A Quantitative Study on Packing Density and Pozzolanic Activity of Cementitious Materials Based on the Compaction Packing Model

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Abstract. A brief introduction to the theoretical basis of compaction packing model (CPM) and an over-view of the principle of the specific strength method provided the starting point of this study. Then, research on quantitative relations was carried out to find the correlation between the contribution rate of the pozzolanic activity and the contribution value of packing density when CPM was applied to fine powder mixture systems. The concept of the contribution value of the packing density being in direct correspondence with the contribution rate was proved by the compressive strength results and SEM images. The results indicated that the variation rule of the contribution rate of the pozzolanic activity is similar to that of the contribution value of packing density as calculated by CPM. This means the contribution value of the packing density could approximately simulate the change tendency of the contribution rate of the pozzolanic activity, which is of significant value for the future of mix designs for high and ultra-high performance concrete.

Key words: compaction packing model; specific strength method; packing density; pozzolanic activity; mix design

1. Introduction

In recent years, new components such as fibers, and various gel and chemical admixtures have been added to composite materials like high performance and ultra-high performance concrete, resulting in complicated concrete mix designs [1]. There have been high-performance concrete mix designs and optimization methods proposed [2-6], but the results are not yet perfect. More research is needed to fully understand the role of mineral admixtures and chemical additives, and the ideal mix proportions [7]. Due to the wide variety of high performance concrete admixtures and their computational complexity, the accuracy and economy of mix designs could be greatly improved with the use of computer technology [8]. It has become increasingly urgent to make full use of modern computer technology the relatively mature theory to establish a coordinated, reasonable, science-based method to develop high-performance concrete mix designs [9, 10, 11]. In this investigation, the quantitative relations between the packing density and the pozzolanic activity of high performance concrete were investigated based on the CPM, which offers a new choice for the mix design of high-performance concrete.
2. Cpm And Specific Strength Method

To date, there has been extensive research conducted to predict the packing density of a mixture of solid particles by using mathematical models in theory. For examples, the Aim-Goff model [12], and Furnas, Toufar, and Dewar models [13, 14] use mathematics for their theories, but most are based on the erroneous assumption that solid particles are of a single size. Compressible Packing Model, referring to CPM models, first proposed by De Larrard of France [15], obtained the actual packing density by calculating the virtual packing density according to the stacking structures of solid particles of different particle size distributions and by solving a nonlinear equation based on compacting Index K [15]. The CPM model considers the particle size distribution and the impact of different stacking methods on the particle packing density. It has been used worldwide, generally for making high performance concrete. Another valuable tool is the MATLAB program, which is prepared on the basis of the CPM model, and can calculate the particle packing density of multivariate powder mixtures [16, 17].

The specific strength method was initially used to describe the pozzolanic activity of mineral powders in high strength (HSC) and high performance concrete (HPC) [18], in which the strengths are governed by both the strength contribution of the hydration effect and of the pozzolanic activity. To explore the quantitative strength contribution of the two parts, indicators including the specific strength (Rspecific), the contribution rate of the pozzolanic activity (Ppozzolanic,%), the contribution rate of hydration (Phydration,%), and activity indicators (A) were put forward. Inactive admixtures were required to prepare a cement concrete to obtain a specific strength, which was used as the reference to calculate the strength contribution value and its contribution rate of the pozzolanic activity and hydration, respectively. This method has proved practical to quantitatively analyze the strength contribution of the pozzolanic effect of mineral powder in HPC. Less attention, however, was paid to the quantitative influence on the packing density [18].

In this investigation, the specific strength method was employed to quantitatively analyze the contribution value of packing density and the strength contribution rate of the pozzolanic activity. The difference between the packing density of the concrete with mineral powder or without mineral powder can be defined as the contribution value of packing density (Dpacking), which can be calculated in the equation:

\[ D_{\text{packing}} = D_{\text{doped}} - D_{\text{reference}} \]  

\( D_{\text{doped}} \) refers to the packing density of concrete mixed with mineral powder; 
\( D_{\text{reference}} \) refers to the packing density of the reference concrete;

Based on the CPM model, the packing density contribution was analyzed and the quantitative relationship between the packing density and the pozzolanic reaction was established accordingly in this study.

3. Experimental Program

3.1 Materials

| Performance          | PC   | SS   | FAI  | SF  |
|----------------------|------|------|------|-----|
| Density (g/cm³)      | 3.14 | 2.94 | 2.4  | 2.21|
| Average particle     | 13.9 | 12.96| 5.14 | 0.12|
| diameter (µm)        |      |      |      |     |

Table 1 Physical properties of the four cementitious materials
A type of Portland cement (PC), 52.5 grade, ordinary granulated blast furnace slag (SS), class I fly ash (FAI), silica fume (SF), and quartz sand were used in this study. The physical properties of PC, SS, FAI, and SF were tested and the results are shown in Table 1.

Quartz sand was 40-70 mesh with a density of 2.626g/cm³. Moreover, polycarboxylate superplasticizer was used in the study with its water reduction rate of approximately 30%.

### 3.2 Test Procedures

#### 1) Mixtures

In the tests mentioned in this paper, the total mineral powder dosage was controlled unchanged at 30%, with 5% as a variable. (If one mineral powder dosage increased from 5% to 25%, another was reduced from 25% to 5%.) To facilitate this study, the water/glue ratio throughout the test was fixed at 0.25, the sand/cement ratio at 1.3; and the superplasticizer dosage amounted to 1.0% of the cementitious material; the specific mixing ratios are shown in Table 2.

| Item | PC (%) | SF (%) | SS (%) | FAI (%) | packing density | compressive strength 3days (MPa) | compressive strength 28days (MPa) |
|------|--------|--------|--------|---------|-----------------|-------------------------------|-------------------------------|
| W0   | 100    | --     | --     | --      | 0.7489          | 47.3                          | 67.4                          |
| W1   | 70     | --     | --     | 30      | 0.7722          | 40.1                          | 72.0                          |
| W2   | 70     | 5      | --     | 25      | 0.7818          | 45.6                          | 80.3                          |
| W3   | 70     | 10     | --     | 20      | 0.7895          | 47.6                          | 84.5                          |
| W4   | 70     | 15     | --     | 15      | 0.7975          | 54.3                          | 87.9                          |
| W5   | 70     | 20     | --     | 10      | 0.8034          | 54.9                          | 91.2                          |
| W6   | 70     | 25     | --     | 5       | 0.8010          | 58.4                          | 89.1                          |
| W7   | 70     | --     | 30     | --      | 0.7518          | 45.3                          | 69.5                          |
| W8   | 70     | 5      | 25     | --      | 0.7567          | 51.5                          | 73.4                          |
| W9   | 70     | 10     | 20     | --      | 0.7708          | 53.5                          | 77.4                          |
| W10  | 70     | 15     | 15     | --      | 0.7789          | 56.7                          | 81.2                          |
| W11  | 70     | 20     | 10     | --      | 0.7846          | 59.8                          | 84.5                          |
| W12  | 70     | 25     | 5      | --      | 0.7932          | 61.1                          | 86.9                          |
| W13  | 70     | 30     | --     | --      | 0.7985          | 49.4                          | 87.9                          |

#### 2) Test Procedures

(1) Compressive strength test methods:

The specimen size was 40mm × 40mm × 160mm, and the compressive strength was measured and calculated according to ISO 679:2009 Cement - Test methods - Determination of strength. All samples were formed in the curing room for one day with mold conservation. Conservation was maintained after the mold was taken apart in the curing room for three days and 28 days, and then its compressive strength was tested.

(2) Packing density calculation:

A laser particle size analyzer was first used to measure each powder particle size distribution, and then the calculation of the packing density of the mineral powder mixtures was achieved through the MATLAB CPM model. For the convenience of the calculation of the packing density of the entire
concrete, it was assumed that the remaining packing density of the mix of each size was equal.

(3) SEM analysis:
The sample was observed and analysed by electron microscopy, which integrated micro and macro perspectives and provided a more detailed description of the test results.

4. Test Results And Analysis

4.1 Test Results
Table 2 lists the test results of the respective mix ratios.

4.2 Results and Discussion
1) Analysis of Packing Density and Compressive Strength
The changing trend of packing density and compressive strength of the PC-FAI-SF and the PC-SS-SF ternary systems at various mineral powder dosages are shown in Figure 1 and Figure 2, respectively, as calculated by the CPM model.

As can be seen in Figure 1, in the ternary system of the PC-FAI-SF, as the SF content increases and FAI dosage decreases, the packing density gradually increases. When the silicon ash content is kept at 20%, and the FAI content at 10%, the bulk density of the system reaches its maximum, which is due to the fact that the average particle diameters of the three materials (silicon ash, fly ash and cement), were in three different magnitudes, and thus the micro aggregate gradation was optimized for close packing and filling, with the mineral powder compacting effect at its full potential. The trend of the packing density of the ternary system of PC-FAI-SF that is revealed in Figure 2 presented a roughly similar change to the compressive strength trend at 28 days, which indicated that the compressive strength growth of the concrete at 28 days was closely related to the mineral powder compacting effect. On the contrary, the compressive strength growth of the concrete at three days was slightly different than the change of packing density. It was, in essence, a linear upward trend, which might be that the age of the concrete was too short and its early strength was primarily determined by the contribution of the cement hydration and the pozzolanic reaction of the silica fume.
Fig. 1 Effect of SF dosage on packing density

For the PC-SS-SF system, as can be seen in Figure 1, the silica fume increases, and the packing density shows a slightly different trend from that of PC-FAI-SF. It is largely straight up, with a significantly lower value than that of the PC-FAI-SF system. This is due to the fact that the particle size of the slag was larger than that of fly ash, and roughly the same size as the cement particle, resulting in a slightly lower grading of the PC-SS-SF than that of the PC-FAI-SF system. However, the composite incorporation of the slag and silica fume is still better than concrete with slag only. For the ternary system of PC-SS-SF, the growth trend of the compressive strength at three days and 28 days was consistent. In particular, its three-day compressive strength, as well as its compressive strength after hardening for 28 days, coincided with the bulk density trends, indicating that the growth of the concrete compressive strength was closely related to the mineral powder compacting effects.

Through a comparative study of the PC-FAI-SF and PC-SS-SF systems, it was found that the compacting effect of the mineral powder in the two systems was directly related to the compressive strength of the concrete, which shows that the increase of compressive strength of high-performance concrete is a result of both the mineral powder compacting effect and the pozzolanic reaction of secondary hydration. In the early stages, the hydration of the mineral powder plays a major role in the strength of the concrete, but in the latter stages, the compressive strength of the concrete depends primarily on the mineral powder compacting effect. As shown in Figure 2, when the same amount of silica fume was added, the compressive strength of the early PC-FAI-SF system was slightly lower than that of the PC-SS-SF system. However, as it aged, the compressive strength of the PC-FAI-SF system gradually became higher than that of the PC-SS-SF system. This is because in the early stage, the slag was more active than the fly ash, and the C-H generated by the cement hydration reaction became an alkaline activator, which accelerated the secondary hydration of the slag. The participation of the fly ash in the secondary hydration was much slower, contributing little to the compressive strength of the concrete in its early stages. Even though the PC-SS-SF system did not show good particle size distribution, the link between bulk density and compressive strength was masked by the chemical activity of the silica fume and slag in the early stages, thus the compressive strength of the early PC-SS-SF was greater than that of the PC-FAI-SF system. As it aged, the fly ash became more...
active and increased its contribution to the compressive strength, and complemented the particle gradation of the PC-FAI-SF system. In the late stages, the compressive strength of the PC-FAI-SF system was ultimately higher than that of the PC-SS-SF system. In addition, the silica fume itself largely improved the early strength of the concrete. As the silica fume increased, the development of the early strength gradually increased. For example, the PC-SS-SF system, when mixed with 5% silicon ash, showed the strength at three days was increased by 13.7%, and when the dosage was increased to 15%, the strength increased to 25.2% at three days.

Figure 3 shows the reference concrete and electron microscopy images of the ternary systems at 28 days. Graph (a) is the electron micrography of the reference concrete without mixing any powder. More coarse pores can be found between the hydration products of the reference concrete, and many flocculent CSH gel and slender rod-shaped crystals of ettringite can also be seen; apart from those, the layered Ca(OH)₂ crystal structure is relatively free and features overall looseness. Graph (b) shows the concrete of the PC-FAI-SF system under the electron microscope; the structure is clearly denser, the amount of fibrous calcium silicate hydrate gel is significantly reduced, and the gel is filled with a large number of mixed particles of various dimensions. Graph (c) shows the concrete of the PC-SS-SF system under the electron microscope; fibrous calcium silicate hydrate can be seen, but the fibers are smaller and more uniformly distributed; in addition, the fibers are interweaved and form a dense mesh, and in places where there is no fiber distribution, the gel is very dense, reflecting a high compressive strength when considered from a macro perspective.

In summary, when a certain dosage of mineral powder is added to the PC-FAI-SF and PC-SS-SF, the greater the bulk density of the mineral powder, and the greater the compressive strength of the system, proving a close correlation. And whether it was from a macro perspective or from the microscopic point of analysis, the packing density and compressive strength of the PC-FAI-SF system were superior to that of the PC-SS-SF system.

2) Pozzolanic Effect Analysis

Figures 4 and 5 indicate the changes of the contribution rate of the pozzolanic activity on the mineral powder under conditions of various aging times corresponding to SF dosage changes.

Judging from Figures 4 and 5, in the test dosage range, although doped species and aging times varied within the two systems, in general, the contribution rate of the pozzolanic activity on various systems increased with the increase of the dosage of silica fume.
Fig. 4 Effect of SF dosage on pozzolanic effect (at 3d)

For the ternary system of PC-FAI-SF, when the silica fume dosage exceeded 20%, the contribution rate of the pozzolanic activity at 28 days decreased slightly, which might be the reason that when the compacting effect of the mineral powder increased, the contribution rate of the pozzolanic activity decreased with the decline in the compacting effect. It is worth noting that the fly ash itself became relatively more active with age, and the particle size was between that of cement and silica fume, which was conducive to being denser and more closely stacked. Meanwhile, as it had a greater density than the silica fume, the fly ash was more beneficial to the strengthening of the cement. Thus, the pozzolanic activity of the mineral powder after 28 days presented a much more significant increase than that of the mineral powder after three days.

Fig. 5 Effect of SF dosage on pozzolanic effect (at 28d)
In the ternary system of PC-SS-SF, as the slag particle size was on the same order of magnitude as the cement, it did not work for dense packing. On the contrary, the silica fume particles could only be filled in a passive way between the slag particles, which helped fill the pores and condense the mix, thus contributing to the improvement of the cement strength. Therefore, in the ternary system, as the silica fume increased, the packing density was greater, and the contribution rate of the pozzolanic activity was greater.

A comparison of the two systems shows that in the instance of the same dosage of silica fume, the contribution rate of the pozzolanic activity at 28 days within the PC-SS-SF system was less than that of the PC-FAI-SF. However, at three days, its contribution rate was greater than that of the PC-FAI-SF system and the contribution rate of the pozzolanic activity tended to rise with the increase of silica fume. This is because fly ash is slightly less active than slag, and the early involvement of the secondary hydration reaction is slow. For the two systems, the chemical activity of the mineral powder plays a dominant role in the contribution rate of the pozzolanic reaction strength in the early stages, while the role of dense particles is not obvious. Although the particle size distribution of the PC-FAI-SF system is better than that of the PC-SS-SF system, the contribution rate of the pozzolanic effect strength in the early PC-SS-SF system is always greater than that of the PC-FAI-SF system. With the passage of time, advantages of the particle size distribution in the PC-FAI-SF system is reflected and the particle compacting effect plays a major role in the contribution rate of the pozzolanic activity. The changes of the contribution rate of the pozzolanic activity and the packing density gradually coincided in the two systems in the 28-day test, as is shown in Table 3. Thus, at 28 days, the contribution rate of the packing density can be modeled as the contribution rate of the pozzolanic activity.

Table 3 Specific strength, contribution rate of pozzolanic effect and of packing density

| Item | Specific strength of concrete cement quantity (MPa) | Specific strength of pozzolanic effect (MPa) | Contribution value of pozzolanic effect strength (%) | contribution value of packing density (10²) |
|------|-------------------------------------------------|------------------------------------------|-----------------------------------------------|------------------------------------------|
|      | 3days | 28days | 3days | 28days | 3days | 28days |                                 |                                  |
| W0   | 0.473 | 0.674  | --    | --     | --    | --     | 0                                |                                  |
| W1   | 0.573 | 1.029  | 0.100 | 0.355  | 17.43 | 34.47  | 2.33                             |                                  |
| W2   | 0.651 | 1.147  | 0.178 | 0.473  | 27.39 | 41.25  | 3.29                             |                                  |
| W3   | 0.680 | 1.207  | 0.207 | 0.533  | 30.44 | 44.17  | 4.06                             |                                  |
| W4   | 0.776 | 1.256  | 0.303 | 0.582  | 39.02 | 46.33  | 4.86                             |                                  |
| W5   | 0.784 | 1.303  | 0.311 | 0.629  | 39.69 | 48.27  | 5.45                             |                                  |
| W6   | 0.834 | 1.273  | 0.361 | 0.599  | 43.30 | 47.05  | 5.21                             |                                  |
| W7   | 0.647 | 0.993  | 0.174 | 0.319  | 26.91 | 32.12  | 0.29                             |                                  |
| W8   | 0.736 | 1.049  | 0.263 | 0.375  | 35.71 | 35.72  | 0.78                             |                                  |
| W9   | 0.764 | 1.106  | 0.291 | 0.432  | 38.11 | 39.04  | 2.19                             |                                  |
| W10  | 0.810 | 1.160  | 0.337 | 0.486  | 41.60 | 41.90  | 3.00                             |                                  |
| W11  | 0.854 | 1.207  | 0.381 | 0.533  | 44.63 | 44.17  | 3.57                             |                                  |
| W12  | 0.873 | 1.241  | 0.400 | 0.567  | 45.81 | 45.71  | 4.43                             |                                  |
| W13  | 0.706 | 1.256  | 0.233 | 0.582  | 32.98 | 46.33  | 4.96                             |                                  |

Through the above analysis, the contribution rate of the pozzolanic activity on the composite mixed mineral powder is not only related to the activity of the mineral powder itself, but also to the bulk
density of the concrete, which shared a similar variation to the contribution rate of the pozzolanic activity. Therefore, it is reasonable to model the contribution rate of the pozzolanic activity by the packing density contribution value.

3) Bulk Density Modelling the Pozzolanic Effects

It was shown in the study that there was good correlation between the mineral powder bulk density and the contribution rate of the pozzolanic activity, and the latter varied according to the change of the former. Thus, a mathematical fitting tool was used to fit the relationship between the contribution rate of the pozzolanic activity and the contribution value of the mineral powder intensity of each system under the conditions of 28 days and acquired the corresponding fitting formula. Although there was a certain correlation between its bulk density contribution value and the contribution rate of the pozzolanic activity for the concrete at three days, the actual engineering significance was of little use due to its short life span. As such, this article’s aim was to fit the relationship between the contribution rate of the mineral powder and its contribution value to bulk density at 28 days.

Figure 6 and Table 4 show the fitting diagram and the corresponding parameters and formulas between the contribution value of the packing density and the contribution rate of the pozzolanic activity under the standard curing conditions of 28 days.

As can be seen from Figure 6 and Table 4, the relationship between the contribution rate of the packing density and the contribution rate of the pozzolanic effect was a polynomial equation, with the correlation coefficient exceeding 98.5%, which shows a good correlation.

![Fig. 6 Relation between contribution rate of pozzolanic effect and packing density (at 28d)](image)

### Table 4 Relation between contribution rate of pozzolanic effect and packing density (at 28d)

| System Name | Fitting Formula | Correlation Coefficient | Standard Differential |
|-------------|-----------------|--------------------------|-----------------------|
| PC-FAI-SF   | $y=26.77+5.23x-0.24x^2$ | 0.99252 | 0.33797 |
| PC-SS-SF    | $y=33.38+2.78x+0.015x^2$ | 0.98742 | 0.63601 |

A coefficient of a polynomial equation is the slope of the curve, representing the rising trend of the packing density contribution value per unit to the contribution rate of the pozzolanic activity. Obviously, the better the particle size distribution, the more noticeable the contribution value of the packing density on the contribution value of the pozzolanic activity, with the equation coefficient of the PC-FAI-SF system equation coefficient greater than that of the PC-SS-SF system. However, the quadratic coefficient of the PC-FAI-SF system is negative, and this is because the increase in the bulk
density reveals a downward trend on the increase of the contribution rate of the pozzolanic activity within the PC-FAI-SF system.

In summary, the packing density had a strong correlation with the contribution rate of the pozzolanic activity of the composite mixed mineral powder. When cured for 28 days under standard conservation conditions, the contribution value of the packing density can be used to calculate the contribution rate of the pozzolanic activity, which provides a foundation for the future design of high-performance concrete mix.

5. Conclusion

The two systems, PC-FAI-SF and PC-SS-SF, were both mixed with dosages of different materials. Under a certain dosage, a greater packing density of the mineral powder correlated with a greater compressive strength of the system. And whether it was from a macro perspective or micro perspective, the packing density and compressive strength of the PC-FAI-SF system were greater than those of the PC-SS-SF system.

The contribution rate of the pozzolanic activity of the composite mixed mineral powder was not only related to the activity of the mineral powder itself, but also to the bulk density of the concrete, which shared a similar variation to the contribution rate of the pozzolanic activity. Therefore, it was reasonable to model the contribution rate of the pozzolanic activity by the packing density contribution value.

The packing density had good correlation with the contribution rate of the pozzolanic activity of the composite mixed mineral powder. When cured for 28 days under standard conservation conditions, the contribution value of the packing density could be used to calculate the contribution rate of the pozzolanic activity, which provided a new idea for the pursuit of future designs of high-performance concrete mix and even the ultra-high-performance concrete mix.

References

[1] M.F Alves, R.A Cremonini, D.C.C Dal Molin (2004) A comparison of mix proportioning methods for high-strength concrete. Cement and Concrete Composites, 26 (6): 613-621.
[2] F. de Larrard (1990) A method for proportioning high-strength concrete mixtures. Cement and Concrete Research, 12 (2): 47-52.
[3] H.-G. Kessler (1994) Spheres model for gap grading of dense concretes. BFT, (11): 73-75.
[4] F. de Larrard, T. Sedran (1994) Optimization of ultra high-performance concrete by the use of a packing model. Cement and Concrete Research, 24 (6): 997-1009.
[5] Chul-Hyun Lim, Young-Soo Yoon, Joong-Hoon Kim (2004) Genetic algorithm in mix proportioning of high-performance concrete. Cement and Concrete Research, 34 (3): 409-420.
[6] M.I. Khan (2012) Mix proportions for HPC incorporating multi-cementitious composites using artificial neural networks. Construction and Building Materials, 28 (1): 14-20.
[7] Konstantin Sobolev (2004) The development of a new method for the proportioning of high-performance concrete mixtures. Cement and Concrete Composites, 26 (7): 901-907.
[8] Ma Baoguo, Wang Xingang, Li Xiangguo, Wang Kai (2005) Design of High-performance concrete and its existing problems. Concrete, (2): 12-15.
[9] F. de Larrard, T. Sedran (1996) Computer-aided mix-design: Predicting final results. Concr. Int, (10): 39-41.
[10] Dewar J D (1999) Computer Modelling of Concrete Mixtures. London: E & FN Spon.
[11] M.F.M. Zain, Md. Nazrul Islam, Ir. Hassan Basri (2005) An expert system for mix design of high performance concrete. Advances in Engineering Software, 36 (5): Pages 325-337.
[12] R. Ben Aïm, P.Le Gof (1968) Effet de paroi dans les empilements désordonnés de sphères et application à la porosité de mélanges binaires. Hand-book Powder Technology, 29 (4): 281-290.
[13] W. Toufar, E. Klose, M. Born (1977) Berechnung der Packungsdichte von Kornemischen Aufbereit.-Tech,
[14] Jones M R, Zheng L, Newlands M. D (2002) Comparison of particle packing models for proportioning concrete constituents for minimum voids ratio. Materials and Structures, 35 (2): 301-309.

[15] De Larrard F, Sedran T (2002) Mixture-proportioning of High-performance Concrete. Cement and Concrete Research, 32 (10): 1699-1704.

[16] J.Q.Gong. Study on the gradation effect of ultra-high performance concrete: (Ph.D. Hunan University, China 2007).

[17] D.Mao. Study on the gradation effect of mineral powder in cement-based composite cementitious materials: (MS. Hunan University, China 2004).

[18] Pu Xincheng (1998) A numerical analysis of pozzolanic effect of the high-strength and high-performance concrete. Concrete, (6): 13-23.