Neutron imaging of moisture transport, water absorption characteristics and strength properties for fly ash/slag blended geopolymer mortars: Effect of drying temperature

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Project Title : Innovations of neutronics in geopolymeric blends for sustainable building applications

1- Cement industry consumes virgin natural resources and requires high energy during the manufacturing processes. Namely, ***the global statistics indicate that***

A- about 260 million tons of cement are needed annually for construction. Because limestone is the main source of ordinary Portland cement production, acute limestone shortages may occur within the next 25–50 years.

AA- Every single ton of cement requires roughly 1500 kg of raw ingredients

B- Cement based materials (industry) is anticipated to be the second highly used material globally, after water .

C- PC is the third highly energy-intensive material on the globe, after steel and aluminum products, accounting for 7% of all energy consumed by worldwide industry - Every single ton of cement requires roughly 80 kilowatt-hours of power

2- It results in an enormous carbon dioxide (CO_2) emission causing some environmental problems . It is true that ***the production of PC alone emits 1.35 billion tons of greenhouse emissions each year***

4- To achieve sustainability in building materials, the extraction and overuse of raw materials and carbon footprint associated with the production of cement should be reduced.

5- To develop an environmentally friendly building materials, wastes such as FA, GGBFS, rice husk, ….etc as well as metakaolin(MK) are the best candidate.

Therefore, one has to find alternative materials

Geopolymers

Geopolymers are

1- A low carbon binders with less energy requirement and CO_2 emission in comparison to cement-based materials.

2- Geopolymers are synthesized by mixing alkaline activator with alumino-silicate precursor (FA, slag , RHA, MK) to form a binding material.

3- Geopolymers in some cases are better than cement in strength, durability, and drying shrinkage and heat, acid and sulfate resistance

The problem

1- Moisture transport in construction building materials including cement and geopolymer- based materials is a crucial physical process for their service life and durability since water itself as well as biological and dangerous chemical substances transported by it can cause and induce a series of deterioration processes.

2- Moisture transport besides it dependence on the properties of the flowing liquid, it depends on both the pore size distribution and hydrophilicity of the porous matrix.

3- Geopolymer- based materials are characterized with rich ranges of pore sizes and porosities and their porous matrix have strong hydrophilicity.

4- The hydroxyl groups (^{-}OH) resulting from chemical activation of precursors during syntheses of geopolymers is responsible for the hydrophilicity of the geopolymer matrix.

Moisture uptake is prompted in these GPs materials.

1- Dissolved ions carried by water such as chlorides and sulphates accelerate dissolution of the alkali metal ions and cause corrosion of steel reinforcement.

2- Freeze–thaw cycles cause deterioration processes such as expansion, cracking, ….etc of the concretes.

3- Biofilms can be formed due to metabolic reactions of microorganisms with the porous matrix of the GPs. Microbial induced concrete degradation (MICD) which is enhanced when the GPs are in contact with water sources such as waste water.

4- Acids are formed due the dissolution of CO_2 and SO_2 gases, from the environment, in water of pore solution leading to acid attacks of the matrix and steel reinforcing the GPs (deterioration processes).

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5- Efflorescence is a very important damage processes that takes place frequently in GPs. Origin of efflorescence is the drying of wetted or partially wetted GPs. During drying of GPs , soluble alkali metal ions can be carried by evaporated water to their outer surfaces. The soluble ions diffused to the surface may react with the Co_2 from atmosphere leading to formation of carbonated products such as sodium carbonate.

6- Moisture variation (gradient) in mortars and/or concretes due to water absorption and/or drying is the primary cause of deformations (drying shrinkage and drying creep). Moisture occupying pore space induces stresses on the matrix of mortars and/or concretes. So, moisture gradient in the pore space causes stress gradients, resulting in cracking.

7- Cracks induced by drying facilitate absorption of moisture carrying aggressive chemical compounds which may impact durability. Therefore, because of its importance for durability of GPs, capillary moisture absorption and distribution in Gps should be of prime concern.

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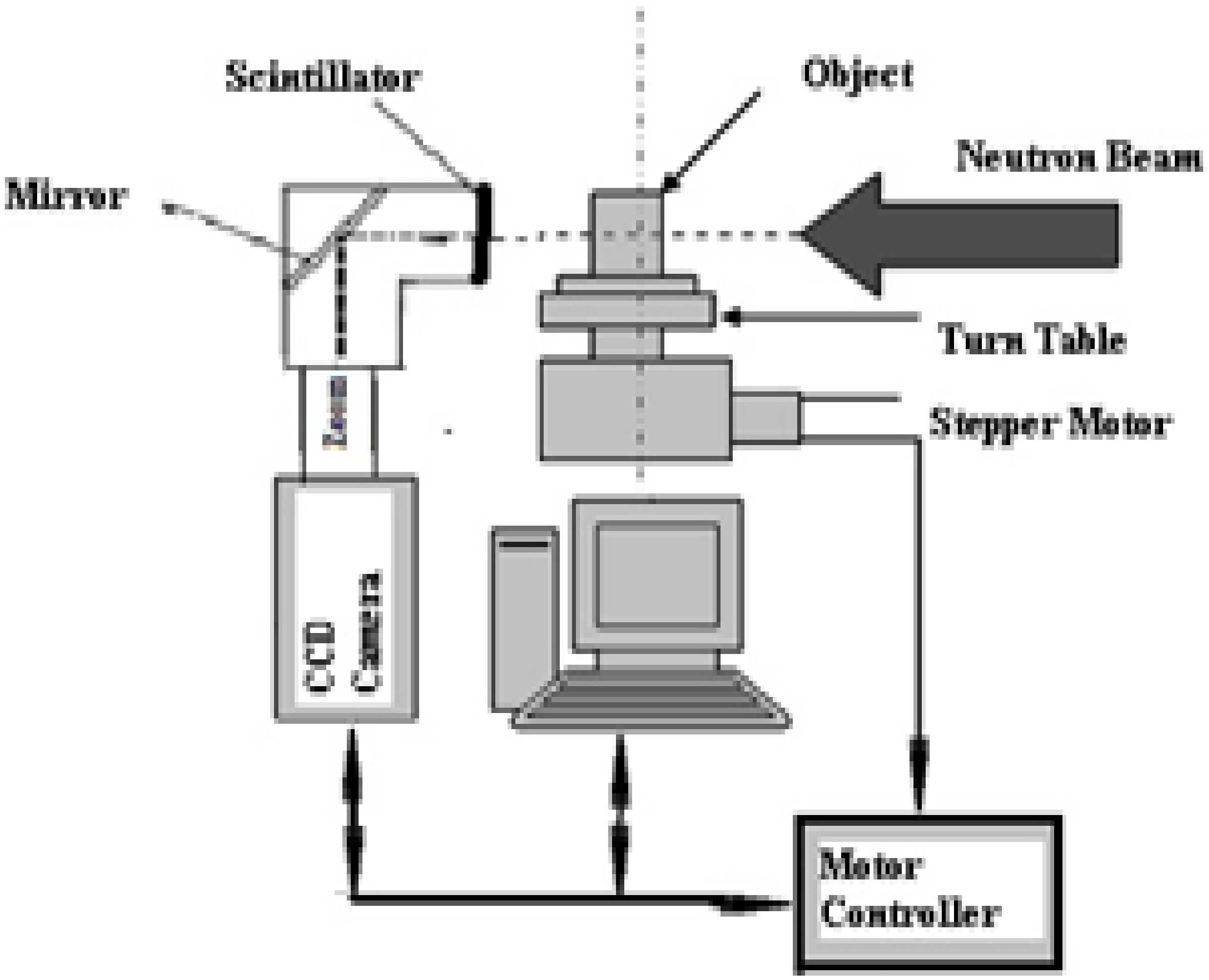
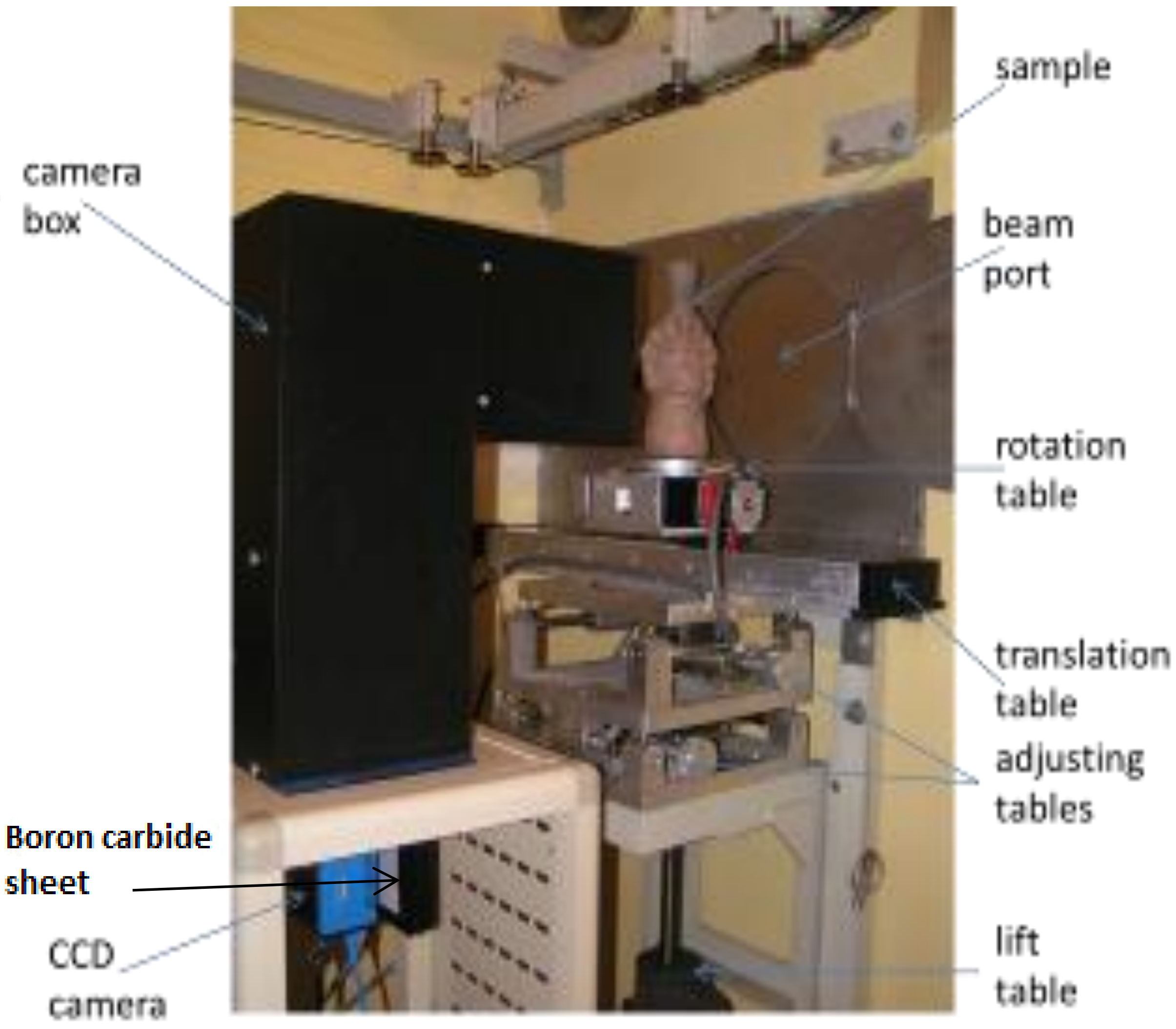
Neutron radiography (NR) is a powerful non-destructive method for measuring spatial water distribution in many porous materials.

The thermal neutrons crossing the sample are attenuated because of their absorption and scattering by nuclei of elements of the sample investigated. Since water or moisture contains hydrogen atoms that have a large scattering cross-section, water content and distribution in any porous sample can be determined using NR.

The Neutron radiography and tomography At ETRR-2





Characteristics of NR/T at ETRR-2

Dose rate intensity at beam outlet	*3.3 Sv/hr at 13.3 M watt thermal*
Thermal neutron flux	*8.6* $\times 10^{7}$ *n/cm*^{2}*. sec*
Fast neutron flux	*1.6* $\times 10^{7}$ *n/cm*^{2}*. sec*
Cd ratio	*10*
L/D ratio	*117*
Facility resolution	*188* μ*m*

Detector Components

1-Al-Box: light tight -box, 2- Scintillation screens (ZnS (Ag)-6LiF) 3- Mirror: 2 mm thick glass plate coated with Al and TiO2 , 3- Pco.2000 CCD camera system

Aim and motivation

1- Many durability issues were studied for FA/slag-based GPs; however, studies on water absorption in these GPs focused on the measurements of sorptivity, water absorption, porosity, and volume of permeable voids

2- The impact of drying temperature and percentage of slag in FA-based GPS on capillary water absorption was not studied before using NR. Such a study is needed and can be used in durability and service life models and the performance evaluation of GPs.

Therefore,

1- The effect of the percentage of slag in FA/slag-based geopolymer mortars and the impact of drying temperature on the capillary water absorption process is studied using NR

2- The NR experiments revealed the visualization of the water absorption processes. Water content distribution, sorptivity and capillary penetration coefficient for the FA/slag-based- GPs were determined from the NR measurements.

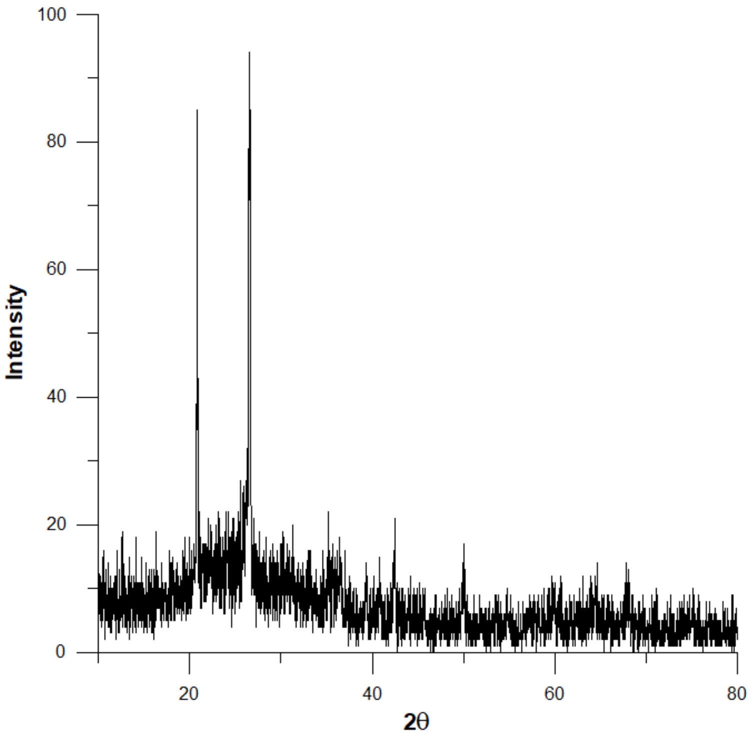
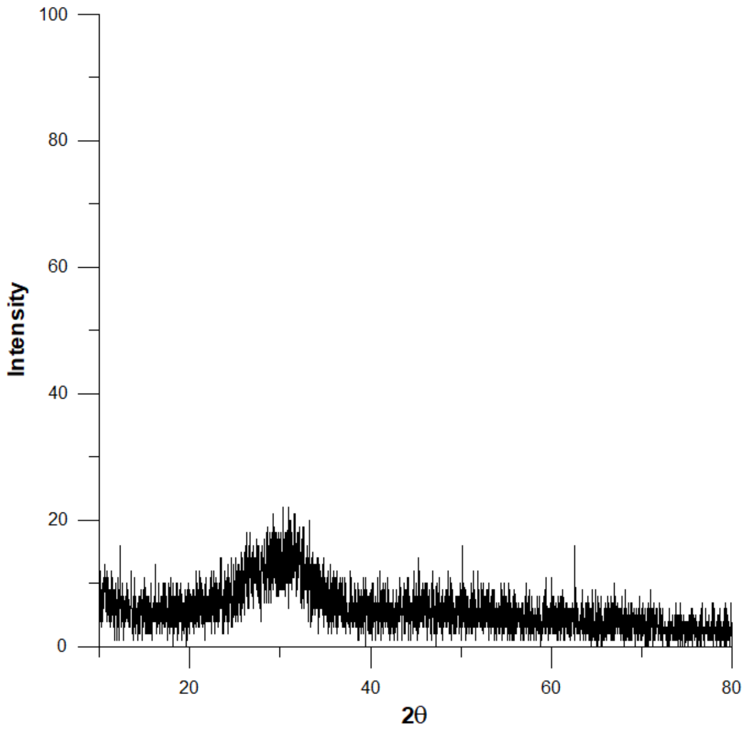
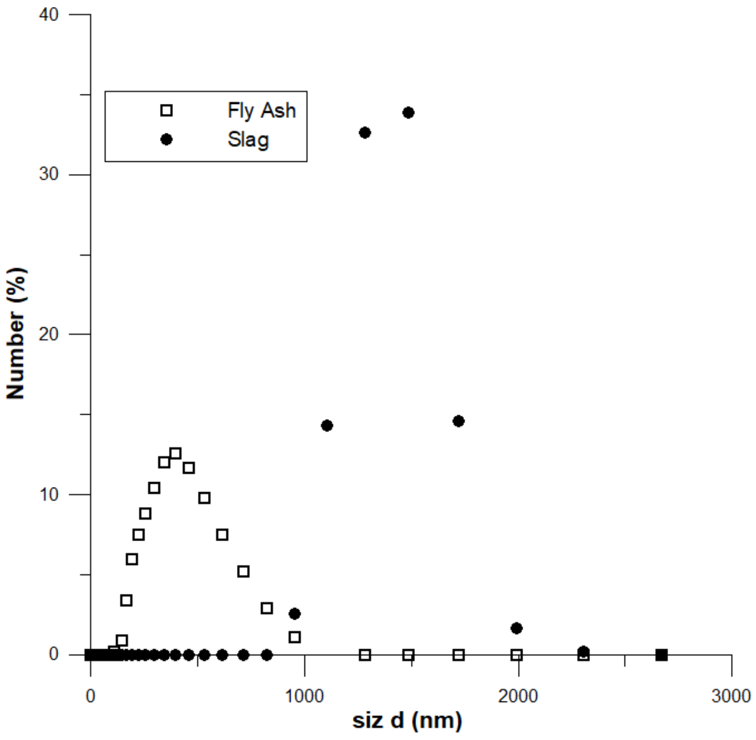
3- Modelling the NR results obtained in terms of the second Fick's law of diffusion and determining water diffusivities were established. The effect of the percentage of slag in FA/slag-based geopolymer mortars on water absorption, volume of permeable voids, sorptivity and mechanical properties were determined.

Sample preparation

- Fly ash -based geopolymer mortar containing 0, 10,20,30,40 and 50% slag were prepared and cured at room temperature.

Chemical compositions of fly ash (FA) and ground granulated blast furnace slag (slag)

Oxide	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	MnO	MgO	CaO	P_2O_5	Na_2O	K_2O	Cl	SO_3	LOI
Fly ash (%)	56.70	36.40	2.64	1.22	0.038	0.818	0.717	0.400	0.549	<0.01	<0.01	0.34	
Slag (%)	37.87	14.01	2.67	0.60	0.44	8.38	33.04	<0.01	0.17	0.97	<0.01	1.31	0.30

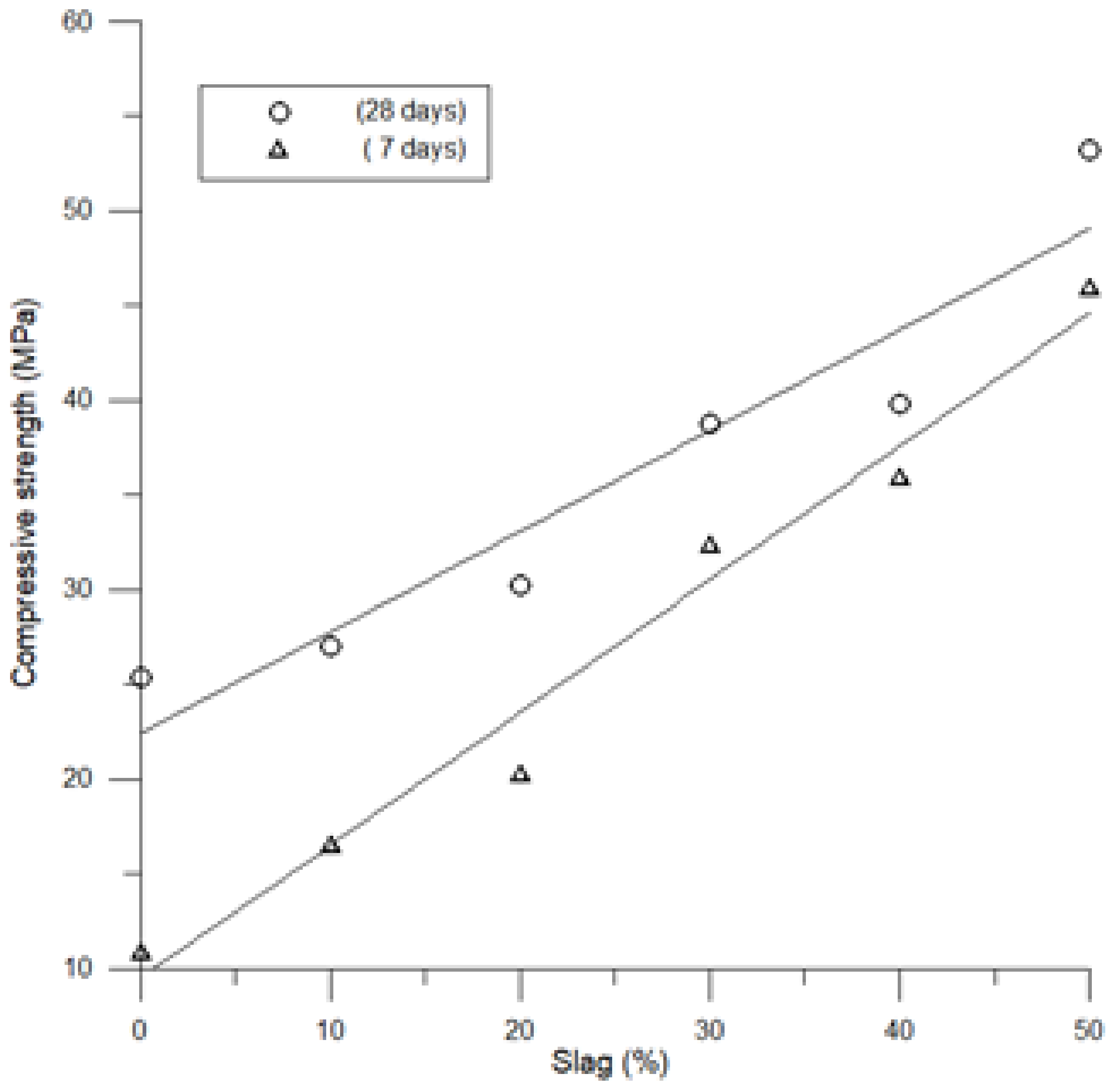
		
XRD for FA	XRD for slag	Particle size distribution for FA and slag

Mechanical tests

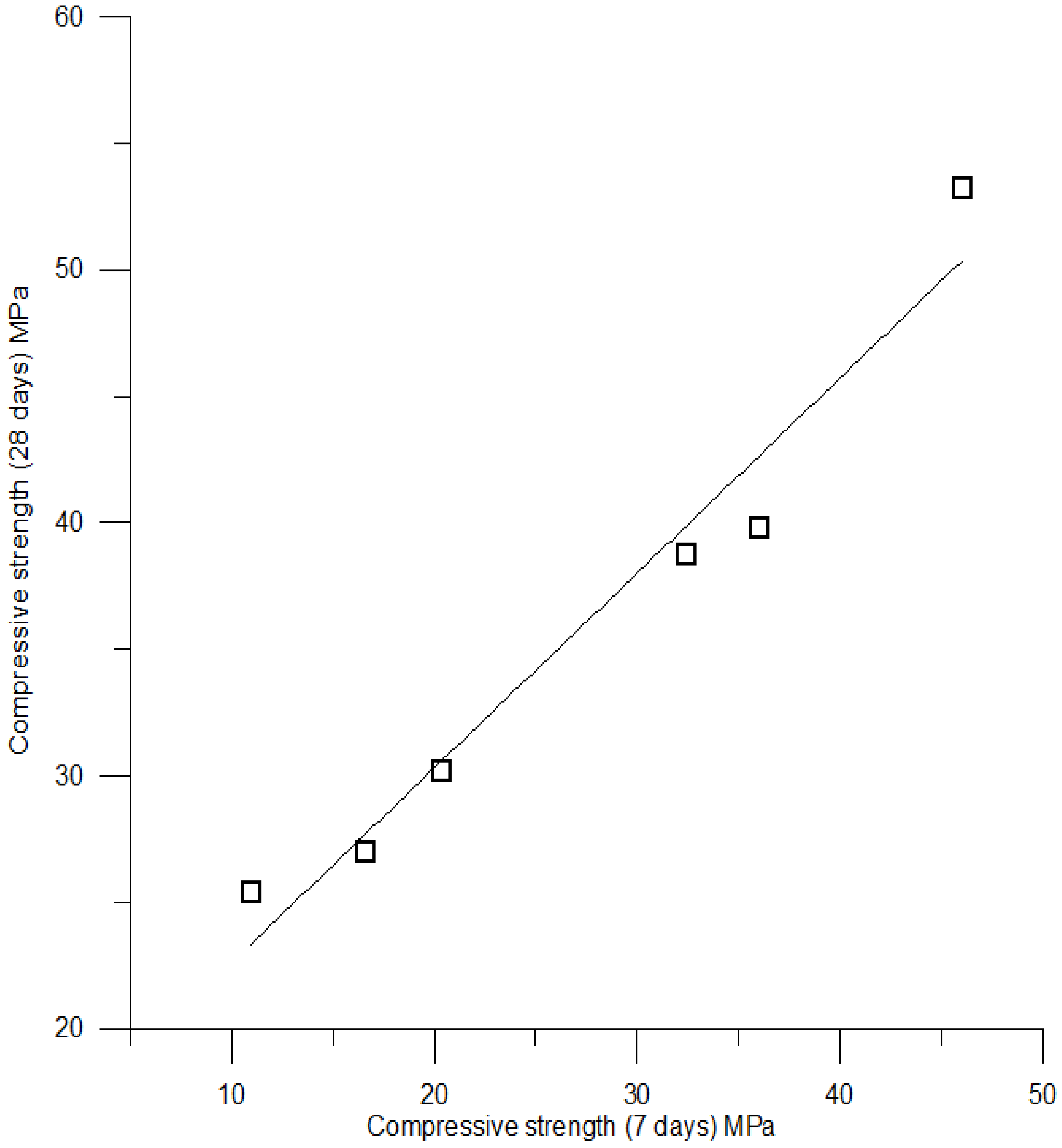
Flexural test

Compression test

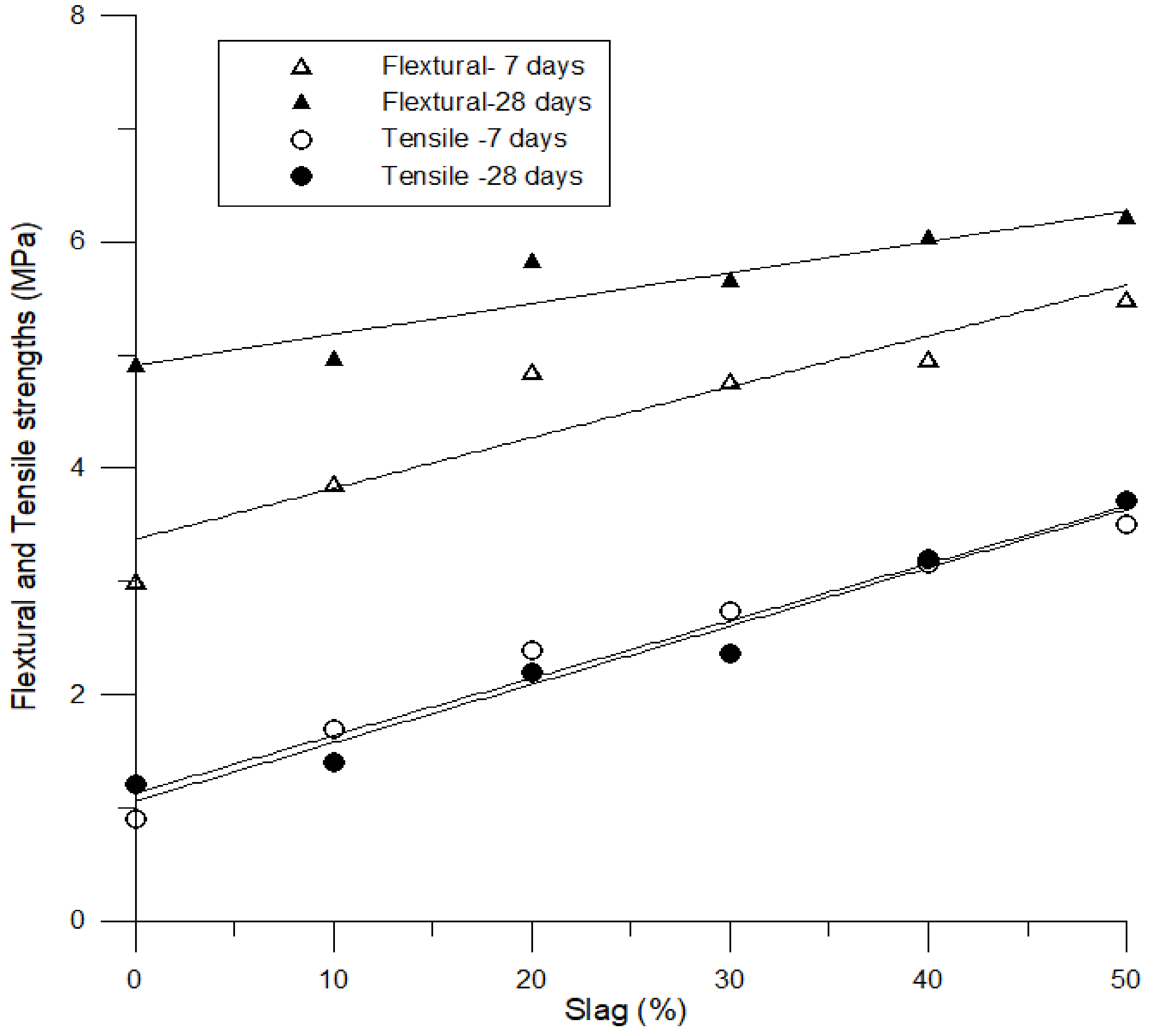
Splitting tensile test

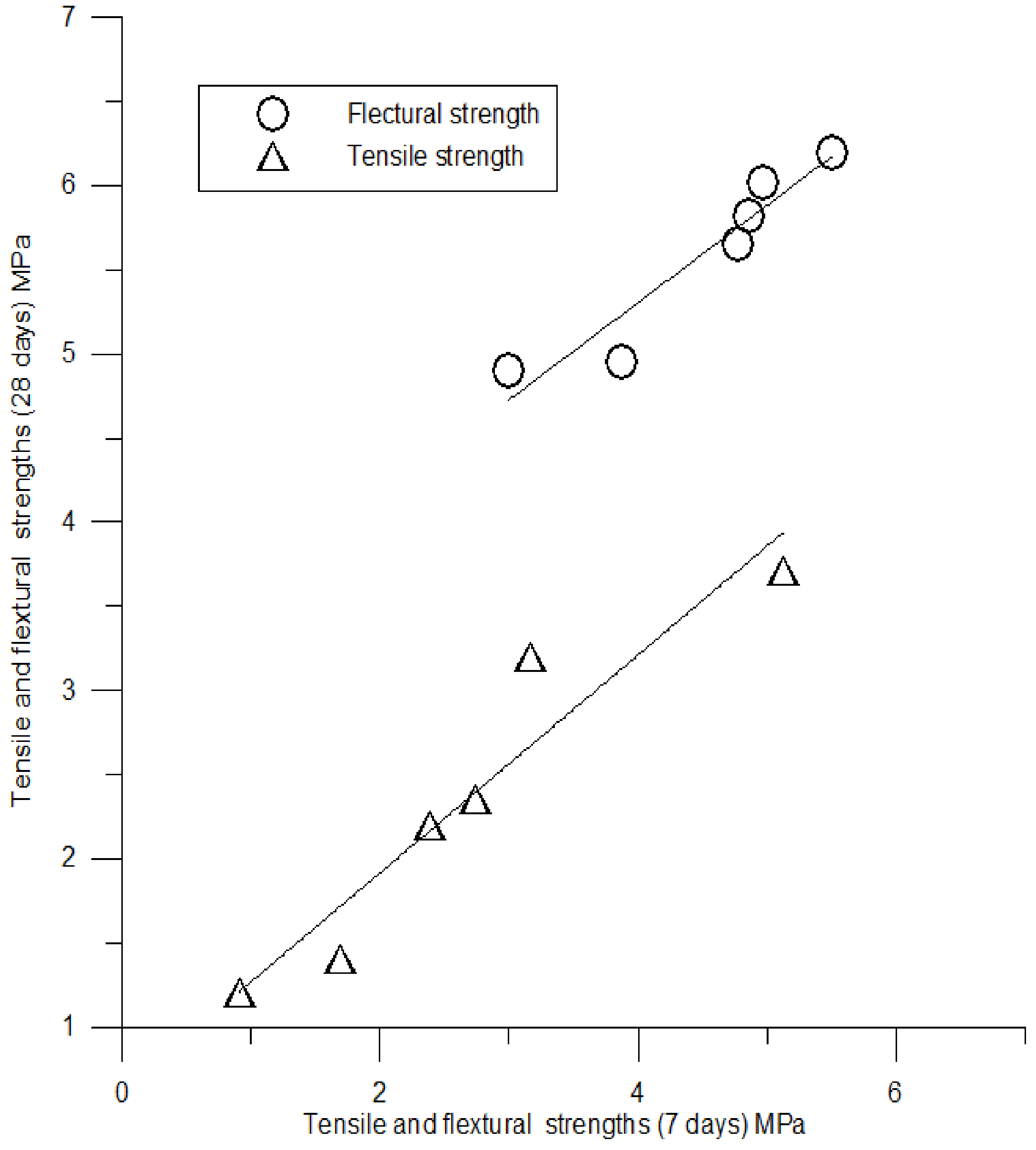


Compressive strength (MPa)
Slag (%)
(28 days)
(7 days)



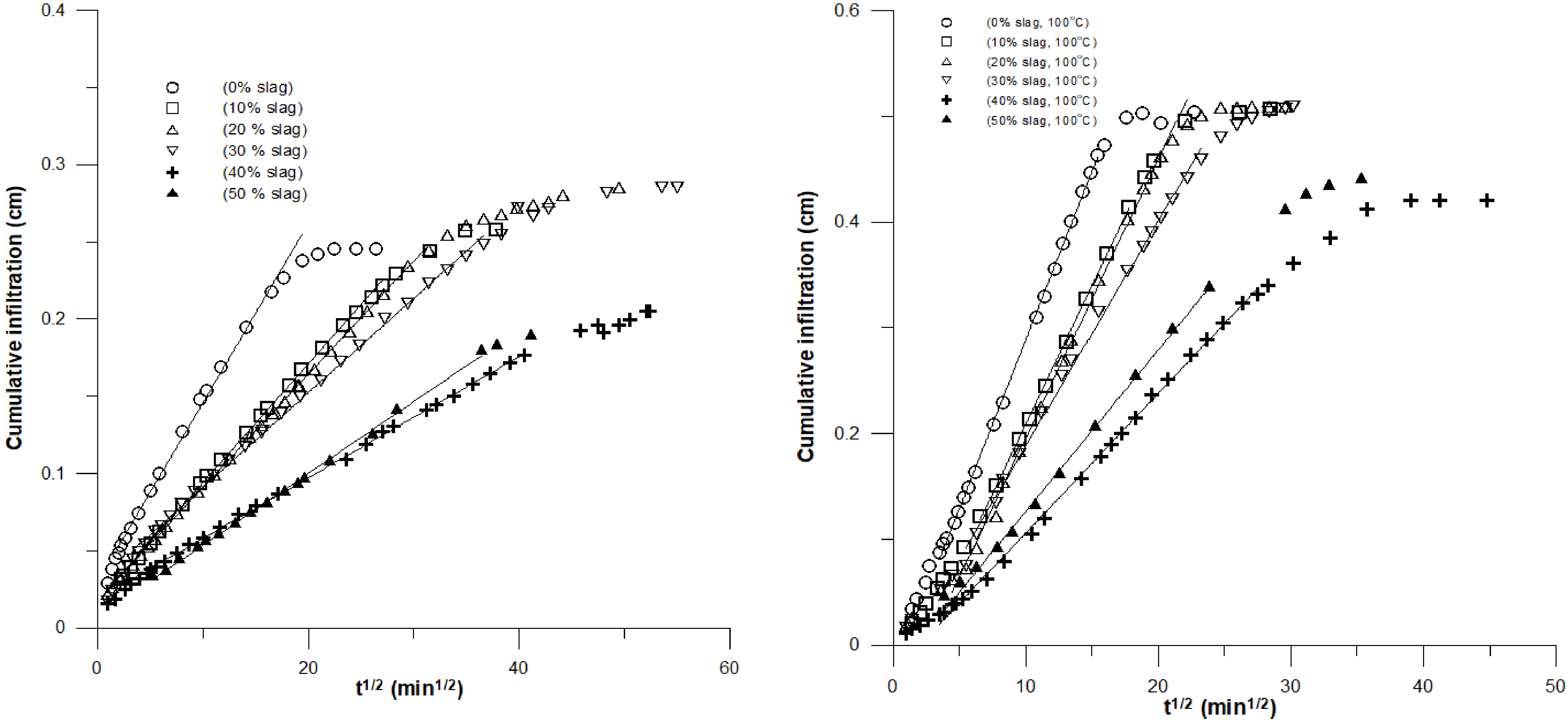
Compressive strength (28 days) MPa
Compressive strength (7 days) MPa


Flextural- 7 days
Flextural-28 days
Tensile -7 days
Tensile -28 days
Flextural and Tensile strengths (MPa)
Slag (%)
0
10
20
30
40
50
0
2
4
6
8

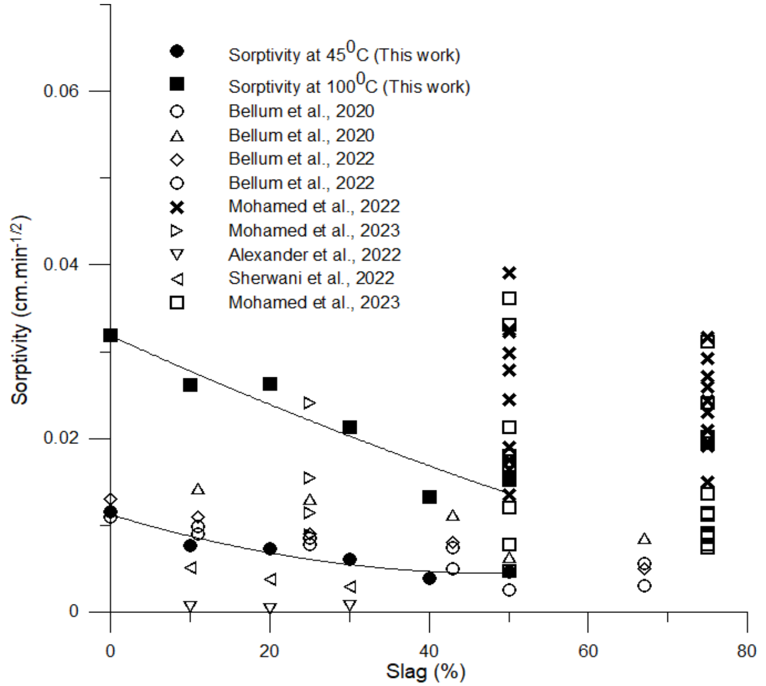

Flectural strength
Tensile strength
Tensile and flextural strengths (28 days) MPa
Tensile and flextural strengths (7 days) MPa
0
2
4
6
1
2
3
4
5
6
7

Durability : Capillary moisture absorption (sorptivity)

Cumulative water absorption I (cm) can be determined as follows $I = W/A = St^{1/2} + a$

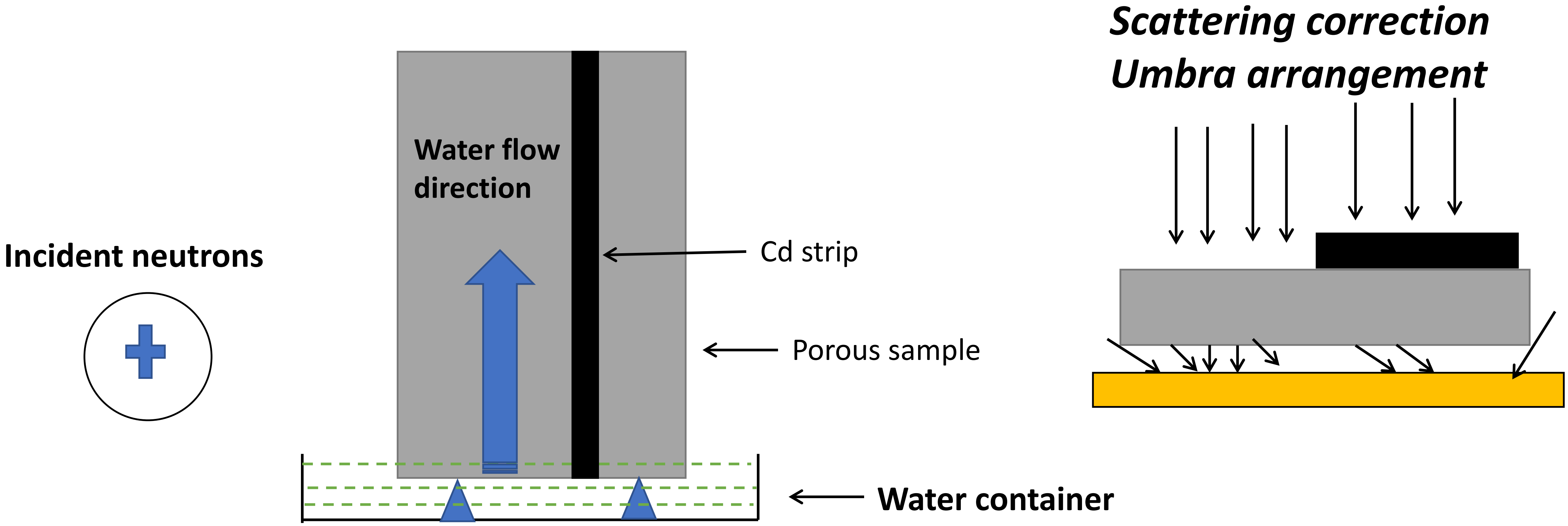


Sample	Sorptivity x10^{-3} (cm.min$^{-1/2}$)			Water absorption, W_1 (%)	Water absorption after immersion boiling W_2(%)	Volume of permeable voids, VBV (%)	Dry bulk density, D (gm/cm^3)
	45^{o}C	100^{o}C	S_{100}/S_{45}				
F100G0	11.60	31.88	2.75	4.95	7.82	16.32	2.09
F90G10	7.70	26.23	3.41	4.81	6.92	14.61	2.11
F80G20	7.33	26.14	3.57	4.7	6.61	13.98	2.11
F70G30	6.12	21.27	3.48	4.6	5.91	12.92	2.19
F60G40	3.92	13.21	3.37	4.28	5.12	11.00	2.21
F50G50	4.65	15.26	3.28	3.93	4.55	10.28	2.26

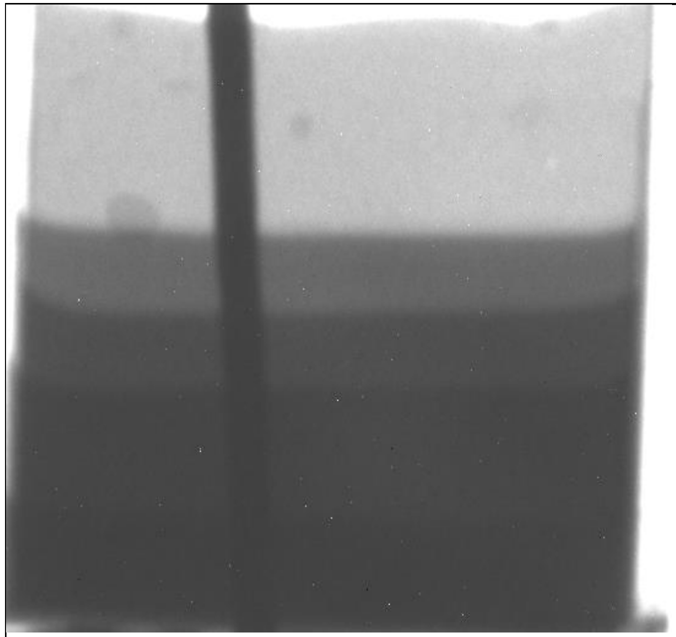


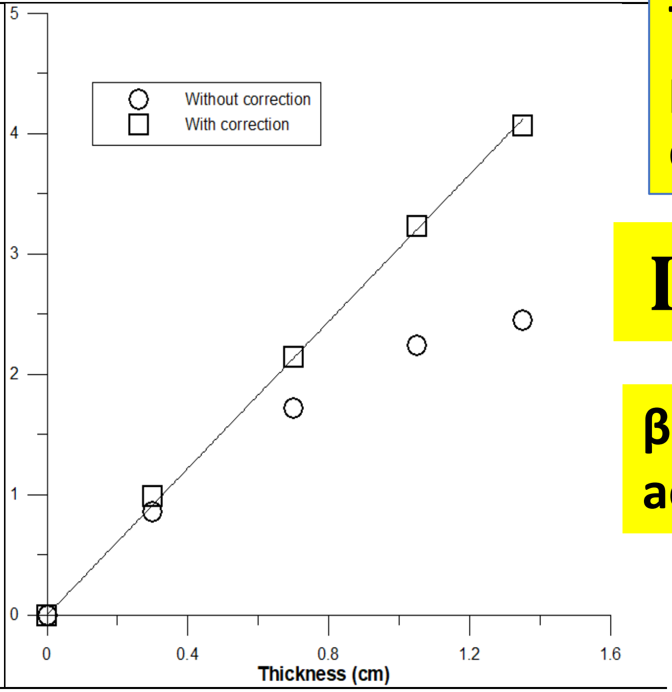
$$S_{100} = 2.5xS_{45} + 5.13$$

Neutron imaging experiment



As the absorption time elapses, water is absorbed by capillary forces in the porous samples.





The transmission of neutrons through a porous sample containing water can be described using the Lambert-Beer law

$$\mathbf{I_w = I_o \ exp(- \textstyle\sum_w x_w - x^2 \beta)}$$

β is a correction parameter which takes into account the neutron scattering

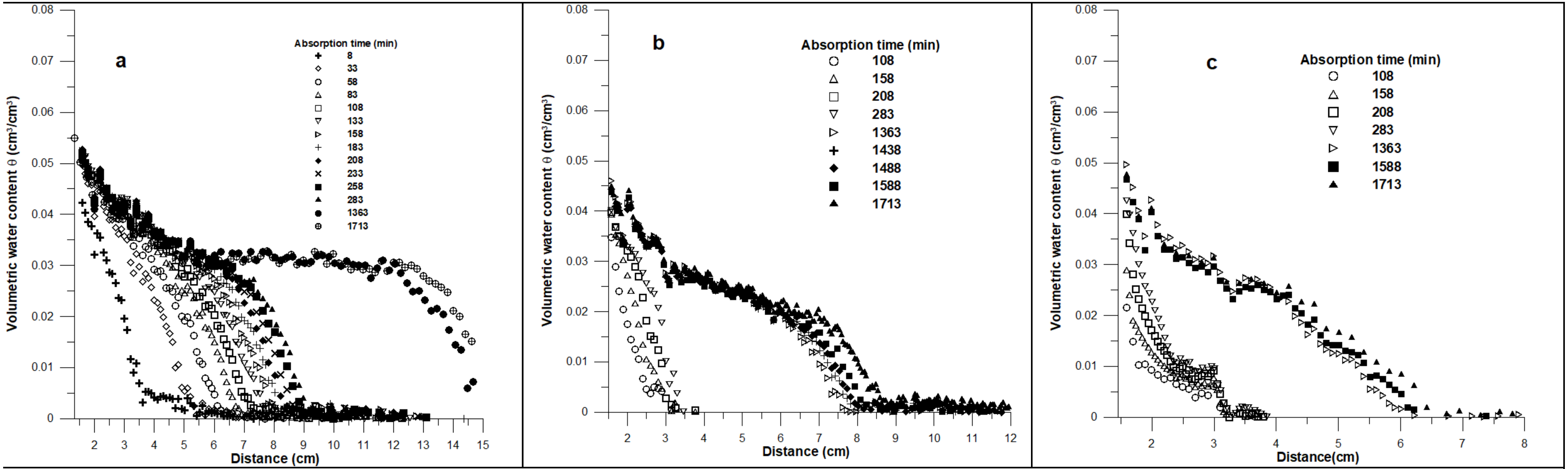
$$x_w = -\frac{\Sigma_w}{2\beta} - \sqrt{\left(\frac{\Sigma_w}{2\beta}\right)^2 - \frac{1}{\beta} ln(\frac{I_w}{I_o})}$$

$$\boldsymbol{\theta(x,t) = \frac{x_w(x,t)}{L_b}}$$

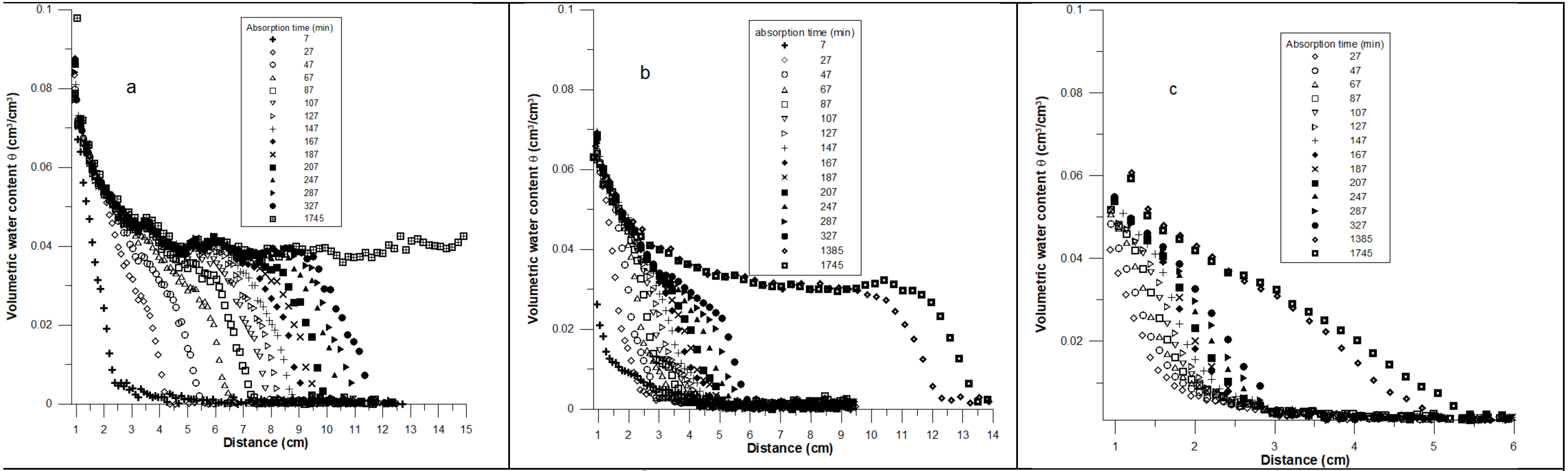
time	0 % slag	10% slag	20 % slag
7 min			
158			

283			
1713			

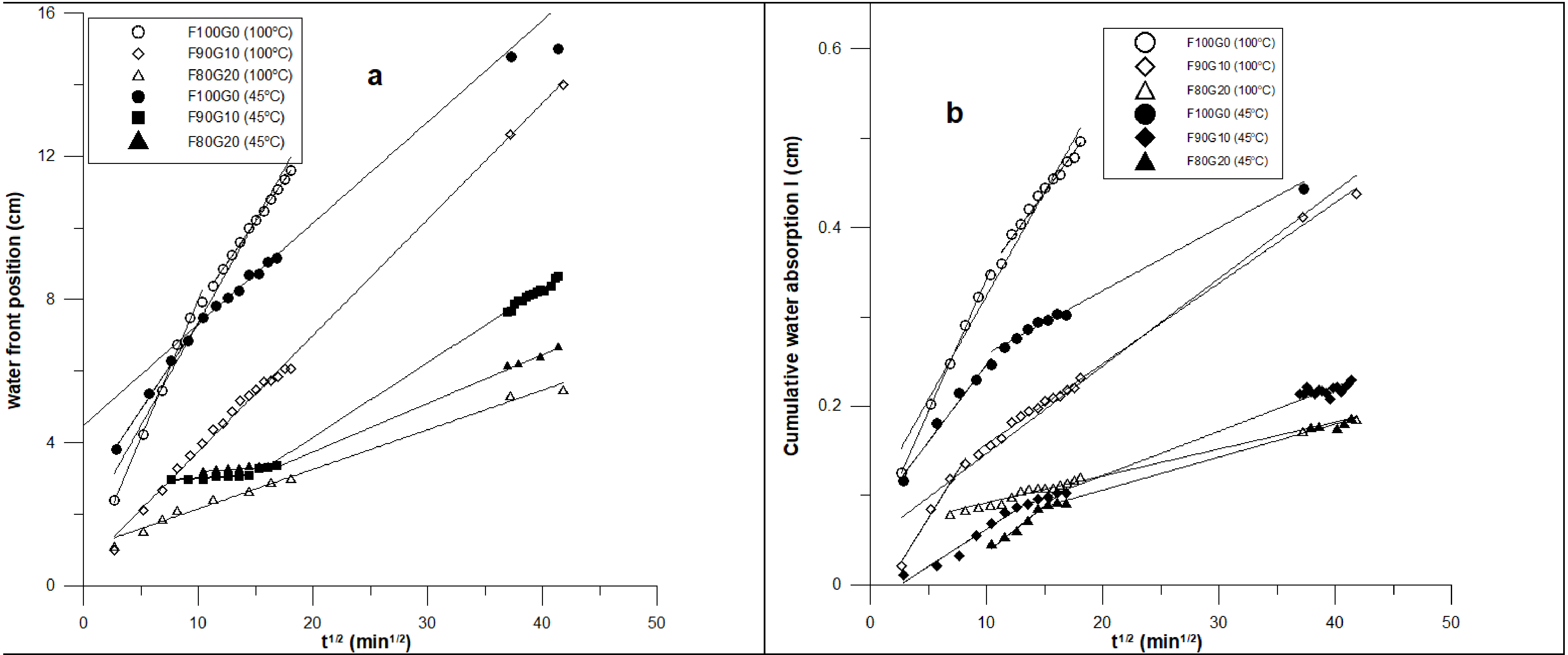
Processed NR images – we see only water. The samples were canceled



Water profiles for the mortar samples dried at 45^{0}C: a) F100G0, b) F90G10 and c) F80G20.



Water profiles for the mortar samples dried at 100^0C: a) F100G0, b) F90G10 and c) F80G20.



Progress of waterfront positions a) and cumulative water absorption I (b) versus the square root of absorption for fly ash- based mortar containing 0, 10, and 20% slag dried at 45 ^{0}C and 100 ^{0}C along with fit lines of straight-line equation

Sample	$k \times 10^{-3}$ (cm/min$^{1/2}$)		$S \times 10^{-3}$ (cm/min$^{1/2}$)	
	k_{NR45}	k_{NR100}	S_{NR45}	S_{NR100}
F100G0	266 (481.5, 282.3)	572.7 (768.5, 465.2)	17.22, 7.07	23.2 (29.45, 18)
F90G10	(20.5, 208.2)	324	8.38, 4.96	9.8 (20.9, 9)
F80G20	(19.8,135.6)	110.5	9.39, 3.81	3.01

The NR results obtained confirm the gravimetric measurement results. The k and S-values follow the same trend as the values of sorptivity, water absorption, water absorption after immersion and boiling, and volume of permeable voids for fly ash/slag mortars. All these parameters decreased and increased as the slag content in the samples investigated and drying temperature increased, respectively

Validity of the second Fick's law of diffusion

The assumptions required for modelling water absorption processes in porous media based on the second Fick's law of diffusion are:

1) homogeneous porous medium, 2) the water absorption in one dimension and into a semi-infinite porous medium with a constant concentration boundary condition, (3) homogeneous distribution of the initial moisture content, and (4) negligible gravitational effects

These assumptions were fulfilled in the present study since

1- the samples investigated seemed to the naked eye homogenous,

2- the water absorption process was in one dimension,

3- the distribution of the initial moisture content is homogeneous because the sample sizes are small and gravity forces can be neglected for many building materials,

4- and applying Fick's law of diffusion for modelling the results obtained was justified since the cumulative water absorption I (cm) and waterfront position followed the $t^{1/2}$ -scaling.

Theory of unsaturated moisture flow

The second Fick's law of diffusion, which describes moisture and water absorption in partially saturated porous media can be used to justify the $t^{1/2}$ -scaling of cumulative water absorption and waterfront position. It can be described in one dimension x by the partial differential equation (neglecting the gravity):

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}\left(D(\theta)\frac{\partial\theta}{\partial x}\right)$$

The diffusivity is a function of the physical properties of the moving liquid, such as contact angle, viscosity, and surface tension and the porous matrix

Assuming that the Boltzmann variable ($\phi = \frac{x}{\sqrt{t}}$) is valid, namely waterfront, x scales with $\sqrt{t}$, water diffusivity can be obtained as

$$D(\theta) = -\frac{1}{2}\frac{d\phi}{d\theta}\int_0^{\theta}\phi\, d\theta$$

The (θ -ϕ) profiles must converge to a single master curve under the assumption that $x = \phi\sqrt{t}$. Assuming the validity of the Boltzmann transformation, the diffusion-based approach can be used to model water absorption in porous materials.

However, in particular porous materials, liquid transport processes can deviate from $\sqrt{t}$- scaling. In this case, anomalous diffusion can be utilized as an alternative method. According to the anomalous diffusion approach. the general form of equation is:

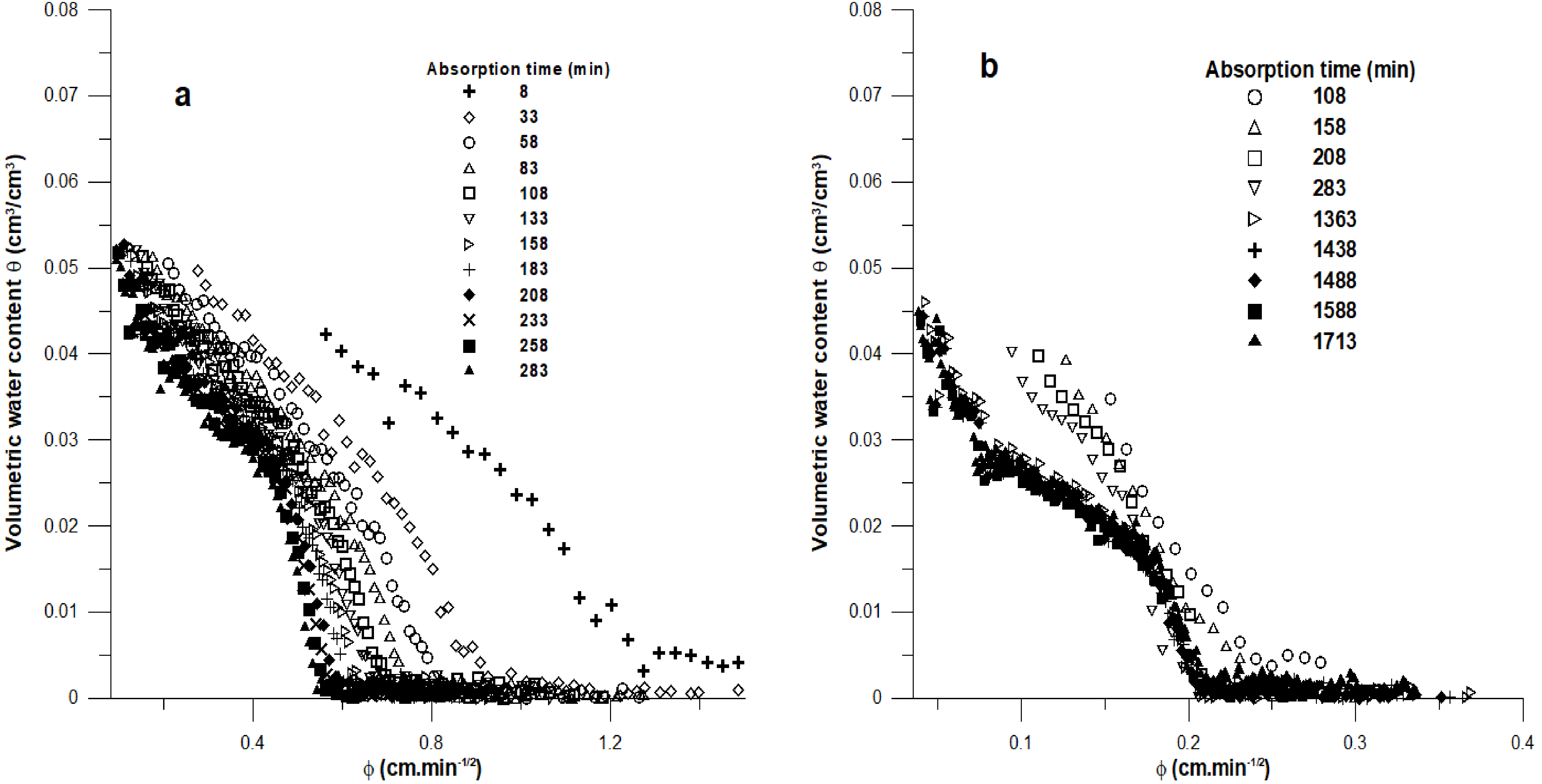
$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}\left(D(\theta)\left|\frac{\partial\theta}{\partial x}\right|^{\alpha}\right)$$

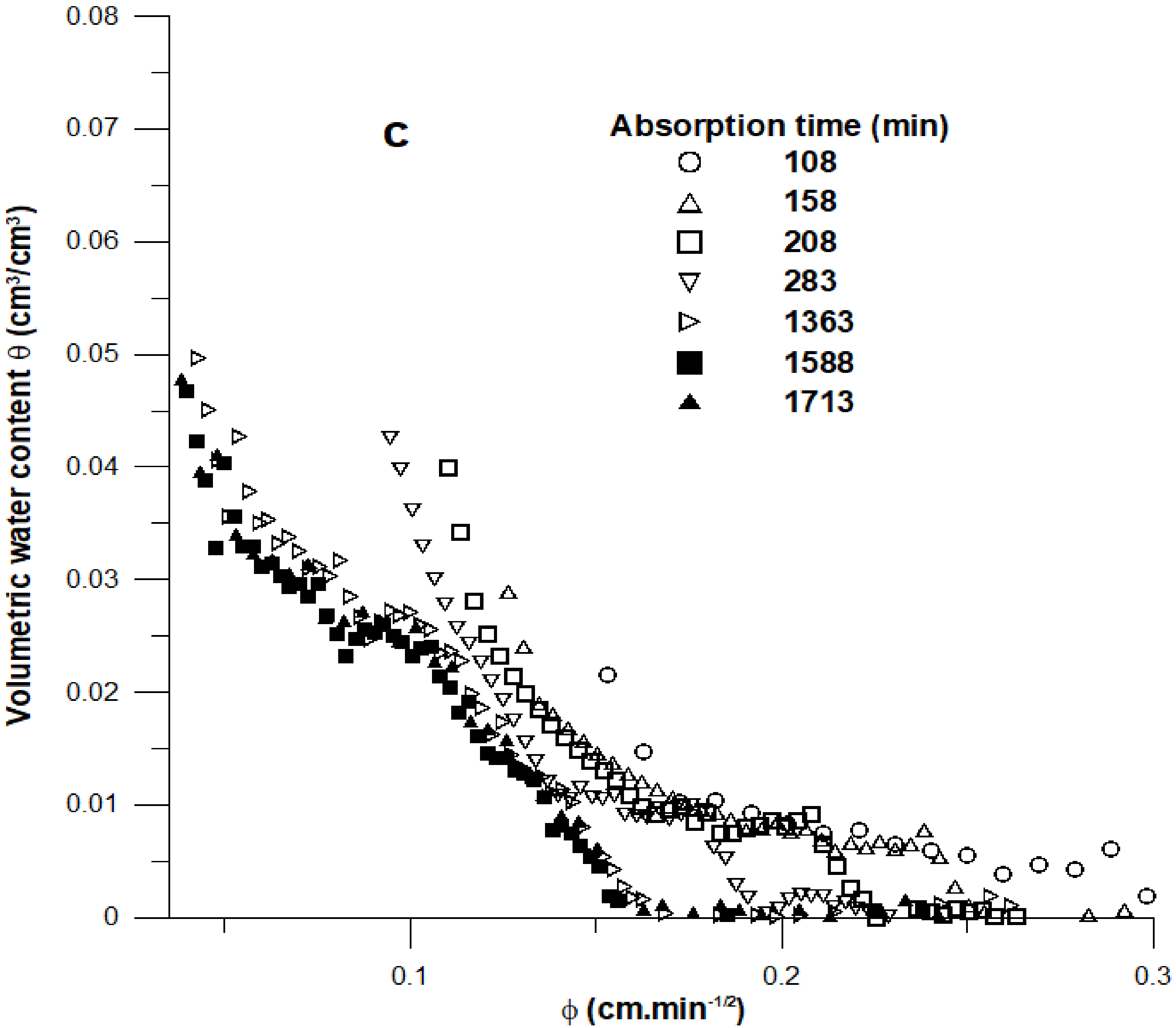
where α is a real number. Also, the Boltzmann variable ($\phi = \frac{x}{\sqrt{t}}$) is modified to $\gamma = \frac{x}{t^n}$

where the waterfront is assumed to scale to t^n. The anomalous diffusivity can be derived from equations as

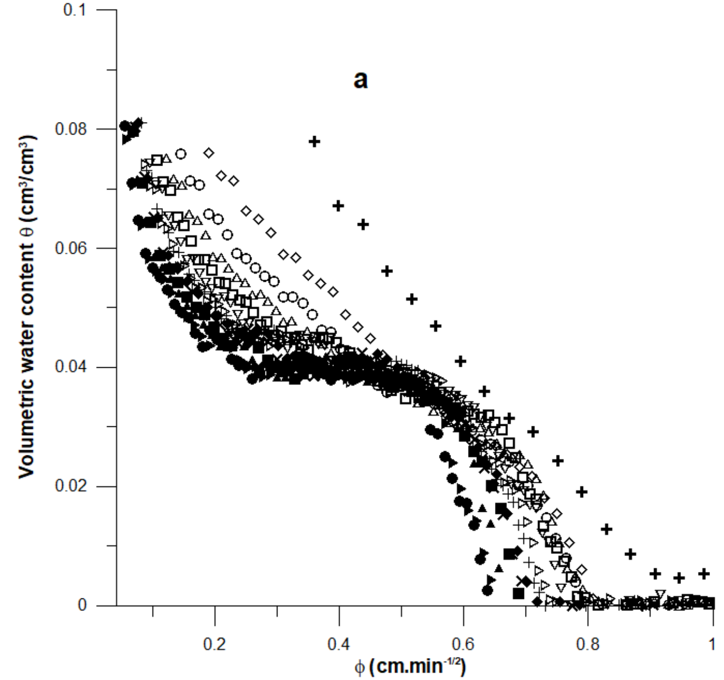
$$D(\theta) = -nt^{(1+\alpha)n-1}\left|\frac{d\gamma}{d\theta}\right|^{\alpha}\int_0^{\theta}\gamma\, d\theta$$

This means that the success of application of Fick's law for modelling the results requires that the water profiles $\theta - \phi$ should converge to single master curves.

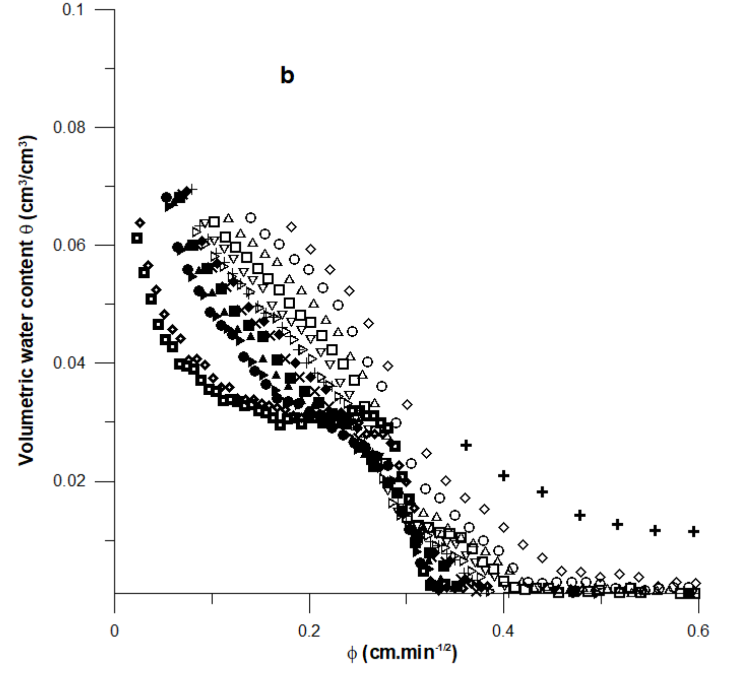




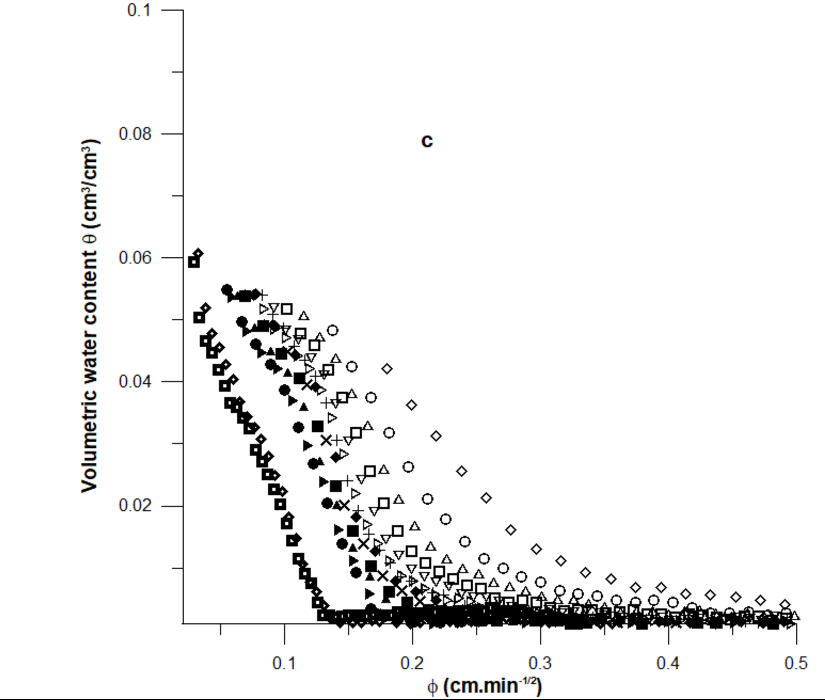
The $\theta - \phi$ profiles for the F100G0, F90G10 and F80G20 samples dried at 45^{0}C



The $\theta - \phi$ profiles for the F100G0 sample dried at 100^0C.



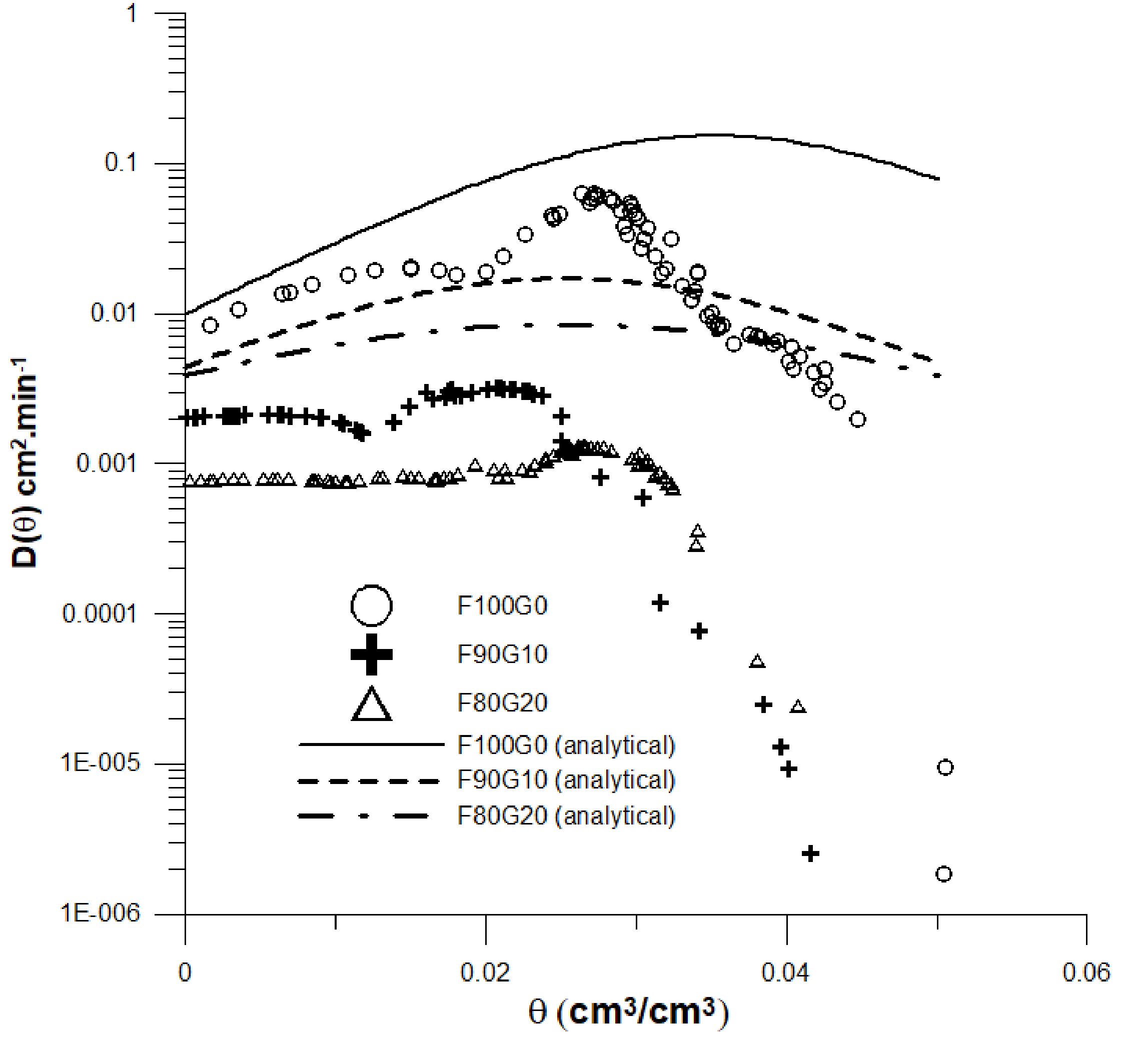
The $\theta - \phi$ profiles for the F90G10 sample dried at 100^0C.

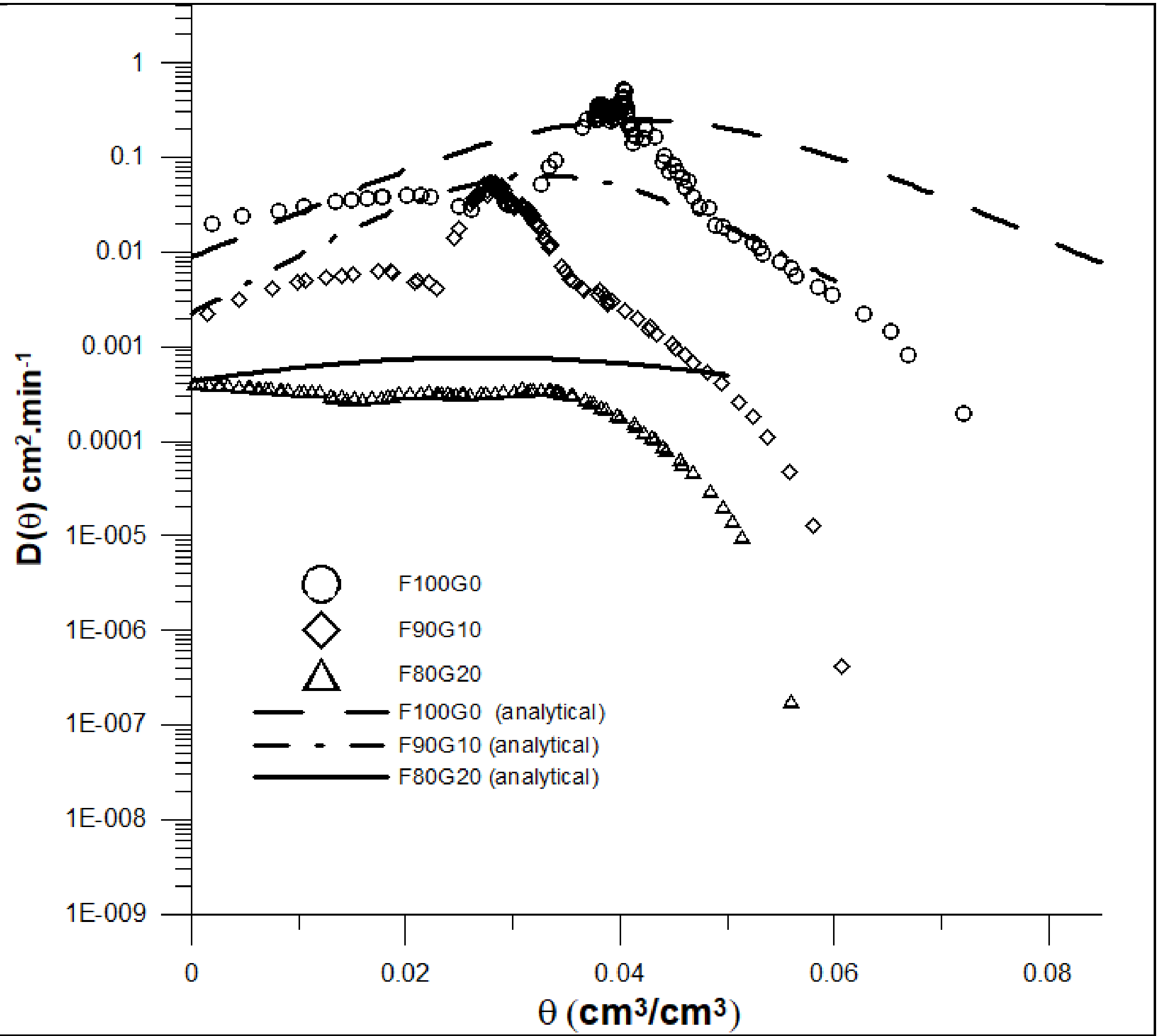


The $\theta - \phi$ profiles for the F80G20 sample dried at 100^0C.

The $\theta - \phi$ profiles do not collapse to master curves. Therefore, Fick's law of diffusion fails to model completely the water absorption processes and such failure can be attributed to the breakdown of at least of one condition and/or assumption among the assumptions required for applying the Fick's law. However, at the high absorption times, the collapse of the $\theta - \phi$ profiles seemed to be better than at the small absorption times. So, Fick's law can be used partially for modeling the results at high absorption times.

$$D(\theta) = -a(exp((\theta - 1)/b) + 1)/4\theta^2\ exp((\theta - 1)/b))$$





water diffusivity for the mortar samples dried at 45°C	water diffusivity for the mortar samples dried at 100°C

Conclusions

The NR results revealed that :

1- The water profiles, $\theta(x,t)$ at the highest water content are characterized with steep gradients along the water flow directions.

2- The water absorption processes for the samples dried at 100 ^{0}C are mainly faster than those dried at 45^{0}C.

3- These results can be used in durability and service life models and the performance evaluation of GPs.

4- The waterfront positions and the cumulative water absorption followed approximately $t^{1/2}$ -scaling. The values of capillary penetration coefficients k and sorptivities S were determined. The results can be divided into an initial and final stages for the water absorption processes.

5- As the slag content in the geopolymer mortar samples increased, both the values of k and S for the samples dried at 100^{0}C and 45^{0}C decreased. This can be attributed to the condensation of the pore structure of the mortar samples as the slag content increases. The k_{NR100} and S_{NR100} -values are mainly higher than the k_{NR45} and S_{NR45} – values.

6- The NR results obtained confirmed the gravimetric measurement results. The k and S-values followed the same trend as the values of sorptivity, water absorption, water absorption after immersion and boiling, and volume of permeable voids for fly ash/slag mortars. All these parameters decreased and increased as the slag content in the samples investigated and drying temperature increased, respectively.

7- There are good agreements between the values of sorptivities determined by the gravimetric and NR methods for the fly ash-based mortar samples dried at 45^{0}C. However , remarkable deviations were noticed for some samples dried at 100 ^{o}C. Such deviations can be attributed to drying preconditions.

8- The results obtained in this work highlight the need to reevaluate the drying procedures adopted in many standard durability testing methods when applied to alkali-activated materials. The dimension of the sample, and type of binders used should be parameters in choosing the optimum drying conditions.

9- The present results could be used to provide recommendations and simplified procedures for the drying conditions for fly ash/slag based- geopolymer mortars for the measurement of sorptivity.

10- The NR results obtained in the present study for the water absorption processes in the mortar samples provided new as well as independent experimental evidences for the enhancement of the microstructure and refining the pore space for the fly ash-based geopolymer mortar as its content of slag increased. Additionally, new experimental evidences of both high rate of water absorption (high values of k) and high amount of water absorbed (high values of S) due to modifying (widening) the pore space available for water flow as a result of drying were provided.

11- The $\theta - \phi$ profiles do not collapse completely to master curves. The collapse of the $\theta - \phi$ profiles at the high absorption times is better than at the small absorption times. At the high absorption times, the second Fick's law of diffusion can be used for modeling the results.

12- Water diffusivities were determined analytically and via differentiation and integration of the smoothed results. As the slag content and drying temperature increased in the mortars, diffusivity decreased and increased, respectively.

13- The diffusivity results presented in showed that as the content of slag in the fly ash-based-geopolymer mortars increased the water absorption behavior changed from D(θ)-dependent on water content to D(θ)- independent for most of the water absorbed, except at the highest water content. D(θ) decreased abruptly at the highest water contents.

14- The results obtained in this work showed evidences of anomalous diffusion forthcoming paper

1- El Abd el al., 2024. **Characteristics and neutron imaging of capillary water absorption for metakaolin and steel fiber reinforced slag based-geopolymer mortars, Journal of Building Engineering 82, 107960**

2- El Abd et al.,2020. Penetration of water into cracked geopolymer mortars by means of neutron radiography, Construction and Building Materials 256, 119471

3- El Abd et al., 2021. Implementation of capillary penetration coefficient on water sorptivity for pourus building materials: An experimental study, Construction and Building, Materials 298,123758

4- El Abd et al., 2019. Water sorptivity of unsaturated fractured sandstone: Fractal modeling and neutron radiography experiment , Advances in Water Resources 130, 172–183

Thank you in advance