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## Research Paper

### Effect of supplementary cementitious materials on the air permeability of concrete

**Samuel Olufemi FOLAGBADE \***

Department of Building, Obafemi Awolowo University, Ile-Ife, Nigeria

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#### ABSTRACT

In order to support the use of cement combination concrete in construction, this paper investigated the resistance of Portland cement and some binary and ternary cement concretes containing fly ash, silica fume and metakaolin to air permeability at equal water/cement ratios and strengths. At equal water/cement ratios, while fly ash binary cement concretes have higher coefficients of air permeability than Portland cement concrete due to delayed pozzolanic reactivity, silica fume and metakaolin binary cement concretes have comparable coefficients with Portland cement concrete due to their higher fineness, improved particle packing and higher pozzolanic reactivity. Consequently, the ternary cement concretes have coefficients comparable with that of Portland cement concrete. At equal strengths, while silica fume and metakaolin binary cement concretes have higher coefficients than Portland cement concrete, fly ash binary cement concretes have lower coefficients which reduced with increasing content of fly ash. Consequently, all the ternary cement concretes have lower coefficients which reduced with increasing total replacement level due to the beneficial effect of fly ash. Hence, high volume fly ash would be required to increase the resistance of concrete to air permeability at equal strength.

## 1 Introduction

The use of cement combinations (Portland cement partially replaced with supplementary cementitious materials) in concrete came as a means of supporting the sustainability principle in construction. However, the pozzolanic reaction of these supplementary cementitious materials which depends on the  $\text{Ca}(\text{OH})_2$  produced by the hydration reaction of Portland cement content would be delayed till enough  $\text{Ca}(\text{OH})_2$  is produced. Despite the intrinsic hydraulicity of these supplementary cementitious materials, well-proportioned cement combinations that would improve the strength development and durability performance of concrete at reduced cost and reduced embodied carbon-dioxide content are available [1, 2]. But, while BS EN 197-1 [3] permits the use of supplementary cementitious materials like silica fume, metakaolin (a natural calcined pozzolana), fly ash, limestone powder and ground granulated blast-furnace slag (GGBS) of up to 10%, 35%, 55%, 35% and 95% respectively, data from the European Ready Mixed Concrete Organisation [4] showed

\* Corresponding author. Tel.: +234 8140969822.

E-mail address: samuelfolagbade@yahoo.com

that, at total consumption levels of less than 20%, the supplementary cementitious materials (majorly fly ash and GGBS) are under-utilised. Hence, to support the use of cement combination in concrete, this paper investigated the air permeability coefficients of concretes containing binary and ternary combinations of Portland cement with fly ash, silica fume and metakaolin.

Fly ash is cheap and readily available [5, 6] but it has poor strength and durability performance at early ages [7, 8]. On the other hand, while silica fume and metakaolin have higher costs [1, 2] and workability problems [9-11], they have better strength and durability performance than fly ash at equal water/cement ratios due to their higher pozzolanic reactivity [12-14]. Hence, the ternary cement concretes containing Portland cement, fly ash and silica fume or metakaolin will ensure that the shortcomings of these supplementary cementitious materials are complemented [7, 15, 16].

The deterioration processes in concrete involve the ingress of aggressive agents through the surface zone [17]. Hence, the resistance of concrete against air penetration could be used as a means of determining the resistance of the surface zone of concrete against carbon-dioxide penetration, carbonation of concrete and loss of passivating layer provided by  $\text{Ca}(\text{OH})_2$  to protect the embedded reinforcing steel against corrosion. Furthermore, while concrete is specified in practice on the basis of strength, most researches in literature have been conducted on the basis of water/cement ratio. Hence, this paper investigated the resistance to air permeability of binary and ternary cement concretes containing fly ash, silica fume and metakaolin at equal water/cement ratios and strengths.

## 2 Experimental materials

The cement used for the experiments was ordinary Portland cement (PC, 42.5 type) conforming to BS EN 197-1 [3]. The supplementary cementitious materials included siliceous or Class F fly ash (FA) conforming to BS EN 450-1 [18], silica fume (SF) in a slurry form (50:50 solid/water ratio by weight) conforming to BS EN 13263-1 [19] and metakaolin (MK) conforming to BS EN 197-1 [3]. The properties of the cement and the supplementary cementitious materials are presented in Table 1. The aggregates consisted of 0/4 mm fine aggregates and 4/10 mm and 10/20 mm coarse aggregates. The coarse aggregates were uncrushed and they come in varied shapes. The 4/10 mm aggregates have rough texture and the 10/20 mm aggregates were smooth. The physical properties of the aggregates are presented in Table 2.

**Table 1: Properties of cement and Pozzolans**

Property	Cements			
	PC	FA	SF	MK
Blaine fineness, $\text{m}^2/\text{kg}$	395	388	*	2588
Loss on ignition, % <sup>a)</sup>	1.9	6.1 <sup>b)</sup>	2.7	0.9
Particle density, $\text{g}/\text{cm}^3$	3.17	2.26	2.17	2.51
% retained by 45 $\mu\text{m}$ sieve <sup>b)</sup>	-	11.0	-	-
Particle size distribution, cumulative % passing by mass <sup>c)</sup>				
125 $\mu\text{m}$	100	100	100	100
100 $\mu\text{m}$	98.2	99.2	100	100
75 $\mu\text{m}$	93.2	96.5	100	99.8
45 $\mu\text{m}$	81.8	87.0	100	99.4
25 $\mu\text{m}$	57.1	66.2	98.8	96.0
10 $\mu\text{m}$	30.1	40.6	93.8	76.2
5 $\mu\text{m}$	13.5	24.1	87.5	50.7
2 $\mu\text{m}$	5.6	10.9	85.5	18.2
1 $\mu\text{m}$	2.9	4.8	78.7	4.7
0.7 $\mu\text{m}$	1.3	1.9	50.7	1.4
0.5 $\mu\text{m}$	0.2	0.3	10.5	0.1

\* Fineness for SF = 15,000-30,000  $\text{m}^2/\text{kg}$  [20]

a) In accordance with BS EN 196-2 [21]

b) In accordance with BS EN 450- 1 [18]

c) Obtained with the Laser Particle Sizer

Concrete was designed using the BRE Design Guide [22], saturated surface-dry (SSD) aggregates and a free water content of 165 kg/m<sup>3</sup> at the water/cement ratios of 0.35, 0.50 and 0.65. Potable water, conforming to BS EN 1008 [23], was used for mixing, curing and testing the concrete specimens. A superplasticiser (based on carboxylic ether polymer) conforming to EN 934-2 [24], was applied during mixing to achieve a consistence level of S2 defined by a nominal slump of 50–90 mm in accordance with BS EN 206- 1 [25].

**Table 2: Properties of fine and coarse aggregates**

Property	Aggregates <sup>1)</sup>		
	Fine	Coarse	
	0/4 mm	4/10 mm	10/20 mm
Shape, visual	-	Varied	Varied
Surface texture, visual	-	Rough	Smooth
Particle density <sup>2)</sup>	2.6	2.6	2.6
Water absorption, % <sup>3)</sup>	1.0	1.7	1.2
% passing 600 µm sieve	55.0	-	-

1) Aggregates were obtained from Wormit Quarry.  
 2) In accordance with BS EN 1097- 6 [26]  
 3) In accordance with BS EN 1097- 6 [26], Laboratory-dry condition

### 3 Experimental methods

Concrete was prepared to BS EN 12390-2 [27] and the specimens were cast, cured under a layer of damp hessian covered with polythene for 20–24 hours, demoulded and cured in water tanks maintained at about 20°C until the tests' dates. Tests were carried out on hardened concrete specimens to determine the cube compressive strength and air permeability coefficients of the cement combination concretes at the water/cement ratios of 0.35, 0.50 and 0.65. Cube compressive strength was obtained in accordance with BS EN 12390-3 [28] using 100mm cubes at the curing age of 28 days. Since absorption into concrete is a function of the drying temperature and immersion duration [29], air permeability coefficient was determined with specimens oven-dried to constant mass at 105±5°C to ensure a uniform basis for the comparison and repeatability of the results as the microstructure of the test specimens would not be adversely affected.

The University of Dundee method [30] was used to obtain the outlet volume rates of flow from which the coefficients of air permeability of the concrete specimens were calculated at various inlet pressures. The layout of the test apparatus is shown in Figure 1. The intrinsic coefficients of air permeability for the specimens were then extrapolated from the plottings of these coefficients at infinite inlet pressure.

This method involves testing 54 mm diameter core specimens of about 50 mm thick obtained from water-cured 100 mm cubes. Two specimens each were tested at the curing ages of 28, 90 and 180 days at the water/cement ratios of 0.35, 0.50 and 0.65. Prior to testing, the specimens were oven dried to constant mass at about 105±5°C and allowed to cool to room temperature in a desiccator containing silica gel. At each test age, the surface of each specimen was greased (taking care not to apply grease to the end faces) and placed in the Hoek-Franklin cell with the cast face down to receive air. This is illustrated schematically in Figure 2.

A constant oil pressure of 600 psi (4.2 N/mm<sup>2</sup>) was applied to the circumference of the specimen to provide an air-tight seal. Uni-axial air pressure was applied by means of a compressor to the specimen at the inlet pressures of 110, 90, 70, 50, and 30 psi (i.e. 0.76, 0.62, 0.48, 0.35 and 0.21 N/mm<sup>2</sup> respectively) and the corresponding readings on the outflow meter (outlet pressures and outlet volume rates of flow) were recorded when equilibrium (steady flow) was obtained. The corresponding coefficients of permeability (K) obtained with the aid of Equation 1 [30] were plotted against the reciprocal of the corresponding outlet pressures and the intrinsic coefficient of permeability was obtained as the intercept when the line of best fit was extrapolated to touch the coefficients' axis.

$$K = \frac{2\eta LP_2 Q_2}{A(P_1^2 - P_2^2)} \quad (1)$$

where A = cross-sectional area of the specimen in  $m^2$

L = thickness of specimen in m

$\eta$  = dynamic viscosity of air ( $1.79 \times 10^{-5} \text{ Ns/m}^2$ )

$P_1$  = inlet pressure ( $\text{N/m}^2$ )

$P_2$  = outlet pressure ( $\text{N/m}^2$ )

$Q_2$  = outlet volume rate of flow ( $\text{m}^3/\text{s}$ )

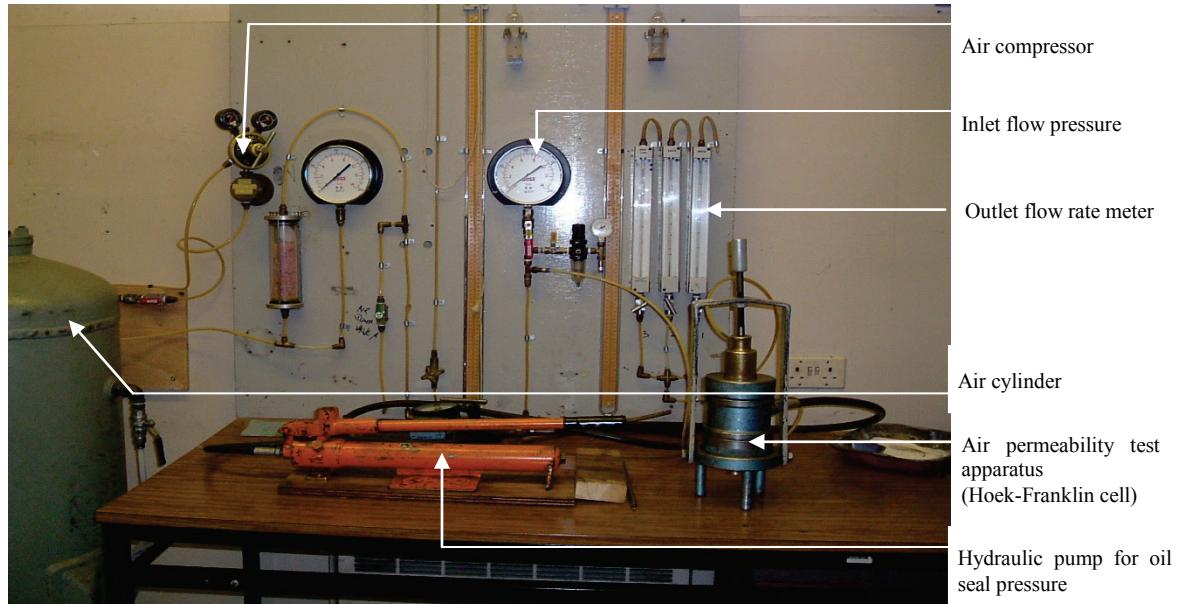


Figure 1: University of Dundee installation for air permeability test

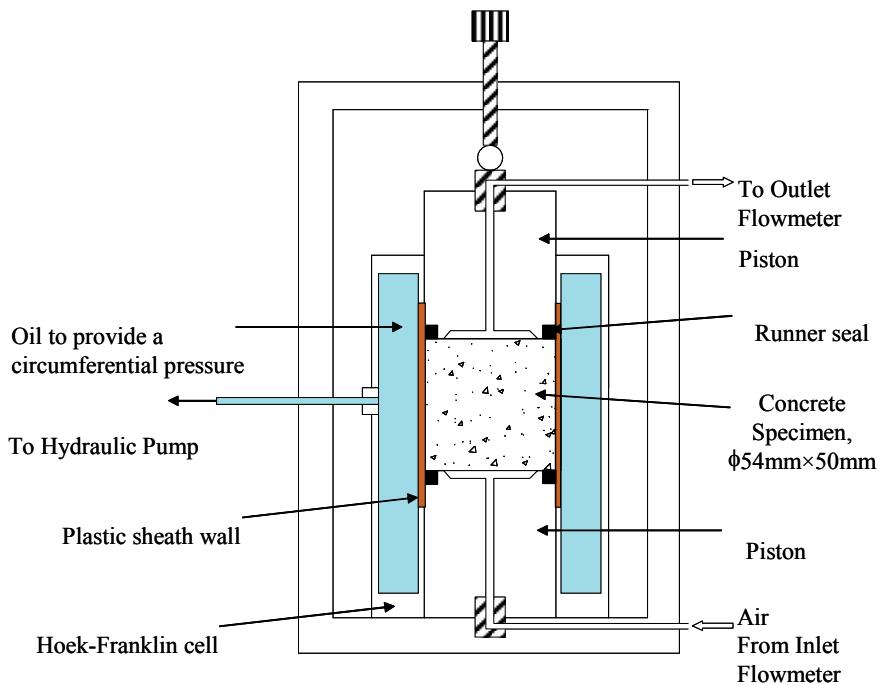
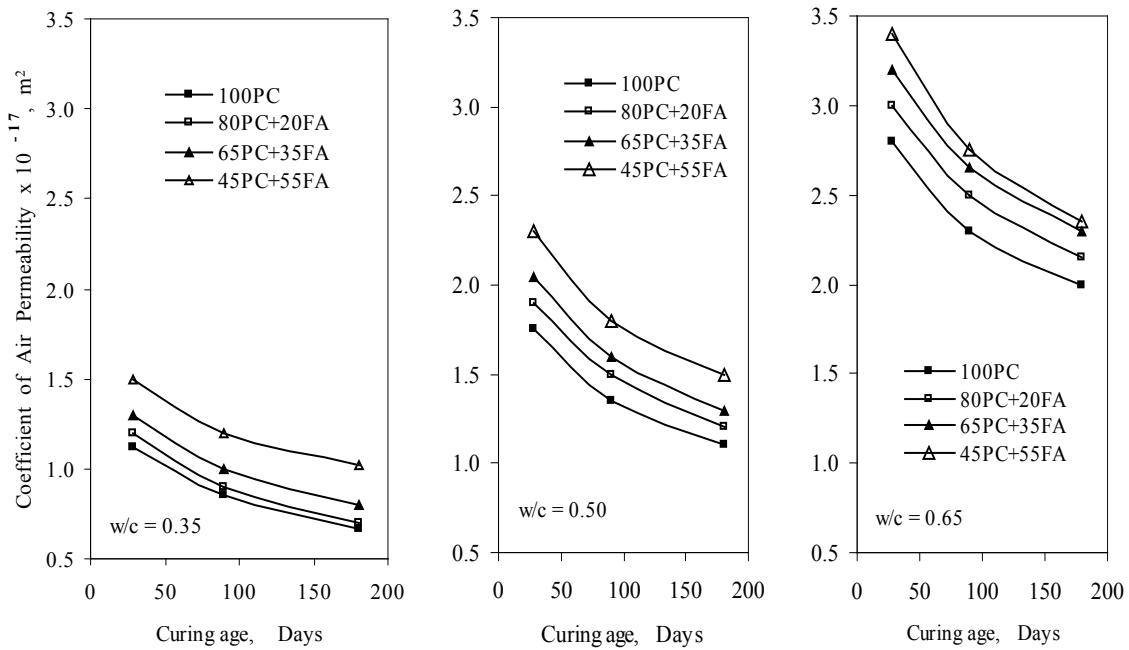


Figure 2: A schematic layout of the air permeability test apparatus

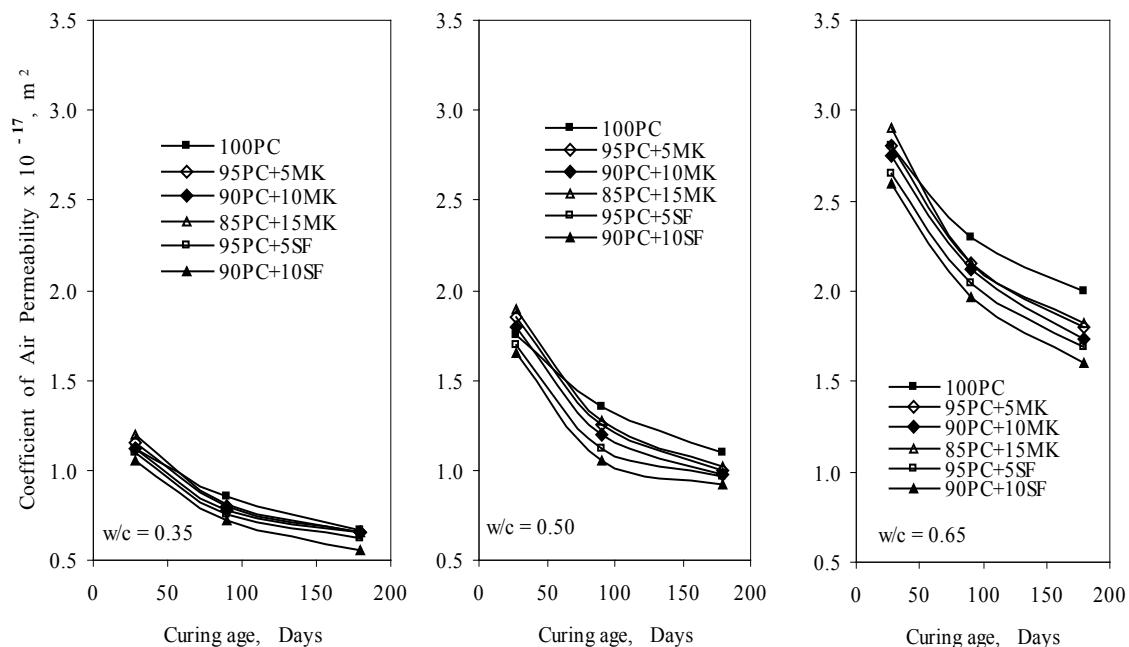
## 4 Analysis and discussion of results

### 4.1 Coefficients of air permeability of binary cement concretes at equal water/cement ratios

Figures 3 and 4 present the coefficients of air permeability of Portland cement concrete and fly ash, silica fume and metakaolin binary cement concretes at the water/cement ratios of 0.35, 0.50 and 0.65.



**Figure 3: Coefficients of air permeability of Portland cement and fly ash binary cement concrete at the water/cement ratios of 0.35, 0.50 and 0.65**



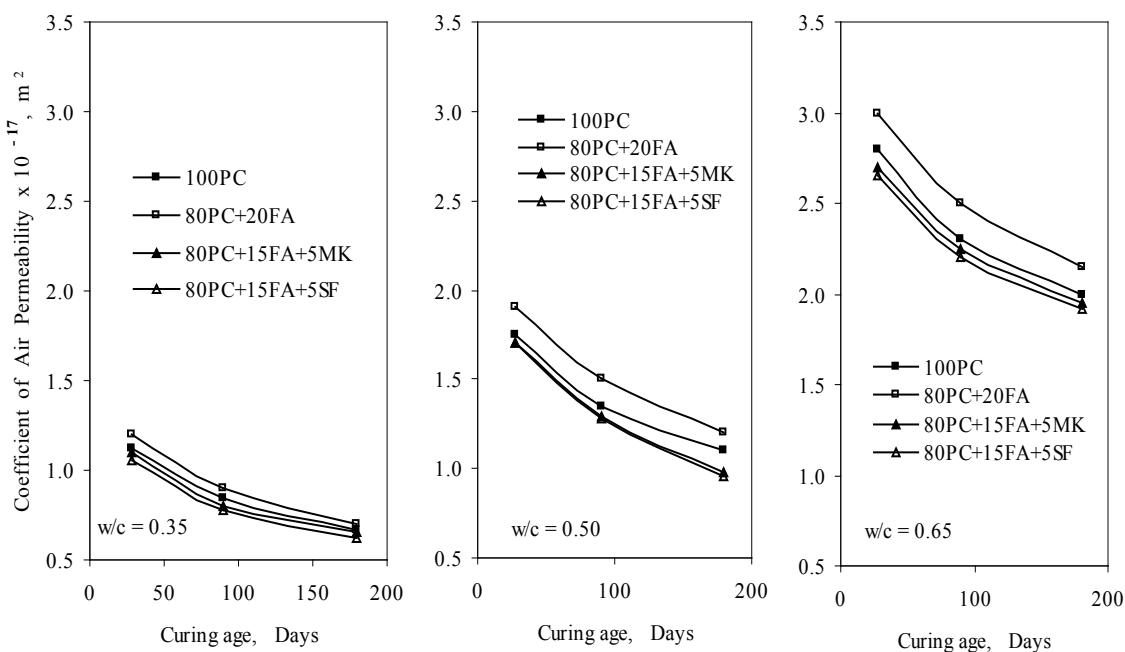
**Figure 4: Coefficients of air permeability of Portland cement and silica fume and metakaolin binary cement concretes at the water/cement ratios of 0.35, 0.50 and 0.65**

As expected, the Figures show that coefficient of air permeability increased with increasing water/cement ratio and reduced with increasing curing age. Compared with Portland cement, Figure 3 shows that the coefficients of air permeability of fly ash binary cement concretes increased with increasing content of fly ash. The increase must be due to the reduction in Portland cement and  $\text{Ca}(\text{OH})_2$  contents (dilution effect) and the delayed pozzolanic reactivity of fly ash [8, 31].

Figure 4 also shows that the coefficients of air permeability of silica fume and metakaolin binary cement concretes were comparable with that of Portland cement at 28 days and the coefficients reduced with increasing curing age such that 180 days, the values were lower than that of Portland cement concrete. Compared with Portland cement and fly ash binary cement concretes, the lower coefficients of air permeability of silica fume and metakaolin binary cement concretes must be due to their higher fineness, improved packing ability and the presence of more nucleation sites to improve pozzolanic reactivity and reduce the pore size and porosity within the paste and at the interface transition zones between the paste and the aggregates despite the dilution effect [14, 32, 33]. Compared with metakaolin, silica fume binary cement concretes have lower coefficients of permeability, at equal replacement levels, and this must also be due to the higher fineness of silica fume (Table 1) resulting in better packing of cement particles and improved pozzolanic reactivity.

#### 4.2 Coefficients of air permeability of ternary cement concretes at equal water/cement ratios

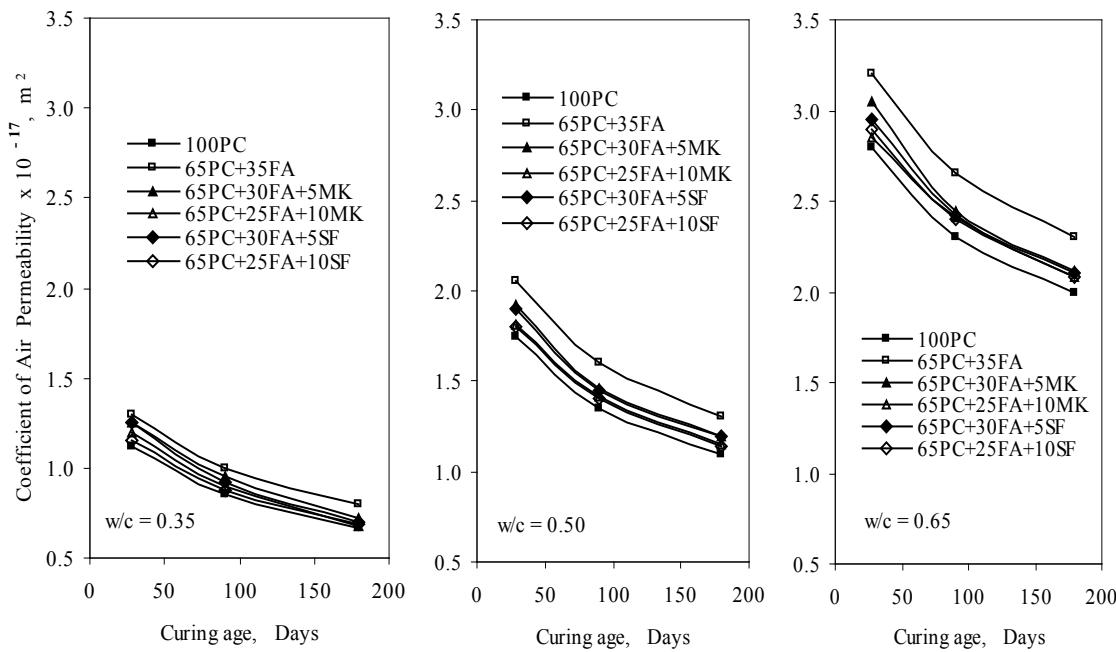
Figures 5-7 present the coefficients of air permeability of Portland cement, fly ash binary cement and silica fume and metakaolin ternary cement concretes at the total replacement levels of 20, 35 and 55% and water/cement ratios of 0.35, 0.50 and 0.65. The ternary cements were obtained by partly replacing the fly ash contents of the fly ash binary cements with either silica fume or metakaolin (Table 3).



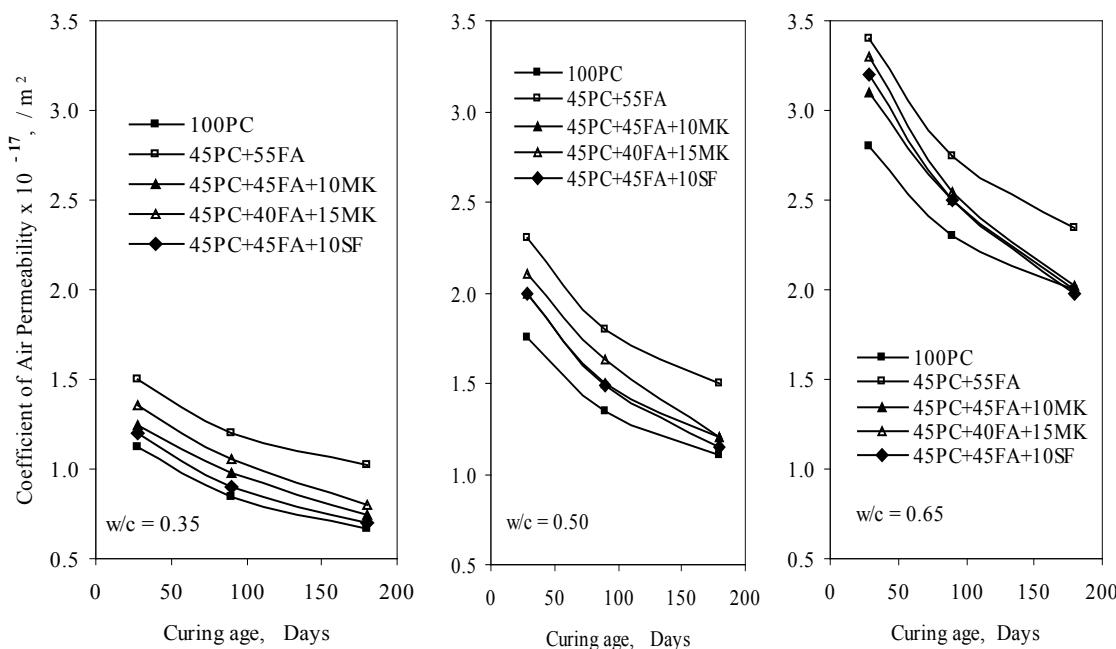
**Figure 5: Coefficients of air permeability of Portland cement, fly ash binary cement and silica fume and metakaolin ternary cement concretes at 20% total replacement level and water/cement ratios of 0.35, 0.50 and 0.65**

Compared with the respective fly ash binary cement concretes, the addition of silica fume and metakaolin as ternary cement components reduced the coefficients of air permeability of concrete with increasing content at all the test ages. Compared with Portland cement concrete, the ternary cement concretes have lower coefficients at the total replacement level of 20%. The coefficients increased with increasing total replacement level such that they become higher than that of Portland cement concrete at 28 days at the total replacement levels of 35 and 55%. The increase in the coefficients, with increasing total replacement level, must be due to the reduction in Portland cement content and the amount of  $\text{Ca}(\text{OH})_2$  available for the pozzolanic reaction of the supplementary cementitious materials [8, 31]. However, due to improved pozzolanic reactivity with curing age, the coefficients of the ternary cement concretes at 35 and 55% total replacement

levels reduced with increasing age such that at 180 days they become comparable with that of Portland cement concrete. Hence, the improved performance of the ternary cement concrete would be due to improved packing and high pozzolanic reactivity of the ternary cement components (i.e., silica fume and metakaolin) within the paste matrix and at the interface zone between the paste and the aggregates [14, 32, 33]. Also, in the same token, the higher fineness of silica fume must have also resulted in the lower coefficients of the silica fume ternary cement concretes than the metakaolin ternary cement concretes at equal replacement levels and water/cement ratios.



**Figure 6: Coefficients of air permeability of Portland cement, fly ash binary cement and silica fume and metakaolin ternary cement concretes at 35% total replacement level and water/cement ratios of 0.35, 0.50 and 0.65**



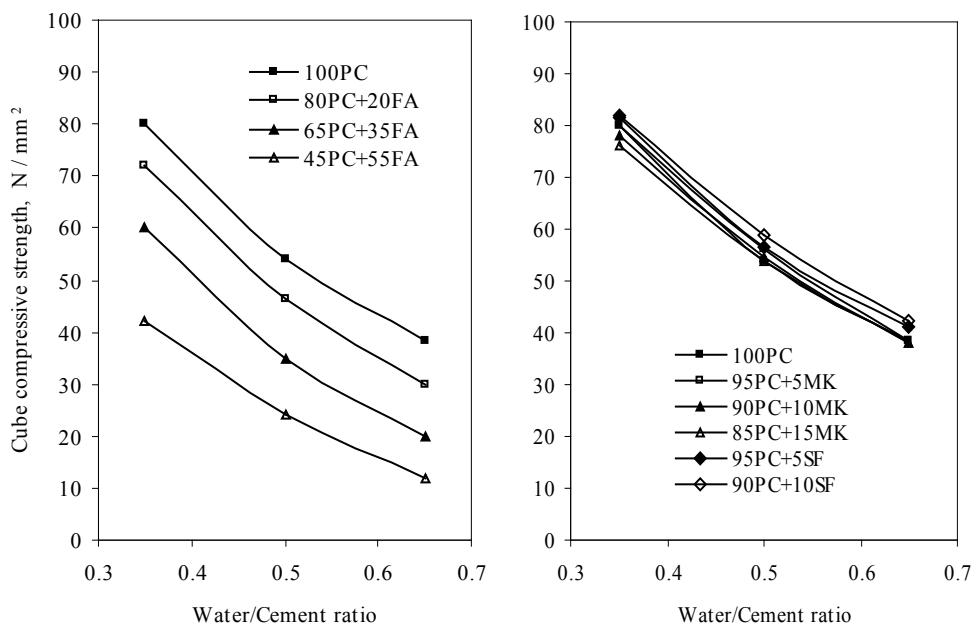
**Figure 7: Coefficients of air permeability of Portland cement, fly ash binary cement and silica fume and metakaolin ternary cement concretes at 55% total replacement level and water/cement ratios of 0.35, 0.50 and 0.65**

#### 4.3 Coefficients of air permeability of cement combination concretes at equal strengths

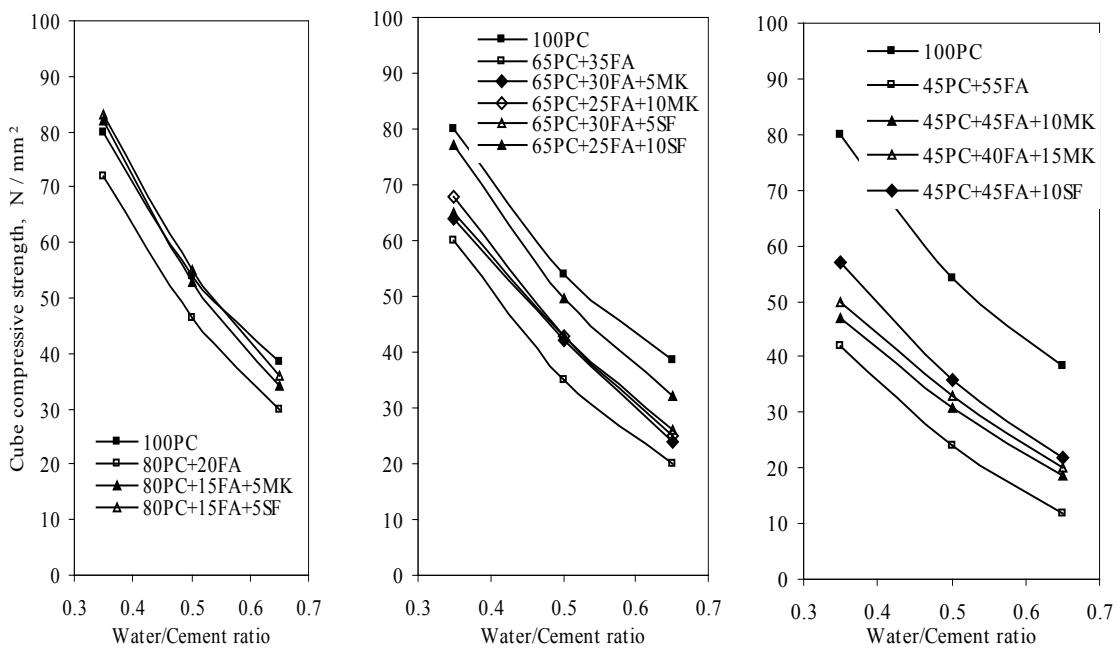
Figures 8 and 9 show that cube compressive strengths of concretes, at 28 days, reduced with increasing water/cement ratio. Compared with Portland cement, while the addition of fly ash as a binary cement component reduced concrete strength with increasing content, the addition of silica fume and metakaolin resulted in comparable strengths with Portland cement concrete. Compared with the fly ash binary cement concretes, the addition of silica fume and metakaolin as ternary cement components resulted in ternary cement concretes with improved strength.

Since concrete is specified in practice on the basis of strength at 28 days, the coefficients of air permeability of the concretes were examined at equal 28-day strengths. The cube compressive strengths (Figures 8 and 9) and the coefficients of permeability of the concretes at the curing age of 28 days (Figures 3–7) at the water/cement ratios of 0.35, 0.50 and 0.65 were interpolated to obtain the coefficients of air permeability of concretes at the 28-day strengths of 35, 40, 45 and 50 N/mm<sup>2</sup> (Table 3). Apart from falling within the range of strengths that would commonly be used in practice, these strengths also satisfy most of the strength requirements in BS EN 206-1 [25] and BS 8500 [34, 35].

Table 3 shows that the coefficients of air permeability of concretes, at equal strengths, were achieved at different water/cement ratios and they reduced with increasing strength. Compared with Portland cement, all the fly ash binary cement concretes have lower coefficients of air permeability, at equal strengths, and these values decreased with increasing content of fly ash up to 55%. On the other hand, all the silica fume and metakaolin binary cement concretes have higher coefficients than Portland cement concrete. Table 3 shows that while the coefficients of air permeability of the binary cement concretes increased with increasing content of silica fume, they reduced with increasing content of metakaolin (MK) up to 10% and become higher at 15% MK content. Compared with the respective fly ash binary cements, the ternary cement concretes have higher coefficients at equal strengths. However, all the ternary cement concretes have lower coefficients than Portland cement concrete and the coefficients reduced with increasing total replacement level due to the beneficial effect of fly ash at equal strengths. Hence, high content of fly ash would play a major role in reducing the air permeability of concrete specified on the basis of strength. These results indicate that, if appropriately proportioned, the use of cement combination would result in better resistance of concrete to permeation at equal strengths [36, 37]. Table 3 also shows that, at equal strengths, metakaolin ternary cement concretes have lower coefficients of air permeability than silica fume ternary cement concretes at equal replacement levels.



**Figure 8: Cube compressive strength of Portland cement and fly ash, silica fume and metakaolin binary cement concretes at the water/cement ratios of 0.35, 0.50 and 0.65**



**Figure 9: Cube compressive strength of Portland cement and ternary cement concretes at the water/cement ratios of 0.35, 0.50 and 0.65 and total replacement levels of 20, 35 and 55%**

**Table 3: Coefficients of air permeability of concretes at 28-day strengths of 35, 40, 45 and 50 N/mm<sup>2</sup>**

Mix combination	Coefficient of Air Permeability $\times 10^{-17}$ , m <sup>2</sup>							
	35 N/mm <sup>2</sup>		40 N/mm <sup>2</sup>		45 N/mm <sup>2</sup>		50 N/mm <sup>2</sup>	
	w/c	Coeff.	w/c	Coeff.	w/c	Coeff.	w/c	Coeff.
100%PC	*	*	0.63	2.64	0.57	2.19	0.53	1.93
80%PC+20%FA	0.59	2.51	0.55	2.22	0.51	1.96	0.47	1.73
80%PC+15%FA+5%MK	0.63	2.55	0.59	2.25	0.55	1.99	0.51	1.76
80%PC+15%FA+5%SF	*	*	0.61	2.36	0.57	2.09	0.53	1.85
65%PC+35%FA	0.50	2.05	0.46	1.81	0.43	1.65	0.40	1.51
65%PC+30%FA+5%MK	0.55	2.25	0.51	1.98	0.47	1.75	0.44	1.60
65%PC+25%FA+10%MK	0.56	2.18	0.52	1.93	0.48	1.71	0.45	1.56
65%PC+30%FA+5%SF	0.56	2.28	0.52	2.02	0.48	1.79	0.45	1.63
65%PC+25%FA+10%SF	0.61	2.56	0.57	2.26	0.53	1.99	0.49	1.74
45%PC+55%FA	0.40	1.74	0.36	1.55	**	**	**	**
45%PC+45%FA+10%MK	0.47	1.82	0.42	1.56	0.37	1.33	**	**
45%PC+40%FA+15%MK	0.48	1.98	0.43	1.70	0.39	1.51	**	**
45%PC+45%FA+10%SF	0.50	2.00	0.46	1.75	0.42	1.53	0.39	1.38
95%PC+5%MK	*	*	0.63	2.66	0.58	2.33	0.54	2.08
90%PC+10%MK	*	*	0.62	2.54	0.57	2.21	0.53	1.97
85%PC+15%MK	*	*	0.62	2.68	0.57	2.33	0.53	2.08
95%PC+5%SF	*	*	*	*	0.60	2.30	0.55	1.98
90%PC+10%SF	*	*	*	*	0.62	2.38	0.57	2.05

\* Required water/cement ratio is higher than 0.65 (0.65 is the highest water/cement ratio tested)

\*\* Required water/cement ratio is lower than 0.35 (0.35 is the lowest water/cement ratio tested)

## 5 Conclusion

This study has investigated the effect of fly ash, silica fume and metakaolin on the air permeability of Portland cement concrete at equal water/cement ratios and strengths and the following conclusions have been drawn:

1. Coefficients of air permeability increased with increasing water/cement ratio and reduced with increasing curing age and compressive strength.
2. At equal water/cement ratio, fly ash binary cement concretes have considerably higher coefficients of air permeability than Portland cement concrete due to delayed pozzolanic reactivity. Silica fume and metakaolin binary cement concretes have coefficients comparable with that of Portland cement at 28 days and lower coefficients at later ages due to their higher fineness resulting in better particle packing and improved pozzolanic reactivity. Consequently, the ternary cement concretes have lower coefficients of air permeability than fly ash binary cement. Also, the ternary cement concretes which have higher coefficients than Portland cement at 28 days improved with age such that the coefficients became comparable with that of Portland cement concrete at 180 days.
3. At equal strengths, while the addition of silica fume and metakaolin as binary cement components increased the coefficients of air permeability of concrete with increasing content, fly ash binary cement as a binary cement component reduced the coefficients with increasing content. Consequently, the coefficients of air permeability of the ternary cement concretes were lower than that of Portland cement and they reduced with increasing total replacement level due to the beneficial effect of fly ash.
4. At equal replacement levels, silica fume (with increasing content) has higher resistance to air permeability than metakaolin at equal water/cement ratios. However, at equal strengths, metakaolin has higher resistance to air permeability than silica fume and while the resistance reduced with increasing content of metakaolin, it increased with increasing content of silica fume.

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