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Diffusivity of saturated ordinary Portland cement-based materials: A critical review of experimental and analytical modelling approaches

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ABSTRACT

This paper provides a comprehensive overview of existing experimental and modelling approaches to determine effective diffusion coefficients of water saturated ordinary Portland cement-based materials. A dataset for diffusivity obtained from different experimental techniques have been presented for cement paste, mortar and concrete. For cement paste at low porosities, diffusivity reported by different authors varies up to a factor of five and electrical resistivity measurements for low capillary porosity are up to one order of magnitude higher compared to other techniques. Experimental data of mortar and concrete reveals predominant influence of increasing tortuosity due to aggregates and limited influence of interface transition zone. Hence, a particular emphasis has been placed on assessing predictability of diffusivity models for cement paste on a larger dataset collected in this paper. It has been observed that all predictive models have similar level of accuracy and fail to predict electrical resistivity data at low capillary porosity as these models are not calibrated using electrical resistivity data.

Contents

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Notations

A Cross-sectional area of the sample
a, a_1, a_2, a_3 Fitting parameter representing coefficients in different models
C Concentration
C_s Concentration at the upper compartment (inlet)
C_o Initial concentration in sample
C_b Bound concentration
D_a Apparent diffusivity (often referred to as non-steady state diffusivity in literature)
D_{CSH} Diffusivity of C-S-H phase
D_e Effective diffusivity (often referred to as steady-state diffusivity in literature)
D_p Pore diffusion coefficient
$D_e^p, D_{e,i}^p, D_e^{m/c}$ Effective diffusivity of cement paste in general, cement paste of the i^{th} layer and mortar/concrete respectively
D_e^{itz}, D_e^{agg} Effective diffusivity of ITZ and aggregates
$D_{e,n}$ Effective diffusivity of the composite with n layer spheres in case of multicoated sphere model
$D_{e,i}$ Effective diffusivity of i^{th} layer in case of transfer matrix method
D_{ea} Effective diffusivity of an equivalent aggregate system specific to [154] model
D_{hm} Diffusivity of hypothetical homogeneous medium containing equivalent aggregate system and cement paste specific to [154] model
D_{gp} Pore water diffusivity in gel pores
D_{HD-CSH}, D_{LD-CSH} Diffusivity of HD and LD C-S-H respectively
$D_{inn}, D_{int}, D_{out}$ Diffusivity of inner, intermediate and outer layer in case of multicoated sphere model
D_s Diffusivity of solid phase in generalized effective media theory
D_0 Diffusivity in capillary pore water
E_c total electric charge passed the sample
F Faraday number
H() Heaviside function
I current passed through the sample
J flux
J_{ss} Steady-state flux
L Length of sample
m Percolation exponent
M Molar mass of species
n Fitting parameter representing power in different models
Q Cumulative quantity at outlet
Q_c Charge passed
R Ideal gas constant
R_d Retardation factor
r_i Radius of i^{th} layer in case of transfer matrix method
r_{agg} Radius of aggregates
r_c Outer radius of the composite sphere applicable to transfer matrix method
t Time
t_i Components of the transfer matrix applicable to transfer matrix method
t_{itz} Thickness of ITZ
t_{lag} Time –lag (through-diffusion)
T Temperature
T Transfer matrix applicable to transfer matrix method
U Voltage
u Darcy velocity
w_{ea}, w_p Weighting factors related to equivalent aggregate system and cement paste, respectively
x Distance
z Valency number
ϕ Capillary porosity
ϕ_c Threshold capillary porosity
ϕ_{pores}^i Volume fraction of pores in i^{th} phase
ϕ_i^p Volume fraction of i^{th} phase in cement paste
σ_e, σ_0 Effective conductivity and conductivity of pore water respectively
ρ_e, ρ_0 Effective resistivity and resistivity of pore water respectively
$\phi_{gp}^{HD-CSH}, \phi_{gp}^{LD-CSH}$ Volume fraction of gel pores in HD C-S-H and LD C-S-H respectively
$\phi_{AF}^{inn}, \phi_{CH}^{inn}$ Volume fraction of capillary pores, Aluminate phases, portlandite and C-S-H phase respectively in inner layer in case of multicoated sphere models
$\phi_{CP}^{int}, \phi_{AF}^{int}, \phi_{CH}^{int}$ Volume fraction of capillary pores, Aluminate phases, portlandite and C-S-H phase respectively in intermediate layer in case of multicoated sphere models
$\phi_{CP}^{out}, \phi_{AF}^{out}, \phi_{CH}^{out}, \phi_{CSH}^{out}$ Volume fraction of capillary pores, Aluminate phases, portlandite and C-S-H phase respectively in outer layer in case of multicoated sphere models
$\phi_{paste}, \phi_{agg}, \phi_{itz}$ Volume fraction of paste, aggregates and ITZ respectively in mortar/concrete
ϕ_{tot} Total porosity in cement paste (gel pores + capillary pores)
$\phi_{ea}, \phi_{ea,c}$ Volume fraction and critical volume fraction of equivalent aggregate system, respectively
Ψ electrical potential
τ Geometric tortuosity factor
τ_a Apparent tortuosity factor
δ Constrictivity
θ Water content
ε Dielectric permittivity

1. Introduction

Chemical degradation processes of cement-based materials such as chloride-induced corrosion [1], sulphate attack [2,3], carbonation [4–6] and calcium leaching [7–10] are associated with the transport of solutes, either as ions or dissolved gases. One of the important transport mechanisms of solutes is diffusion which is driven by concentration gradients. The rate of diffusion is characterized by diffusion coefficient, commonly referred as diffusivity. Diffusivity is an important parameter for modelling processes related to contaminant transport in cementitious barriers, assessment of long-term behaviour of nuclear waste disposal systems based on cementitious engineered barriers and rebar corrosion in civil concrete structures [11]. Diffusivity is also used as a key durability parameter to define service life of concrete structures [12]. Diffusivity of a porous media at the macroscopic scale is given as [13]:

$$D_e = \frac{\theta D_0 \delta}{\tau^2} \quad (1)$$

where D_e is the diffusivity of ion/gas in porous media also known as effective diffusivity [$L^2\,T^{-1}$]; D_0 is the diffusivity of the solute in pore water [$L^2\,T^{-1}$]; θ is the volumetric water content which is equal to porosity for saturated media [−]; δ is the constrictivity factor [−] and τ is the geometric tortuosity factor [−]. The ratio (D_e/D_0) is commonly