite element method and to be carried out with commonly available software.

The aim is to integrate into the model all the fundamental factors that have an effect in such conditions. Another goal is to form a general view of the usefulness of the conventional methods in estimating the degradation of concrete in the conditions that are of concern here. The subjects considered in the model are carbonation, moisture ingress, chloride penetration, the corrosion of concrete caused by the intrusion of both sulphate and magnesium, and the leaching of cement paste compounds into groundwater. In addition, the effects of concrete admixtures (here silica fume and blast furnace slag) are included in the model. In the modelling the objective is sufficient accuracy for the estimation of durability over periods of hundreds of years, considering the geometry of the structures. Consequently, the model is three-dimensional.

2. DEGRADATION PROCESSES OF REINFORCED CONCRETE IN THE DISPOSAL ENVIRONMENT

In the prevailing conditions at the disposal facility, besides the corrosion of the reinforcement, the corrosion of the concrete, i.e. disintegration, cracking, and spalling, also has to be taken into account. However, the prerequisite for the initiation of the corrosion of the reinforcement will probably be fulfilled first. In this study, in practice, the initiation period of corrosion is examined. Therefore, neither the consequential effects of the corrosion of the reinforcement after the propagation period, resulting in the deterioration of the concrete (loss of reinforcing cross-sectional area and accumulation of corrosion products) nor the controlling factors of corrosion are included in the study. Actually, the confining facilities will fulfil their function as a barrier as long as any solid concrete surrounds them.

The conditions in the surrounding environment of the repository in an underground repository can be divided into three different periods of time: the construction stage, operating
phase, and post-closure period. Each of the periods has various effects regarding the durability of the concrete.

During the construction stage, the concrete may be exposed to the detrimental effect of high temperatures (over 60 °C) if the generation of hydration heat is not managed during the construction. The environmental temperature can be considered as constant after the construction phase, being between 7 and 10 °C, according to the measurements.

During the second period, i.e. the operating phase, dank air, in practice, surrounds the concrete. The estimated length of the operating period will be 50-100 years. During this period, the concrete structure will be exposed to aerial carbonation caused by carbon dioxide in the air, intensified by the optimally relative humidity for carbonation (average values between 60 and 100%). This phenomenon has both positive and negative impacts on the durability of the concrete. In the worst-case scenario, carbonation leads to the corrosion of the reinforcement. The carbon dioxide content in the ambient air at the disposal site is higher than in the open air. The average carbon dioxide content values in the air are between 500 and 600 ppm.

After the repository has been filled up, it will be sealed. Thereafter saline groundwater will gradually fill the disposal zone, exposing the concrete to various mechanisms of deterioration. The most significant aggressive ions associated with the groundwater from a durability point of view are sulphates, magnesium, and chlorides. These ions can be highly destructive for the concrete and the reinforcement, especially when their combined effects are taken into account. In addition to that, the major components of the cement paste can leach out during the interaction with the water.

3. NUMERICAL MODELS FOR ESTIMATION OF CONCRETE DEGRADATION IN DISPOSAL ENVIRONMENT

3.1 Modelling Assumptions and Limitations

In order to develop the model for the estimation of the durability of concrete in the conditions described, some assumptions regarding the factors affecting the destructive phenomena should be made, and the limitations of the model considered. The following assumptions and limitations are applied in the model:

- the modelling is mathematically based on Fick’s diffusion theory and Finite Element Method;
- the boundary conditions are constant during each time period;
- the effects of different deterioration mechanisms on the strength of the structures are not included in the model;
- hydrostatic pressure is not assumed to affect the structures when the repository is submerged;
- the velocity of the water flow in the repository area in its submerged condition is not high enough to cause erosion of the concrete. The flow velocity is assumed to be, however, sufficient to transport away the material leached from the concrete, causing the boundary concentration of the material under consideration to approach zero;
- the time required for filling up the repository with water after it has been sealed is short compared with the designed service life;
- the reactivity of aggregates concerning the alkali-aggregate reactions (AAR) is negligible;
the nuclear wastes have no effects on durability (e.g. increasing of heat, harmful substances resulting from the decomposition of wastes);  
- The concrete is uncracked (excluding the consequences of sulphate corrosion). This assumption requires the structure to have been designed to avoid major stresses. On the other hand, the self-healing of cracks can be expected to be a predominant phenomenon;

3.2 Combined Effects of the Deterioration Mechanisms

The modelling assumptions are applied to the combined effect of the deterioration mechanisms. All the effects are directed to the diffusion coefficients of the substances. The assumed interaction of the degradation mechanisms is presented in Figure 1.

![Figure 1: Assumed interaction of the main degradation mechanisms][1]

3.3 Mathematical Model for the Combined Effects

The numerical models developed according to the theory at the detailed level can be found in [1]. Anyway, the main equations are as follows:

Carbonation

$$\frac{\partial g}{\partial t} = \text{div}[D_g \cdot \text{grad}(g)] + \alpha_4 \frac{\partial c}{\partial t}$$

where,

- $D_g$ = carbon dioxide diffusion coefficient [m$^2$/s]
- $g$ = carbon dioxide concentration [kg/m$^3$]
- $\alpha_4$ = material parameter which reflects the characteristics of the concrete [-]
- $c$ = concentration of CaCO$_3$ [kg/m$^3$]
Moisture ingress
\[ \frac{\partial W}{\partial t} = \frac{\partial W}{\partial H} \cdot \frac{\partial H}{\partial t} = (\text{div}[D_h \cdot \text{grad}(H)]) \]  
where,
- \( W \) = total water content for the unit volume of material [g/g]
- \( H \) = pore relative humidity [-]
- \( D_h \) = humidity diffusion coefficient [m\(^2\)/s]

Chloride ingress
\[ \frac{\partial C_f}{\partial t} = \frac{\partial C_f}{\partial C_t} \cdot (\text{div}[D_{Cl} \cdot \text{grad}(C_f)] + \mu \cdot \frac{\partial W}{\partial t} \cdot C_f) \]  
where,
- \( C_t \) = total chloride concentration [g\(_{cl}\)/g\(_{concrete}\)]
- \( C_f \) = free chloride concentration [g\(_{cl}\)/g\(_{concrete}\)]
- \( \frac{\partial C_f}{\partial t} \) = binding capacity of the concrete [-]
- \( D_{Cl} \) = chloride diffusivity of the concrete [m\(^2\)/s]
- \( \mu \) = unit converter [-]
- \( W \) = total water content for the unit volume of material [g/g]

In saturated concrete the term \( \frac{\partial W}{\partial t} \) vanishes.

Sulphate and magnesium corrosion
\[ R_d = \frac{X_d}{t_d} = E \cdot B^2 \cdot C_e \cdot C_0 \cdot D_{SO4} / \alpha \cdot \tau \cdot (1 - \nu) \]  
where,
- \( R_d \) = degradation rate of concrete [m/s]
- \( X_d \) = degradation depth [m]
- \( t_d \) = time required for degradation [s]
- \( E \) = Young’s modulus of concrete [Pa]
- \( B \) = linear strain caused by one mole of sulphate reacted in 1 m\(^3\) of concrete [m\(^3\)/mol]
- \( C_e \) = external sulphate concentration [mol/m\(^3\)]
- \( C_0 \) = concentration of reacted sulphate as ettringite [mol/m\(^3\)]
- \( D_{SO4} \) = diffusion coefficient of sulphate ions [m\(^2\)/s]
- \( \alpha \) = roughness factor for the fracture path [-]
- \( \tau \) = fracture surface energy of concrete [J/m\(^2\)]
- \( \nu \) = Poisson’s ratio [-]
Leaching of concrete

\[ \frac{\partial (\theta C_{Ca})}{\partial t} = \text{div}[D_{Ca} \cdot \text{grad}(C_{Ca})] - \frac{\partial C_{Ca,s}}{\partial t} \]  

where,

- \( \theta \) = porosity of the cement paste \([\text{m}^3/\text{m}^3]\)
- \( C_{Ca} \) = calcium concentration in the pore solution \([\text{mol}/\text{m}^3]\)
- \( C_{Ca,s} \) = calcium concentration in the solid phase \([\text{mol}/\text{m}^3]\)
- \( D_{Ca} \) = diffusivity of calcium in the porous media \([\text{m}^2/\text{s}]\)

4. PRELIMINARY RESULTS OF THE NUMERICAL SIMULATION

The estimation of the durability of disposal facilities was performed by assuming conservative boundary values and conditions. Carbonation was presumed to last 50 years during the operating phase of the repository. The length of the post-closure period, when the structure is submerged and exposed to various mechanisms of degradation, was assumed to be 500 years. Three different binder combinations with three constant water-to-binder and aggregate-to-binder ratios were used in the simulations. Silica fume and/or blast furnace slag were used partly as a cement replacement in some of the mixes, while some of the mixes consisted of pure sulphate-resisting cement as a binder. The used mixes are presented in Table 1.

The simulation results were compared to the results achieved with conventional methods and laboratory experiments [2]. Summary of the preliminary results calculated through experiments, empirical and FEM models can be found in Table 2.

### Table 1: Mixture proportions of the concretes.

<table>
<thead>
<tr>
<th>Mix [kg/m³]</th>
<th>CEM</th>
<th>SF</th>
<th>BFS</th>
<th>A_ggr</th>
<th>W/st</th>
<th>A_d åd</th>
</tr>
</thead>
<tbody>
<tr>
<td>B1</td>
<td>453</td>
<td>-</td>
<td>-</td>
<td>1811</td>
<td>1.59</td>
<td>5.0</td>
</tr>
<tr>
<td>B2</td>
<td>373</td>
<td>-</td>
<td>-</td>
<td>1867</td>
<td>1.59</td>
<td>3.0</td>
</tr>
<tr>
<td>B3</td>
<td>319</td>
<td>-</td>
<td>-</td>
<td>1918</td>
<td>1.60</td>
<td>1.3</td>
</tr>
<tr>
<td>B4</td>
<td>402</td>
<td>44</td>
<td>-</td>
<td>1797</td>
<td>1.57</td>
<td>16.9</td>
</tr>
<tr>
<td>B5</td>
<td>334</td>
<td>37</td>
<td>-</td>
<td>1877</td>
<td>1.58</td>
<td>12.2</td>
</tr>
<tr>
<td>B6</td>
<td>286</td>
<td>32</td>
<td>-</td>
<td>1907</td>
<td>1.58</td>
<td>9.9</td>
</tr>
<tr>
<td>B7</td>
<td>89</td>
<td>23</td>
<td>336</td>
<td>1787</td>
<td>1.59</td>
<td>5.4</td>
</tr>
<tr>
<td>B8</td>
<td>74</td>
<td>19</td>
<td>279</td>
<td>1862</td>
<td>1.58</td>
<td>4.1</td>
</tr>
<tr>
<td>B9</td>
<td>63</td>
<td>16</td>
<td>238</td>
<td>1913</td>
<td>1.59</td>
<td>3.2</td>
</tr>
</tbody>
</table>

4.1 Carbonation

The carbonation depths according to the FEM model did not exceed the depth of 40 mm, even in the corner zone after 50 years’ exposure. The calculations predicted that plain cements would perform best, whereas cements with blast furnace slag ingredients were inferior.
4.2 Chloride ingress

The computed chloride ingresses through the FEM model were high with every test concrete mix at ordinary reinforcement depths (25-75 mm), exceeding the possible critical corrosion threshold values (0.05% wt of concrete). It should be noted that the threshold values should be determined more accurately in each separate case than they were here. Anyway, the cement containing silica fume has the best resistance to chloride ingress, the critical chloride penetration depth being 82 mm with the lowest water-to-binder ratio. The equivalent depth with blast furnace slag cement with the same 0.35 water-to-binder ratio was 112 mm, whereas the depth was 166 mm with plain cement.

4.3 Sulphate and magnesium corrosion

The sulphate-based degradation according to the FEM model followed almost the same trend as with the chloride ingress that was calculated, except that the blast furnace cement reached the smallest degraded depth, of 31 mm. The smallest depths with silica fume cement were 43 mm and with ordinary cement 111 mm.

4.4 Leaching of concrete

According to the results of the FEM model for an increase in porosity caused by the leaching of cement paste, all the mixes performed well. The maximum increase in porosity occurred with plain cement with the highest water-to-binder ratio, being to a depth of 66 mm from the surface of the concrete. Otherwise, the depths of the increase in porosity were below 50 mm, the lowest being 23 mm with blast furnace slag cement. On the other hand, the calcium leaching depths were the lowest with ordinary cement with a 0.35 water-to-binder ratio, whereas these depths were the highest for blast furnace slag cements, indicating a considerable loss of the strength of the concrete.

Table 2: Summary of the results calculated through experiments, empirical and FEM models.

<table>
<thead>
<tr>
<th>Concrete River</th>
<th>Carbonation [mm]</th>
<th>Leaching [mm]</th>
<th>Sulphate Corrosion [mm]</th>
<th>Chloride Ingress [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>FEM model</td>
<td>Experiments</td>
<td>FEM model</td>
<td>Emp. model</td>
</tr>
<tr>
<td>B1</td>
<td>3</td>
<td>2</td>
<td>36</td>
<td>106</td>
</tr>
<tr>
<td>B2</td>
<td>6</td>
<td>2</td>
<td>50</td>
<td>117</td>
</tr>
<tr>
<td>B3</td>
<td>10</td>
<td>2</td>
<td>66</td>
<td>128</td>
</tr>
<tr>
<td>B4</td>
<td>3</td>
<td>2</td>
<td>36</td>
<td>116</td>
</tr>
<tr>
<td>B5</td>
<td>6</td>
<td>5</td>
<td>41</td>
<td>127</td>
</tr>
<tr>
<td>B6</td>
<td>9</td>
<td>9</td>
<td>49</td>
<td>138</td>
</tr>
<tr>
<td>B7</td>
<td>7</td>
<td>9</td>
<td>23</td>
<td>135</td>
</tr>
<tr>
<td>B8</td>
<td>14</td>
<td>14</td>
<td>26</td>
<td>148</td>
</tr>
<tr>
<td>B9</td>
<td>27</td>
<td>22</td>
<td>30</td>
<td>161</td>
</tr>
</tbody>
</table>

5. CONCLUSIONS

- it is important to study the effect of the interaction between different degradation mechanisms of reinforced concrete when managing the service life of reinforced concrete under disposal conditions;
the deterioration caused by the combined mechanisms is significantly more harmful than the degradation induced by a single mechanism;

- the conventional methods are not able to describe the deterioration of reinforced concrete caused by coupled degradation mechanisms;

- the evaluation of the reliability of the model that was developed is difficult as some of the degradation mechanisms have mainly a long-term effect and test results cannot be found for their estimation. The validation can, however, be performed at some level by using the empirical and conventional methods for comparison;

- the validation of the model requires proper long-term experimental results and further experimental and analytical research is needed to improve the reliability and applicability of the model;

REFERENCES