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MODELING OF CHLORIDE TRANSPORT IN CONCRETE COUPLED WITH MOISTURE MIGRATION IN MARINE ENVIRONMENT BASED ON THERMODYNAMIC APPROACH

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ABSTRACT: The purpose of this research is to predict penetration behavior of chloride ions in concrete under marine environment. Wherein, concrete structures such as wharves, jetties, docks, breakwaters, piers, dolphins, etc, are exposed to salinity of seawater. The complex marine environment is a jumble of perpetual submergence, intermittent cycling of wetting and drying spells by high and low tide ebbs, wave splashing/pounding, and 100% humid chloride impregnated atmosphere. Ingress of moisture, carrying chloride ions, is central to all these phases of marine environment, though with varying degrees of penetration by all the three transport processes- diffusion, convection, and quasi-adsorption. In this research real time marine environment phases are modeled in laboratory controlled conditions. And a series of experiments have been performed with different combinations of chloride concentrations (to represent seawater), temperature variations, and wetting/drying spell cycle durations. The chloride penetration profiles thus generated are compared with analytical chloride penetration profiles of DuCOM- a durability model developed by Concrete Laboratory, The University of Tokyo, Japan.

KEYWORDS: Chloride transportation, moisture migration, service life prediction in marine environment.

1. INTRODUCTION

Concrete is a forefront heavy construction material parallel to steel, but far excels in bulk use. It has no match to its versatility as foundation material, being cast-able in-situ even under water. It is a wonderful invention of construction industry as a simple manageable composite material of matrix form but of heterogeneous mass distribution. Perfection of construction in concrete is fraught with inhibited disabilities of concrete of multifarious nature and extent. On top of all porosity/permeability is the most grievous and confronting. Thus the ingress of deleterious materials (chlorides and sulphates etc.) is facilitated to troublesome extent.

Transport of chloride ions in concrete is commonly known as through diffusion process only. But, in reality all transport processes analogous to that of momentum and heat transfer play their role. Convective transport of chloride ions in concrete is the major accelerated cause of rapid corrosion of reinforcing steel. The durability of concrete is put to test when it is exposed to the harsh marine environment saturated with sodium chloride, a mega source of chloride ions penetration in concrete structures. The prediction of chloride ions penetration concentration and depth posed a great challenge to civil engineers since long, which still

stands satisfactorily unresolved. For this reason, realistic simulation modeling of chloride penetration is utterly enviable to secure serviceable long life (say about 100 years) of the concrete structures built in chloride rich environments.

In this research of chloride ions penetration in concrete; concentration of chloride in environment phase (water and/or air), temperature variation, and moisture migration have been coupled together for four hygral environment conditions in laboratory-controlled climate. The experimental results are compared with the analytical results obtained by the model. And thus, verification of the DuCOM analytic model has been carried out.

2. RESEARCH SIGNIFICANCE

It is hard to seal entire periphery surface with permanent protective coating. They are all short lived in service. In and out flow of chlorides through air and water phases is the biggest challenge threatening reasonable durable life (say 100 years) of reinforced concrete structures under harsh hostile environments, in particular marine environment.

In hygral exposure of marine environment, there is inward and outward movement of moisture, which is a major cause of chloride ions transportation by convection process as commonly known by diffusion process. These two processes need to be essentially accounted together for complete determination of chloride ions influx. And for that, the moisture transport modeling has to be integrated with chloride concentration model for comprehensive study of chloride penetration profiles, which are direct index of long-term service life of concrete. The research in this regard is being pursued vigorously the world over.

Further, in reinforced concrete, steel comes under severe corrosion attack by the ingress of chloride ions, which in return causes premature deterioration of concrete. Success of the research on controlling and modeling the ingress of chloride ions is of utmost importance before concrete becomes state-of-the-art construction material for 21st century developments.

3. DUCOM- A THERMODYNAMIC DURABILITY SIMULATOR MODEL

Presently, many models have been developed to predict the propagation of chloride ions in concrete. Some models are empirical or semi-empirical methods whereby chloride profiles are predicted by analytical or numerical solutions of Fick's 2nd law diffusion equation. Other models wherein an apparent diffusion coefficient is used. Some scientists used in their model, separate physical expressions for chloride transport and chloride binding.

In this research DuCOM model is used, which is the durability computation model developed by Concrete Laboratory- Department of Civil Engineering, The University of Tokyo, Japan. The originality of this model comes from the fact that DuCOM is a composite multipurpose model, which predicts the state of the concrete from its birth, to its entire life. It comprises several sub-models, which work together and exchange data in real time. And thereby the macro level prediction of life span of concrete is derivable from the micro-level modeling. A schematic representation of different modules of DuCOM is shown in figure 1.1.

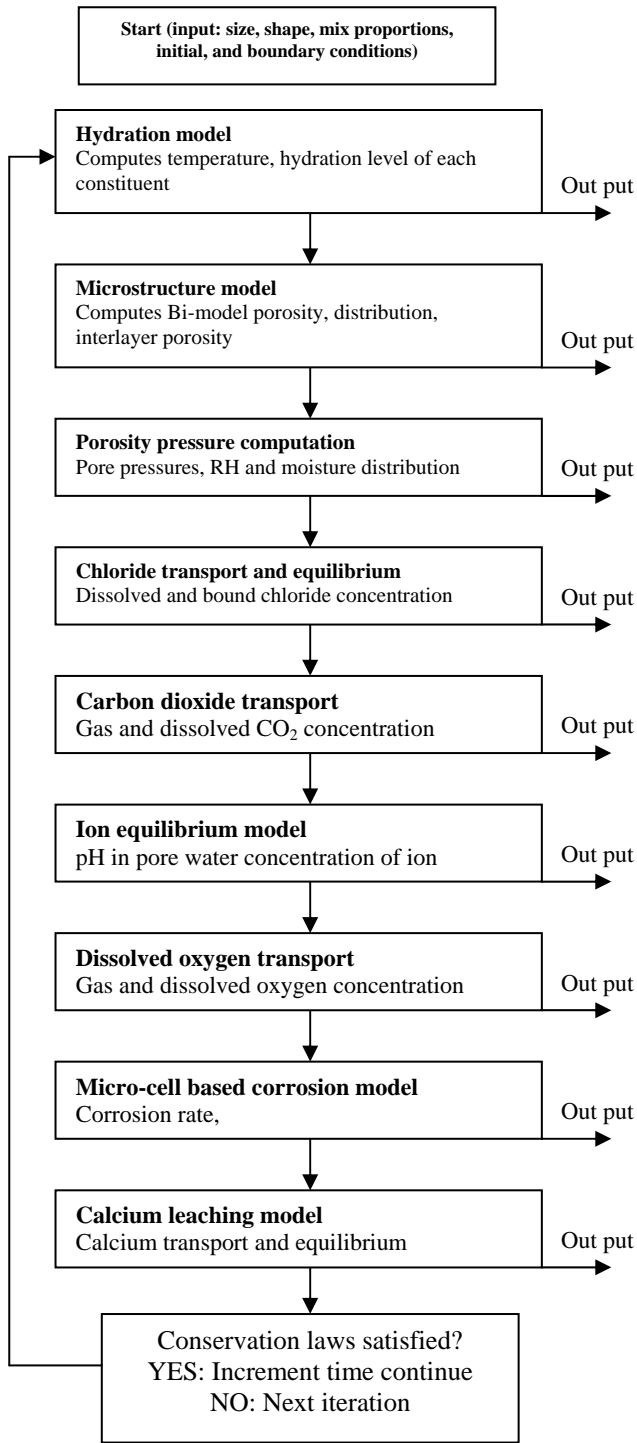


Fig 1.1 Basic Frame of DuCom

The development of multi-scale micro-pore structures at early age are obtained for average degree of cement hydration in the mixture. For any arbitrary initial and boundary conditions, the pore pressure, relative humidity, and moisture distribution are mathematically simulated according to a moisture transport model that considers both vapor

and liquid phases of mass transport. The moisture distribution, relative humidity, and micro-pore structure characteristics in turn control the chloride, carbon-dioxide and oxygen diffusion and rate of carbonation under arbitrary environmental conditions. In this study chloride transport with moisture migration are the primary focal points.

3.1 Modeling of Chloride Transport

Chloride transport in cementitious materials under usual conditions is an advective-diffusive phenomenon. In modeling, the advective transport due to bulk movement or pore solution phase is considered, as well as ionic diffusion due to concentration gradients. Mass balance for free (moveable) chlorides can be expressed as (Ishida 1990a, Takeda and Ishida 2000)

$$\frac{\partial}{\partial t}(\phi S C_{cl}) + \text{div} J_{cl} - Q_{cl} = 0, \quad (1)$$

Where $(\phi S C_{cl})$ is the potential term, from which the porosity and saturation is directly obtained by thermo-hygro physics, Q_{cl} is the sink term for modeling of equilibrium between free and bound chloride, and $\text{div} J_{cl}$ is the chloride flux term. The relationship between free and bound chlorides under equilibrium conditions is model based on several experiments by Ishida et al by measuring concentration of free chlorides by the extracted pore liquid and bound chlorides by XRD. The model is based on Langmuir equation

$$C_b = 11.8 C_f / (1 + 4 C_f) \quad (2)$$

Equation (2) represents the relation between free and bound chlorides for ordinary cement. Where: C_b : bound chloride concentration (Kg Cl/Kg cement), C_f : free chloride concentration (Kg Cl/Kg cement).

$$J_{cl} = -\frac{\phi S}{\Omega} \delta D_{cl} \nabla C_{cl} + \phi S \cdot u \cdot C_{cl} \quad (3)$$

Equation (3) represents the flux term. Where: J_{cl} : chloride flux (mol/m².s), Ω : tortuosity (reduction factor in terms of complex micro-pore structure) δ : constrictivity (takes the effect of pore radius and density of ions) D_{cl} : Diffusion coefficient of chloride ion in the pore solution (m²/s), C_{cl} : concentration of chloride ions in the pore solution (mol/l), $u^T = [u_x \ u_y \ u_z]$: the advective velocity of ions due to the bulk movement of pore solution phase (m/s), C_{ion} : ion concentration (mol/l), ϕ : Porosity of the porous media (m³/m³).

3.1.1 Temperature dependency of chloride flux

The temperature affects in the chloride model is nicely simulated, by considering a temperature dependent diffusion coefficient in the chloride flux equation (3).

$$D_{cl} = RT \frac{\lambda_{ion}}{Z_{cl}^2 F^2} \quad (4)$$

Equation (4) is the mathematical expression to calculate diffusion coefficient of chloride ions, where R: the gas constant (8.314J/Kmol), T: the absolute temperature (K), Z_{cl} : electric charge of chloride ion (-1C), F: Faraday constant (9.65E+4 C/mol), λ_{ion} : ion conductivity (Sm²/mol). λ_{ion} : is a function of temperature and follows the Arrhenius law.

$$\lambda_T = \lambda_{25} \exp \left[-\frac{E_a}{R} \left(\frac{1}{T} - \frac{1}{298} \right) \right] \quad (5)$$

Where: λ_{25} : Ion conductivity at 25°C $\lambda_{25} = 7.63E-3$ (Sm²/mol), E_a : the activation energy for free pore fluid $E_a = 17.6$ (KJ/mol)

From the above formulation and assuming local equilibrium conditions, the distribution of bound and free chloride ions can be obtained, once mix proportions, powder materials, curing and environmental conditions are applied to the analytical system. The detailed description of chloride binding and flux has been explained else where in detail by Maekawa & Ishida 2003.

3.1.2 Chloride Surface Flux Sub-Model

At the boundary level, the surface flux of chloride ions has been modeled taking into account the diffusion and quasi-adsorption flux (Maruya et-al). It has been experimentally known that the concentration of chlorides in the concrete near exposure surface is higher than that of the submerged environment. To simulate this phenomenon, Maruya proposed the condensation model at the surface as shown in fig 1.2

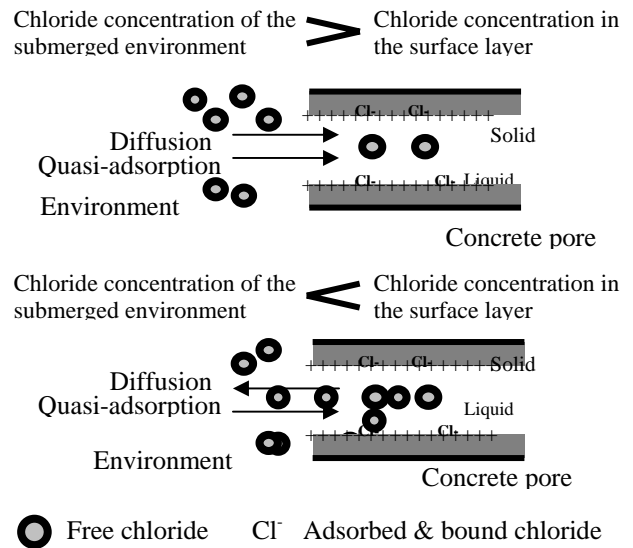


Fig 1.2 Schematic diagram of surface condensation model of chloride Ions

It considers the diffusive movement due to the concentration gradient and the quasi-adsorption phase by electro-magnetic attractive force between the positively charged pore walls and the negatively charged chloride ions. The flux of free chlorides through the boundary surface q_{cl} (mol/m²-s) is described as the summation of the diffusive

component q_{diff} and the contribution of quasi-adsorption q_{ads} . It is expected that the quasi-adsorption flux will decrease as the free and adsorbed chlorides increases, because the migrating chlorides neutralize positive charges on the pore wall. In this system, the flux of quasi-adsorption is described by the following function, in which the flux decreases as a function of the chlorides in the porous medium.

$$Q_{cl} = q_{diff} + q_{ads} \quad (6)$$

$$q_{diff} = E_{cl}(C_{pore} - C_{cl}) \quad (7)$$

E_{cl} has been given a fixed value of 1.0×10^{-3} based on sensitively analysis

$$q_{ads} = K_{cl}(C_{cl}/0.51)^2 \exp(-1.15C_{cl}) \quad (8)$$

C_{cl} : Chloride concentration at the exposure surface,

C_{pore} : chloride concentration in the pore.(mol/l).

For ordinary Portland cement $K_{cl} = 1.5 \times 10^{-3}$

Coefficients in the above equations are determined by experiments using 100% Portland cement. To examine the performance of the chloride surface flux model coupled with moisture transport a comprehensive experimentation series is carried out under laboratory-controlled conditions. In addition to this equation, moisture conductivity in concrete is very important in this regard, since the chloride transportation is dependent on it, as the increase in moisture flux will indirectly affect the diffusion and convection parameters of chloride penetration model.

3.2 Moisture Conductivity Process

The moisture transport model for cementitious materials considers the multiphase dynamics of liquid and gas phases. The contributions of the diffusive as well as bulk movements of moisture have been combined in the model. The total porosity of the cementitious material is divided into interlayer, gel and capillary porosity. The moisture model considers the contributions from each of these components from a thermodynamic viewpoint. The

macroscopic moisture transport characteristics of flow, such as conductivity are obtained directly from the microstructure of the porous media, and this is achieved by considering the mass conservation of moisture capacity, conductivity, pore-structure development, and hydration loss.

The moisture capacity is obtained from pore structures, summation of gel, capillary and interlayer. Moisture conductivity comprises of liquid and vapor phases computed from pore structure. Pore structure development is based on cement particle expansion, and average degree of hydration, and lastly moisture loss due to hydration is directly obtained from hydration model of DuCOM. The moisture flux for both vapor and liquid water can generally be expressed as equation (9)

$$J = -(D_p \nabla P_l + D_T \nabla T) \quad (9)$$

Where, D_p : moisture conductivity (Kg/Pa.m.s) with respect to the pore pressure gradient, and D_T : moisture conductivity (Kg/Pa.m.s) with respect to temperature gradient. Capillary and gel pores are idealized as cylindrical pores. Liquid transport in the porous network would be only through the pores which are capillary condensed. Integrating the liquid flux over the complete microstructure using random pore distribution model, the equation of moisture flux was proposed. A general frame work of mass and moisture equilibrium, flux of moisture in liquid and vapor phases, and overall moisture balance of concrete equations have been presented elsewhere in detail by Maekawa & Ishida 2003

3.2.1 Moisture Surface Mass Flux Sub-model

In this research, the emphasis is laid on the surface mass moisture flux, which consists of a moisture emissivity coefficient, and is formulated after experimentation and much sensitivity analysis. A

value of 5×10^{-5} is given to this constant. The mathematical equation for moisture surface flux E_b is given by equation (10)

$$E_b = 5 \times 10^{-5} (RH_{\text{bound}} - RH_{\text{surf}}) \quad (10)$$

Where

RH_{bound} = relative humidity of the interior

RH_{surf} = humidity of the surrounding environment.

In the above equation (10), the moisture mass flux expression is simplified by considering 99.95% RH as water submergence conditions. The other way to simulate is to apply saturated pore pressure at the surface nodes of the elements. For simplicity, in this paper, the moisture migration results have been obtained from the simplified expression of equation (10)

4. EXPERIMENT

To verify the chloride surface flux model coupled with moisture mass surface flux model, a comprehensive series of experimentation is conducted.

4.1 Material and specimen preparation

Ordinary Portland cement, regular tap water, coarse sand and a W/C of 50% was provided to all specimens. The mix proportion of cement to sand was kept at 1:2.25. Cement content 580Kg/m^3 , water 290Kg/m^3 and sand 1305Kg/m^3 were used. Specimens consist of cylinders 50mm diameter and 100mm height. Curing was done for 28 days in sealed condition at 20 degree Celsius temperature. After curing, the top 10mm slice from the surface of all the specimens was removed to minimize the surface disturbances. Dry cutting was done for

Slicing of the specimens. Two levels of salt concentrations 3% and 6% by mass of water were selected by carefully considering the salinity of world oceans.

4.2 Specimen variables

For each exposure and time duration, same set of temperature and salt concentration is repeated as shown in table 1.1

Environment	Exposure	Temperature	NaCl
Submerged 33hr/9hr/1hr	1 month	20°C	3%
	7 months	20°C	6%
		40°C	3%
		40°C	6%

Table 1.1 Specimen identification table

4.3 Specimen Preparation for Chloride Testing

For the determination of chloride profiles, specimens were tested after 1 and 7 months exposures by potentiometer titration technique. In this experiment, slicing method was used for the determination of chloride contents. Thickness of each slice ($10 \pm 2 \text{mm}$) was measured and specimen loss due to the blade thickness of the cutter was accounted for in measuring the depth of each slice from the surface. After this, the slices of respective specimens were grounded and tested for the chloride contents according to the relevant ASTM standard C1152/C1152M-04 (for acid soluble chloride).

4.4 Environment Exposure Cycle

After removing the top surface slice, all specimens were exposed to laboratory controlled simulated marine tidal environmental conditions. For this purpose, four hygral cycles were designed. The following environmental cycles was applied

Cycle-A

Temperature: $T = 20^{\circ}\text{C}$ and 40°C (Constant)

Duration of cycle: 3days

Wetting period: 1hr

Drying period: 71hrs

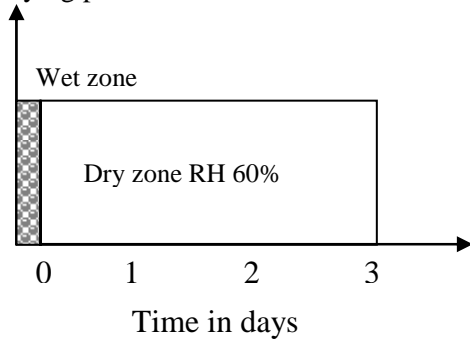


Fig 1.3 1hr wet in 3days

Cycle-B

Temperature: $T = 20^{\circ}\text{C}$ and 40°C (Constant)

Duration of cycle: 3days

Wetting period: 9hrs

Drying period: 63hrs

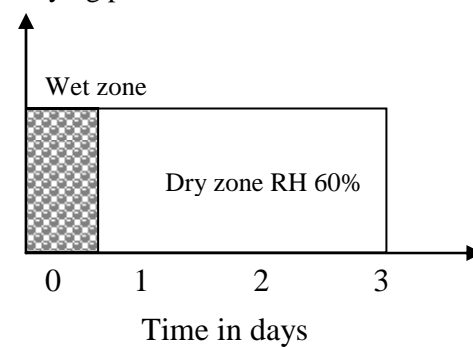


Fig 1.4 9hr wet in 3days

Cycle-C

Temperature: $T = 20^{\circ}\text{C}$ and 40°C (Constant)

Duration of cycle: 3days

Wetting period: 33hrs

Drying period: 39hrs

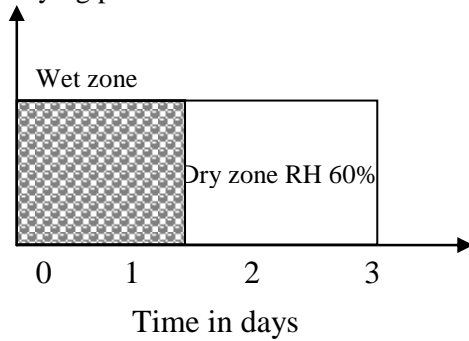


Fig 1.5 33hr wet in 3days

Cycle-D

Temperature: $T = 20^{\circ}\text{C}$ and 40°C (Constant)

Completely submerged

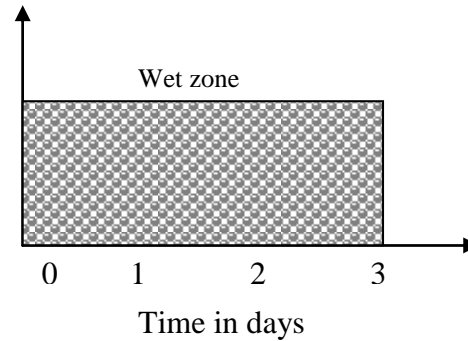


Fig 1.6 Complete submerged case

5. ANALYTICAL BOUNDARY CONDITION

The boundary conditions in the modeling were simulated as precise as in the experiment. In the modeling, only one surface was exposed to the environment to simulate the sealed specimen with one face exposed. For the initial curing period the moisture flux in and out of the specimen was restricted, whereas the heat flux was allowed during curing and environment exposure conditions. The submerged wetting cycle was modeled by providing

99.95% Relative humidity, and drying environment corresponds to 60% RH.

6. EXPERIMENT RESULTS AND DISCUSSION

Experimental and analytical results of chloride transport profile by DuCOM are presented and compared for verification of the model. Due to space limitation, chloride penetration results for 3% NaCl by weight of water are not shown in this paper.

6.1 Chloride Penetration Experiment Results

6.1.1 Submerged case: 1 and 7 months exposure

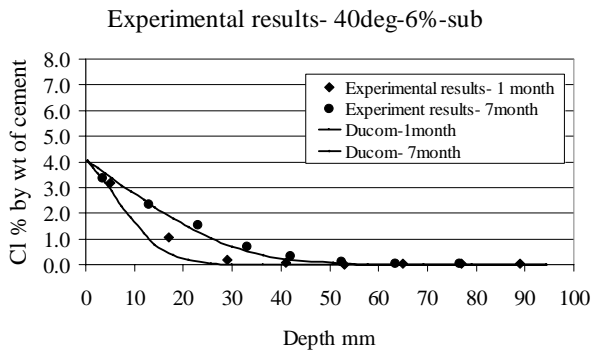


Fig 1.7 Results for chloride penetration profiles for 40°C and 6%NaCl- Experiment and analysis comparison

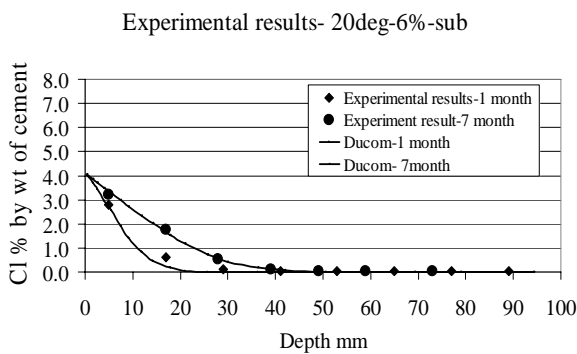


Fig 1.8 Results for chloride penetration profiles for 20°C and 6%NaCl- Experiment and analysis comparison

The experiment results show very good agreement with the analytical model results of DuCOM for 20°C and 40°C with 6%NaCl exposure environment as shown in figures 1.7 and 1.8. The good prediction by the analysis can be seen both for 1 month as well as 7 months environmental exposures. Since completely submerged case corresponds to the pure diffusion, therefore the validity of model in diffusion-dominated environments is confirmed. Same trend was observed in case of 3% NaCl concentration exposures.

When temperature affect was compared among the 20°C and 40°C graphs, it was observed that not so large difference is obtained. The temperature affect is simulated in the model by

introducing a temperature dependent diffusion coefficient D_{cl} . In DuCOM total diffusivity of the concrete is described as the product of D_{cl} and $\phi S \delta / \Omega$ as explained earlier in equation (3).

Temperature	D_{cl} (m^2/s)	$\phi S \delta / \Omega$	$D_{cl} \phi S \delta / \Omega$ (m^2/s)
20°C	$1.77E10^{-9}$	0.451761	$7.99E10^{-10}$
40°C	$3.00E10^{-9}$	0.356169	$8.78E10^{-10}$

Table 1.2 Temperature affect comparison table

The value of $D_{cl} \phi S \delta / \Omega$ for 40°C and 20°C does not differ so much. Thus the compensating affects of porosity and diffusion coefficient, shown by the experiment are nicely reproduced by the DuCOM analytical results, as shown in table 1.2.

6.1.2 33hr wetting case: 1 and 7 months exposure durations

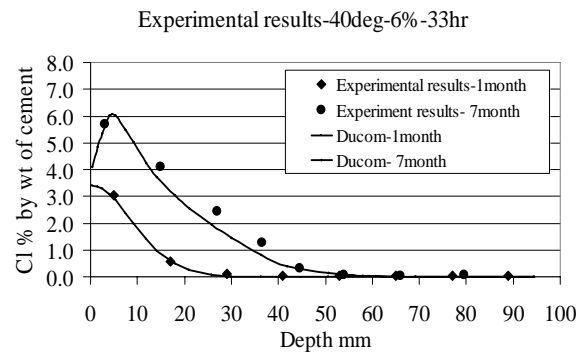


Fig 1.9 Results for chloride penetration profiles for 40°C and 6%NaCl- Experiment and analysis comparison

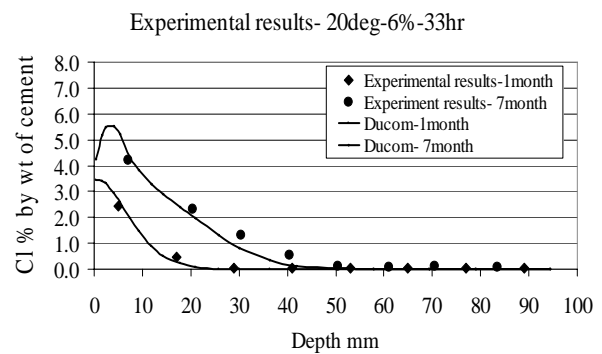


Fig 1.10 Results for chloride penetration profiles for 20°C and 6%NaCl- Experiment and analysis comparison

The results of experiment and model in figures 1.9 and 1.10 are in good agreement like that of submerged zone for 1month and 7month exposures. The same trend was observed for 3%NaCl environment exposures. In this case, the wetting time was nearly equal to that of the drying time, and diffusion and convection phenomenon worked simultaneously for the buildup of chloride ions. A visible peak is formed, which may be due to the simultaneous two-way movement of chloride ions during the drying period. Here the temperature affects become more pronounced as compared to submerged environment, due to the wetting and drying cycle, which generates strong moisture flux, responsible for the convection movements of chloride ions.

For 1month testing, the specimens were picked up after the end of drying period; however for 7months exposure time, the specimens were tested after end of wetting cycle. Both of the above mentioned testing conditions were very nicely predicted by the analytical results of DuCOM, which verify the ability of this model to predict under any variable hygral environment.

6.1.3 9hr & 1hr wetting case: 1 and 7months exposure durations

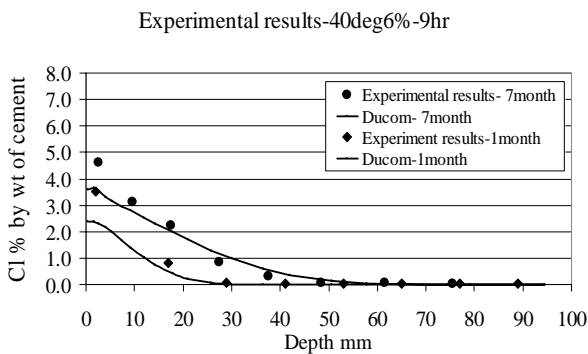


Fig 1.11 Results for chloride penetration profiles for 40°C and 6%NaCl- Experiment and analysis comparison

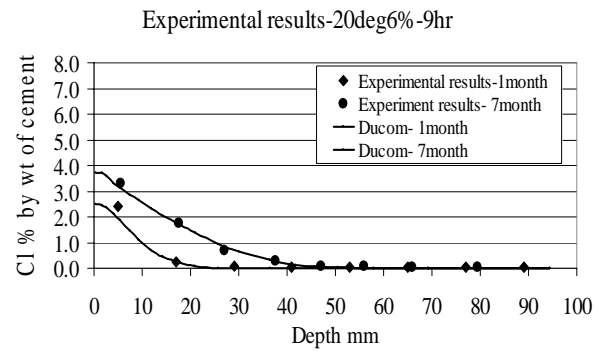


Fig 1.12 Results for chloride penetration profiles for 20°C and 6%NaCl- Experiment and analysis comparison

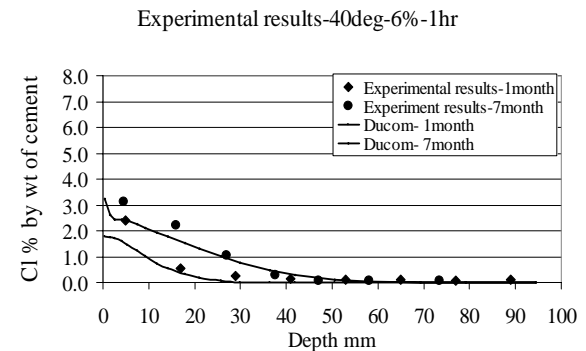


Fig 1.13 Results for chloride penetration profiles for 40°C and 6%NaCl- Experiment and analysis comparison

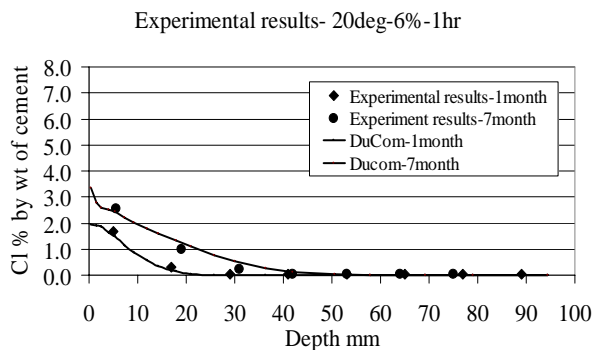


Fig 1.14 Results for chloride penetration profiles for 20°C and 6%NaCl- Experiment and analysis comparison

In these cases DuCOM prediction is also in good agreement with the experiment; however for high temperature environments, slight under-estimation is seen. Here only 6% NaCl results are shown, whereas 3% also follow the same trend. (Figures 1.11 to 1.14). In case of high temperature environment, the surface moisture flux in and out of the specimen is very strong as shown by the moisture migration results in the succeeding section 6.2. Therefore the high surface mass convection flux of moisture drags with it a large amount of chloride ions. However the analytical results does not show this strong

temperature affect as seen in the preceding graphs for 33hr wetting. This small under-estimation may be related to the fact that the moisture surface mass flux has been modeled by considering the simplified moisture mass surface flux equation (10), wherein the submergence is simulated by 99.95% RH, instead of the induced saturated pore pressure, at the surface elements at each cycle.

6.2 Moisture Migration Experiment Results

Moisture absorption of the specimens was also measured during drying and wetting cycles for 1hr, 9hr and 33hr, and the experimental values were compared with the analytical values computed by DuCOM.

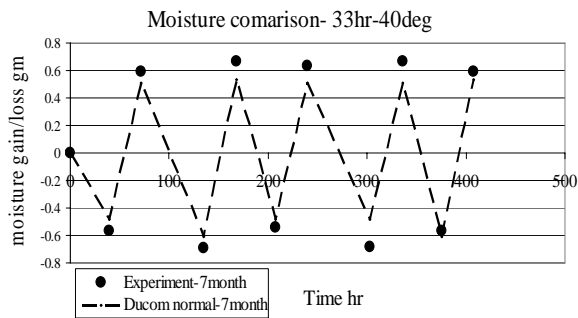


Fig 1.15 Results for moisture migration 33hr wet, 40°C during wetting and drying- Experiment and analysis comparison

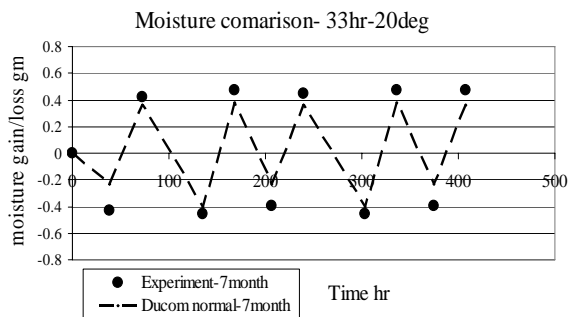


Fig 1.16 Results for moisture migration 33hr wet, 20°C during wetting and drying- Experiment and analysis comparison

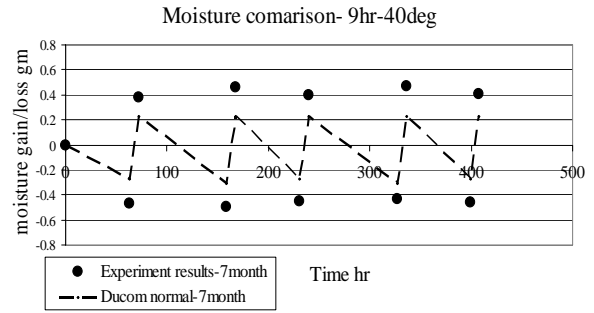


Fig 1.17 Results for moisture migration 9hr wet, 40°C during wetting and drying- Experiment and analysis comparison

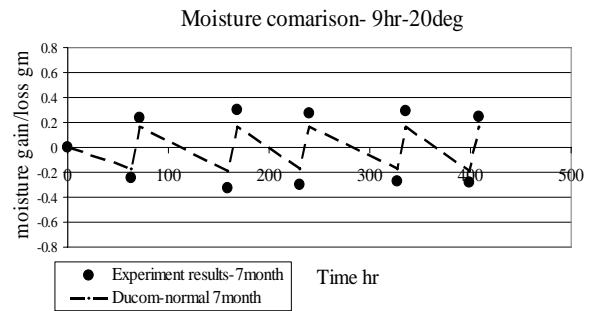


Fig 1.18 Results for moisture migration 9hr wet, 20°C during wetting and drying- Experiment and analysis comparison

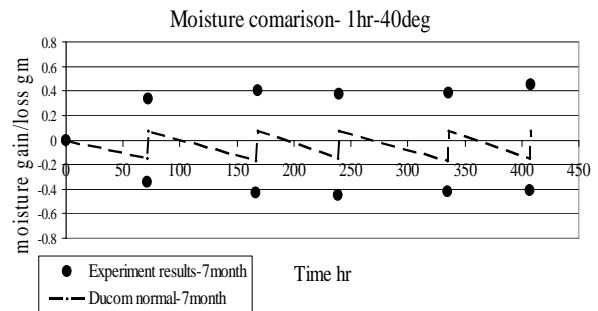


Fig 1.19 Results for moisture migration 1hr wet, 40°C during wetting and drying- Experiment and analysis comparison

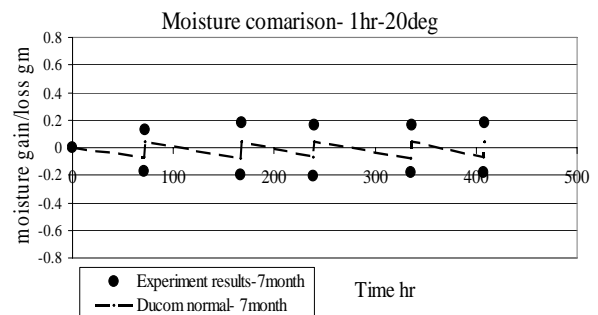


Fig 1.20 Results for moisture migration 1hr wet, 20°C during wetting and drying- Experiment and analysis comparison

The figures from 1.15 to 1.20 correspond to the moisture mass flux movement in and out of the specimen. The analytical results for moisture gain/loss were directly obtained in units of Kg/m^3 and converted to gm by multiplying it with the actual volume of the specimens used in this experiment. The zero on the y-axis does not correspond to the absolute saturation condition, and is taken as the starting point of drying cycle after 7 months exposure. The analytical results for 33hr wetting both for high and low temperature cases, are in good agreement with that of experiment. However for 9hr and 1hr wetting cycles, there is a slight under-estimation between experiment and analytical results. This could be due to the usage of simplified mass moisture flux equation (10) instead of applying saturated pore pressure as explained earlier.

7. CONCLUSIONS

The experimental results verify the DuCOM chloride surface flux model coupled with moisture surface mass flux model under varying wetting and drying environments. It may be noted that pure advective movement of chloride ions is difficult to study; since pure convection is not easy to obtain, as compared to pure diffusion (water submerged case). In all the wetting and drying cases the diffusion and convection affects takes place simultaneously and make thing complicated. The chloride distribution is simulated by directly coupling the chloride penetration with the moisture migration model, which is based on logical and physical laws of convection and diffusion movements.

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