Impact of the porosity of coarse aggregates on the structuration of the paste-aggregate interface: Elementary model study

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Abstract. In the context of sustainable development for concrete production, local aggregates and recycled aggregates should be preferred but they can be porous and of lower quality. As a result, the compressive strength and the durability of concrete are adversely affected. In a preliminary understanding and with a view to accentuating phenomena occurring in the concrete, the Elementary Model (EM composed of paste and gravel, was studied. In this paper, the effect of Water Porosity (WP), moisture state and volume of coarse aggregates, as well as the nature of mineral admixture on the WP of EM was taken into account. Results indicated that volume and water porosity of aggregate are the two most important factors affecting WP of EM. Besides, it was also shown that aggregate porosity has the same trend of impact on the WP of EM while does not have the same trend of impact concerning the effect of the cement paste-aggregate interface on the WP of EM whatever the nature of mineral admixture, water porosity, moisture state and volume of aggregate. Explanatory attempts are proposed from the Scanning electron microscope (SEM) in combination with Energy dispersive spectroscopy (EDS) to support for statistic and ranking analysis.

Keywords: Paste-aggregate interface, Mineral admixture, Porosity.

1 Introduction

The quality of the concrete depends upon the quality of cement paste and aggregates, as well as the bond between them. Related works showed factors that affect the properties of concrete, namely: the nature properties and moisture state of aggregates [1] [2], the incorporation of mineral admixture [1] [3], and the volume content of aggregates [4] [5]. These impact factors play important role on the quality of concrete, but what extent of these impacts are still questionable.

In this paper, at the scale of elementary model (EM), composed by only paste and gravel, the impact of water porosity (WP), moisture state (Saturated Surface Dry (SSD) and Oven Dry (OD) state), and volume of aggregate on WP of EM was qualified. Five aggregates differing in physical and chemical nature were used. Since the

C. Ha-Minh et al. (eds.), *CIGOS 2019, Innovation for Sustainable Infrastructure*,
Lecture Notes in Civil Engineering 54,
https://doi.org/10.1007/978-981-15-0802-8_68

replacement of cement by mineral admixture not only reduces the cement content (for environmental and economic benefits [6]) but also improves the properties of concrete [7], four kinds of mineral admixture were also considered. Statistical method [8] and ranking method [9] were applied to analyze the experimental results. The analysis was then completed by the means of SEM/EDS observations/characterization.

2 Experimental procedure

2.1 Materials:

Raw materials. Table 1 shows the main chemical and physical properties of the cement and the four mineral admixtures used.

Table 1. Characteristics of cement and mineral admixtures.

	Cement I 52.5 N	Metakaolin	Limestone filler	Slag	Fly ash
Physical characteristics					
Density (g/cm^3)	3.09	2.50	2.70	2.90	2.00
Specific surface (cm^2/g)	4600 (Blaine)	16000 (BET)	5960 (Blaine)	4450 (Blaine)	2300 (Blaine)
Chemical / Mineralogical composition (wt. %)					
SiO_2	20.1	93.16	/	37.4	52.5
Al_2O_3	5.2		/	10.8	22.5
Fe_2O_3	3.3	/	/	0.5	8.5
CaO	64.1	0.36	/	43.7	3.5
$CaCO_3$	/	/	95	/	/

The five types of coarse aggregate along with the physical and chemical properties are presented in Table 2.

Table 2. Characteristics of aggregates.

Rank	**Designation**	**Density (kg/m^3)**	**Water porosity (vol. %)**	**Water absorption (wt. %)**	**Mineralogy (>90% by mass)**
1	Concrete Recycled	2580	20,03	5,20	SiO_2, $CaCO_3$
2	Soft limestone	2450	12,45	4,20	$CaCO_3$
3	Siliceous-calcareous	2580	1,20	1,05	SiO_2, $CaCO_3$
4	Hardened limestone	2670	0,88	0,56	$CaCO_3$
5	Siliceous-quartz	2720	0,65	0,51	SiO_2

Elementary Model (EM). The coarse aggregate volume (measured after 24-h immersion in water) was fixed to 3.30cm^3 ± 0.05 (V_1), 7.30cm^3 ± 0.05 (V_2) and 9.80 cm^3 ± 0.05 (V_3), which corresponded to two, four and six selected gravels to be incorporated in the paste. The pre-conditioning to obtain two moisture states were three days immersed in water for Saturated Surface Dry (SSD) state and three days at 105°C for Oven Dry (OD) state.

Five compositions of binder (water/binder ratio = 0.35) were considered. The reference composition was only cement, while the four other ones consisted in the replacement of cement with 15% metakaolin, or 25% limestone filler, or 30% slag or 30% fly ash by mass according to the maximum replacement rates fixed by the European standard EN 206/CN [10].

All EM were produced in cylindrical container (5cm-diameter and 4-cm height), and cured in water at 20°C for 90 days.

2.2 Tests

After the curing period, the WP of EM was measured according to French standard NF-P18-459 [11] (Eq. (1)):

$$WP_{exp} = \frac{M_{air}-M_{dry}}{M_{air}-M_{water}} (\%) \tag{1}$$

M_{air}, M_{dry}, M_{water} : SSD weight, OD weight and hydrostatic weight of EM, respectively.

The SEM/EDS was a useful method to determine the chemical length of the interfacial transition zone (ITZ) based on the variation of the Ca, Si and C/S atomic ratios through 100-point scans from the aggregate phase to the bulk paste (fig. 1).

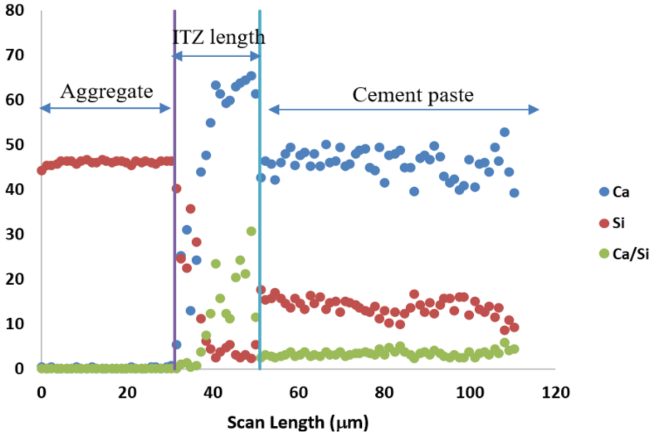

Fig. 1. ITZ chemical length

3 Analysis method

3.1 Statistical method

A multi linear model was performed using the statistical environment R [8] in order to evaluate and compare the impact of the studied factors on the WP of EM. In the present case, there were 8 variables (X) (Table 3, column 2, row 1 to 8) and the expected variable (Y_i) is WP of EM. Our dataset was big enough (450 samples including 3 times repeatability) and satisfied all assumptions of the model (linear relationship between variables (X_i) and expected variables (Y_i), independence between variables (Y_i), normal distribution of the residuals). The coefficient of determination R^2 was used to explain for the changing of the expected variable.

3.2 Ranking method

Aggregates were first ranked by decreasing porosity as presented in Table 2 (column 1). Then, when considering WP of EM, the ranking method [9], based on Kendall's coefficient of concordance, was applied to answer to three questions:

1. Is EM WP ranked in the same order as aggregate WP, whatever the binder nature?
2. Is the effect of the paste-aggregate interface on EM WP ranked in the same order as aggregate WP, whatever the binder nature?
3. Is the chemical length of ITZ ranked in the same order as the aggregate WP, whatever the binder nature?

Since the difference between EM WP or between EM ITZ length was not always significant, the Kendall's coefficient was calculated by considering ties case (Eq. (2)):

$$W = \frac{12.S}{k^2.(n^3-n)-k\sum_1^k(\sum_1^s(t^3-t))} \tag{2}$$

S = Variance of the sums of ranks; k = Judges (five binder natures); n = Contestants (1: EM WP; 2: ITZ effect; 3: ITZ chemical length); t =Tie corresponding to a set s of ranks for a given judge.

4 Results and discussion

4.1 Result from multi linear regression model

When porous aggregates (rank 1 and 2, table 2; 180 EM samples out of 450) and non-porous aggregates (rank 3, 4 and 5, table 2; 270 EM samples out of 450) are analyzed separately, it is clear that when porous aggregates are used, the moisture state and the aggregates WP are factors influencing EM WP. In addition, the incorporation of mineral admixtures such as limestone filler or slag becomes also influential factors.

When considering the five aggregates together, all variables explain for 93.85% the change in the EM WP. The two most important variables, i.e. aggregate volume and aggregate WP, account for approximately 75% and 11% the change in the EM WP, respectively. Among all the remaining variables, only the slag has statistical meaning explaining for 5.58% the change in the expected variable EM WP.

4.2 Result from the ranking method

Questions 1 and 2 (section 3.2) – Results from EM WP measurements. Concerning question 1 (Table 4, column 3), the Kendall's coefficient of concordance (W) in all cases is higher than the threshold at the 0.01 significance level (0.571). Hence, EM WP is classified in the same order as aggregate WP, whatever the nature of mineral admixture, but also the moisture state and the volume of aggregates.

Table 3. Coefficient of determination R^2 of model and variables

Variables		R^2 (%) (single)		
		Non porous aggregate (270 samples)	Porous aggregate (180 samples)	Both kinds of aggregate (450 samples)
X1	Cement	0.00	0.0	0.00
X2	Metakaolin	0.03	0.11	0.00
X3	Limestone	0.06	1.34	0.28
X4	Slag	5.04	8.84	5.58
X5	Fly ash	0.00	0.00	0.00
X6	Moisture state of aggregate	0.02	2.22	0.12
X7	WP of aggregate	0.34	2.34	11.02
X8	Volume of aggregate	87.97	76.92	74.89
Coefficient of determination R^2 (%) (Multi)		95.99	93.70	93.85

Table 4. Values obtained for the Kendall's coefficient of concordance W

Moisture state of aggregate	Aggregate volume	Question 1 (Section 3.2)	Question 2 (Section 3.2)	Question 3 (Section 3.2)	Threshold at 0.01 Significance level
SSD	V_1	0.808	0.333	0.311	0.571
	V_2	0.956	0.068		
	V_3	0.961	0.302		
OD	V_1	0.632	0.124	0.372	
	V_2	0.755	0.193		
	V_3	0.633	0.567		

To answer to question 2, the difference in EM WP between experimental method (Eq. (1) - ITZ taken into account) and analytical calculations (Eq. (3) –ITZ not considered) was calculated. The W values shown in Table 4 (column 4) are lower than the 0.01 significance level (0.571). This enable us to conclude that the effect of the interface on the EM WP is not classified in the same order as aggregate WP whatever the nature of mineral admixture, moisture state and volume of aggregate.

$$WP_{cal} = \frac{WP_{aggregate} \cdot V_{aggregate} + WP_{paste} \cdot V_{paste}}{V_{EM}} (\%) \qquad (3)$$

WP, V - Water porosity and volume of aggregate and paste; V_{EM} - Volume of EM

Question 3 - Results from SEM-EDS observation. The chemical length of the ITZ (fig. 1) was determined by the means of SEM/EDS tests in order to enhance the conclusions of statistical and ranking results pertaining to EM WP measurements. The chemical length of ITZ was then analyzed by ranking method (Table 4, column 5). The resulting W values are lower than the level of significance at 1% (0.571). This indicates that the ITZ chemical length of EM is not ranked in the same order as the aggregate WP whatever the nature of the mineral admixture and moisture state of aggregate.

5 Conclusions

From EM (paste + gravel(s)) investigations, the following conclusions can be drawn:

1. The two most important variables effect on EM WP are aggregate volume and aggregate WP, which account for approximately 75% and 11% the change of EM WP, respectively. The impact of moisture state and incorporation of mineral admixture is important when porous aggregates are used. Particularly, slag presents remarkable influence on the change in the WP of EM, whatever the aggregate WP.

2. Aggregate WP ranking is preserved for EM WP whatever the other parameters studied (mineral admixture nature, moisture state and volume of aggregates) while not preserved when evaluating the effect of paste-aggregate interface on EM WP, due to the mineral admixture nature.

3. The dependence of the paste-aggregate interface structuration on the mineral admixture nature is supported by SEM/EDS characterization enabling the determination of the chemical length of ITZ.

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