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Packing density modeling of blended cement with limestone having different particle sizes

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• A mathematical model simulating the packing density of blended cement was derived.

• Cements with limestone of different particle sizes and various quantities were used.

• The model was derived based on a linear packing density of grain mixtures models.

• The predicted packing density was validated by experimental values.

• The modified model could accurately simulate the packing densities of the blended cements.

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1. Introduction

Over the years, there has been a growing interest in the development of a blended Portland cement, in which the amount of clinker is reduced and partially replaced by mineral additives. Limestone is one of the most attractive additives, because it is considered natural, available and economical. According to EN-197\1, all 27 common types of cement may contain up to 5% minor additional components (MAC), usually limestone. Nonetheless, there are four types of cement permitted to have a higher content of limestone in two replacement ranges, CEM II/A-L and CEM II/A-LL (6–20% limestone), as well as CEM II/B-L and CEM II/B-LL (21–35% limestone). The motivations for attempting to reduce the clinker content are threefold: (1) ecological benefits, due to lower emission of CO₂ into the atmosphere; (2) economic

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A mathematical model simulating the packing density of blended cement with limestone having different particle sizes added in various quantities was derived from models created by Stovall and de Larrard and was developed based on a linear packing density model of grain mixtures. The predicted packing density was validated by experimental values. Following our small and simple modifications, the Stovall and de Larrard models could accurately simulate the packing densities of blended cements with different particle sizes in various combinations, mainly those related to the interaction functions accounting for loosening l(r) and wall w(r) effects and the compaction index, *K*.

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benefits, due to cost reduction; and (3) technological benefits, due to the resulting improvement in the performances of both the cement and the concrete.

Partial replacement of the clinker by mineral additives was found to affect the performances of the blended cement with limestone: the initial and final setting times [1,2]; the hydration rate and degree [3-5]; the compressive strength after 1 day and 28 days [3,6]; the workability of the fresh paste [7]; the water requirement for reaching a normal consistency [8]; and the packing density of the blended cement [7,9,10]. The effect of partial replacement of the cement with additives was mostly shown by the influence of fine powders with sizes smaller than that of the clinker. For example, the addition of nano-limestone to accelerate the hydration of blended cement with fly ash [2] and the effect on the hydration rate and compressive strength caused by the partial replacement of cement with ultrafine particles with an average of $0.07 \,\mu m$ [3] were reported. There are fewer reports on the addition of coarse particle limestone (larger than or equal to the size of the clinker). It was shown by Bentz et al. [2] that the setting time of blended cement with 4.4 μ m limestone was lower than that with







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Nomenclature						
$egin{array}{l} \gamma_i & \ \mathcal{B}_i & \ \mathcal{I}(r) & \ \mathcal{W}(r) & \ \mathcal{Y}_i & \ \mathcal{Y}_$	virtual packing density if component <i>i</i> dominates packing density of component <i>i</i> loosening effect wall effect fractional solid volume of component <i>i</i> limestone particle size ratio	d _i φ Κ α b	diameter of powder particle in size group <i>i</i> real (experimental) packing density compaction index numerical constant value numerical constant value			

larger sized limestone particles (16.4 μ m). Earlier work [11] examined the setting history of blended cements with limestone of different sizes and in different quantities. Limestone particle size was found to have a substantial influence on the surface area and packing density of the resultant blended cement. The packing density has a tremendous effect on the fresh and hardened cement paste and on concrete performances [7–9,11–13], as indicated in many experiments.

The goal of this work is to provide a mathematical tool for predicting the packing density of blended cements with limestone in different sizes and quantities. The packing densities of two types of blended cements with limestone were investigated: limestone having single-sized particles (larger, smaller and similar to the original cement particles) and limestone having various combinations of particle sizes. This model was validated by means of experimental results in our prior works [11,13].

2. Modeling background

Several mathematical models were developed to predict the packing density of cement pastes containing particulate additives, as well as aggregates, in concrete [14–17]. Stovall et al. [17] developed a model to predict the packing density of multi-sized grain mixtures, given in Eq. (1):

$$\gamma_i = \frac{\beta_i}{1 - \sum_{j=1}^{i-1} \left[1 - \beta_i + w(r)\beta_i \left(1 - \frac{1}{\beta_j}\right)\right] y_j - \sum_{j=j+1}^n \left[1 - \frac{l(r)\beta_i}{\beta_j}\right] y_i}$$
(1)

where γ_i is the virtual packing density, if component *i* dominates; \mathcal{B}_i and \mathcal{B}_j are the packing densities of components *i* and *j*; y_i represents the fractional solid volume of component *i*; *r* is the size ratio between components *i* and *j*; and *l*(*r*) and *w*(*r*) are the interaction functions accounting for loosening and wall effects, respectively (described in the following).

Two different effects were introduced by the Stovall model that influence packing density the loosening effect and the wall effect.



Fig. 1. Reduction of the packing density by large (designated 1) and small (designated 3) grains in a ternary mixture [18]. In the diagram, the medium-sized grains (designated 2) are separated by the small-sized grains, causing a loosening effect, while they are being kept apart by the large grains, causing a wall effect.

The loosening effect describes a situation in which small particles present between larger particles, leading to the separation of the larger particles (Fig. 1). The wall effect describes a situation in which large particles cause interstitials in the grain mixture too small to be filled by other-sized particles; leaving voids with low packing of the particles around the perimeters of the larger particles (Fig. 1).

Several models were developed, based on the Stovall model, to predict the packing density of blended cement with different minerals mostly having a single particle size [19,20]. In order to precisely calculate the packing density with such models, one must first predict the loosening and wall effects. Yu et al. [21] developed a model to calculate the interactions between components *i* and *j* for loosening, l(r) and wall, w(r) effects, given in Eqs. (2) and (3), respectively. These equations were formulated based on the packing density of spherical particles only:

$$l(r) = 1 - (1 - r)^{3.3} - 2.8r(1 - r)^{2.7}$$
⁽²⁾

$$w(r) = 1 - (1 - r)^{2.0} - 0.4r(1 - r)^{3.7}$$
(3)

where *r* is the limestone particle size ratio.

de Larrard [18] expressed the loosening and wall effects according to Eqs. (4) and (5), respectively:

$$l(r) = \sqrt{1 - (1 - r)^{1.02}} \tag{4}$$

$$w(r) = 1 - (1 - r)^{1.5}$$
(5)

The loosening effect f and the wall effect g were also defined by Guo et al. [16] according to the following equations:

$$f(i,k) = 0.52 \left[\left(\frac{d_k}{d_i} \right)^{2.8} + 3.15 \left(\frac{d_k}{d_i} \right) \left(1 - \frac{d_k}{d_i} \right)^{2.9} \right] \quad k = i+1, \dots, n$$
(6)

$$g(i,k) = 1.13 \left(1 - \frac{d_i}{d_k}\right)^{1.0}$$
 $k = 1, \dots, i-1$ (7)

where d_i is the diameter of a powder particle in size group *i*.

Guo et al. calculated the packing density by measuring the fluidity of blended cement with slag through a cylinder. The measured packing density was obtained from the amount of water required to reach the same fluidity in samples with different particle-size distributions. The loosening and the wall effects (Eqs. (6) and (7)) were then used to calculate the predicted packing density of the blended cement with slag.

The loosening and wall effects were calculated for the range of particle size ratios presented in Fig. 2, using the three models described above (Eqs. (2)-(7)), of Yu et al., de Larrard, and Guo et al. The calculated values of the loosening effects, based on the de Larrard model, are larger than those obtained from the Yu model up to a size ratio of 0.55; from that point on an opposite trend is observed (Fig. 2a). However, the calculated values obtained by the Guo model are significantly lower than those obtained by the other two models. When comparing the calculated values for



Fig. 2. Interaction function for (a) loosening effect (b) wall effect.

the wall effects (Fig. 2b), a less significant difference is observed between the three models. Following the above comparison, the interaction effects vary for different particulates and must be adjusted relevant to each system, yet all are based on the Stovall linear packing density model for grain mixtures [17].

3. Packing density prediction

3.1. Existing model

In the current study, a packing density prediction was made for blended cements with limestone in various quantities and particle sizes: larger, similar and smaller than that of the plain cement. We based our calculations on the mathematical model developed by Stovall [17] (Eq. (1)), adapting the interaction functions of the loosening and wall effects developed by de Larrard [18] (Eqs. (4) and (5)), originally developed to predict a binary mix that fits well in our case. In the de Larrard model, the degree of compaction was defined by index *K*; the relation between ϕ (the real packing density) and *K* is given in Eq. (8):

$$K = \sum_{i=1}^{n} \frac{\frac{y_i}{\beta_i}}{\frac{1}{\phi} - \frac{1}{\gamma_i}}$$

$$\tag{8}$$

where y_i represents the volume fraction of the limestone in the blended cement; β_i – the packing density of the *n* elementary granular classes in the mixture; γ_i is the virtual packing density of the mixture, when *i* is considered as the main particle size (also see Eq. (1)). The interaction coefficient values 1.02 and 1.50 were calculated for the loosening and wall effects, respectively (Eqs. (4) and (5)) by the calibration of mono-dispersed aggregate systems, using the interaction effects l(r) and w(r) (Eqs. (4) and (5)) and compaction index, *K*, in Eq. (8).

To simulate the packing density of different blended cements with limestone according to Eq. (1), β_i must be determined. Doing so, the real packing density, ϕ , of the blended cement (Eq. (8)) may be defined as calculated by Eq. (9) (following Lecomte et al.) [19]. This method of calculation is based on experimental results that measured the amount of water required to reach a normal consistency according to EN-196/3. The assumption is that this amount of water is a consequence of the free voids between the particles, thus, indicating the packing density of the particles within the different blended cements:

$$\emptyset = \frac{1}{1 + \rho_P \frac{w}{p}} \tag{9}$$

where \emptyset is the experimental packaging density (the real packing density in Eq. (8)), *W* is the water requirement of normal consistency, *P* is the powder weight and ρ_p is the specific density of the dry powder.

Following this procedure (EN-196/3), the amount of water required to reach a normal consistency was measured for several different blended cements. CEM I 52.5 R with average particle diameter of 17 μ m was replaced with 30% limestone powders (>99.8% CaCO₃) having six different particle diameters—smaller than, larger than, or similarly sized to the original cement: 70 μ m, 53 μ m, 25 μ m, 23 μ m, 7 μ m and 3 μ m. For more details see Ref. [11].

The packing density, Ø, for each of the blended cements with the different aforementioned particle sizes and having 30% limestone were calculated using Eq. (9) based on the water demand measured to reach normal consistency, the values are presented in Table 1. The loosening, l(r) and wall, w(r) effects were calculated for all these blended cements according to Eqs. (4) and (5), respectively as were developed by de Larrard. The compaction index, K (Eq. (8)), was taken to be equal to 4.8, as was reported by Lecomte et al. [19] for similar mixtures used in this work (binary mixtures, cement-mineral admixtures). Therefore by knowing all required parameters for limestone amount of 30%, the values of β_i were determined by solving Eqs. (1) and (8). Knowing the β_i values, it became possible to calculate the packing density, γ_i , by means of the Stovall model (Eq. (1)) for blended cements with any limestone content and any particle size. The predicted packing density values of blended cements with 10% and 20% limestone and various sized particles were calculated and presented in Fig. 3. These values are compared with the experimental packing density values calculated based on Eq. (9) (Table 1). For more detailed of the water demand to reach normal consistency related to the particle sizes see Ref. [13].

From Fig. 3, according to the experimental results, an increase in the limestone particle size leads to greater packing densities; however, this is not the same for the predicted results. For the large- and medium-sized limestone particles (70 μ m, 53 μ m, 25 μ m, and 23 μ m), a good correlation between the predicted and the experimental values is observed, but for the small particle sizes (CC7 μ m, CC3 μ m), the predicted values do not fully fit with the experimental ones. Accordingly, the models developed by Stovall and de Larrard can accurately predict the packing densities of blended cements with large- and medium-sized limestone particles, but are less accurate in regard to blended cements with small particles (CC7 μ m, CC3 μ m), smaller than those of the cement.

3.2. Modified model

To better predict the packing densities of blended cements with particle sizes smaller than those found in the original cement, a modification of the existing models is essential. Our approach was to modify the loosening and wall effect values, since those values are highly dependent on the particle arrangement within the blended cement, which, in turn, depends on the ratio between

Table 1

Experimental packing densities and the water demand to reach normal consistency of the tested powders (original cement and limestone) for 10%, 20% and 30% limestone contents calculated by Eq. (9).

Median particle	Limestone content, % (by mass)						
size, µm	10		20		30		
	Packing density, Ø volume solid/total volume	Water demand, gr	Packing density, Ø volume solid/total volume	Water demand, gr	Packing density, Ø volume solid/total volume	Water demand, gr	
70	0.5426	137.5	0.5500	135.0	0.5670	127.5	
53	0.5382	140.0	0.5455	137.5	0.5529	135.0	
25	0.5294	145.0	0.5323	145.0	0.5352	145.0	
23	0.5294	145.0	0.5323	145.0	0.5352	145.0	
7	0.5294	145.0	0.5280	147.5	0.5309	147.5	
3	0.5252	147.5	0.5197	152.5	0.5067	162.5	

The original cement packing density is 0.5223 and the water demand is 147.5 gr.



Fig. 3. Calculated (Eq. (1)) and experimental (Eq. (9)) packing density with (a) 10% limestone and (b) 20% limestone.

the particle sizes. We based our modification on the trend presented in Fig. 2, showing differences in the values of the loosening and wall effects introduced by several models. As such, Eqs. (4) and (5) (developed by de Larrard) were modified to provide more accurate tools that fit better to the systems tested in this work for the prediction of blended cement packing density having a range of particles sizes larger and smaller than that of the original cement investigated in our work. The modified equations for the loosening and wall effects are given as Eqs. (10) and (11), respectively:

$$l(r) = [1 - (1 - r)^{a}]$$
(10)

$$w(r) = 1 - (1 - r)^{b}$$
(11)

where *a* and *b* are numerical constant values.

Knowing the values of the experimental packing density, ϕ (Table 1), the compaction index *K* = 4.8 and the limestone content, 30%, it was possible to determine the values of the constants, *a* and *b* (in Eqs. (10) and (11)). This was done by altering the values of *a* and *b* until the value of γ_i derived from Eq. (1) was equal to the value of γ_i derived from Eq. (1) was equal to the value of γ_i derived from Eq. (1) was equal to the value of α and *b* were found to be 0.306 and 0.800, respectively. Note that the values of *a* and *b* are mainly influenced by the size of the particles, thus different values are suggested here for fine particles than those suggested by de Larrard that dealt with aggregates. The obtained B_i values (in Eqs. (1) and (8)) of the modified model are presented in Fig. 4. The greatest packing densities of the limestone powders are observed for limestone with similar-sized particles to those of the original cement particles, while the limestone with smaller particles showed the lowest packing density.

Fig. 5 compares the modified loosening and wall effect values calculated by Eqs. (10) and (11) to those calculated based on de Larrard (Eqs. (4) and (5)), showing the expected differences between the two models. Compared to the de Larrard values, our modified values are greater for the loosening effect and smaller for the wall effect. It is also interesting to note that according to Fig. 5b the wall effect values verses the particle size ratios are behave slight differently when comparing the two models, de Larrard and the modified one (Eq. (11)). The modified model observed linear increase for the entire presented values, up to ratio 1.0; however according to de Larrard model the well effect values are linearly increase up to particle size ratio of 0.6, but above this ratio the trend is slightly changed showing more moderate increased behavior. These differences in between the two models might be explained based on the different tested systems. In the current



Fig. 4. The modified calculated values of β_i for limestone powders having different sizes.



Fig. 5. Comparison of the modified (a) loosening effect values (Eq. (10)) and (b) wall effect values to those of de Larrard (Eqs. (4) and (5)).

work fine powders were tested, while de Larrard tested aggregates of much larger particle sizes.

The packing density, γ_i , was calculated (Eq. (1)) for all the blended cements with different-sized particles using the modified loosening and wall effect values. These calculated results are compared in Fig. 6 to the experimental packing density values, ϕ (Table 1), of blended cements with 10% and 20% limestone, relative to the limestone particle sizes. A good correlation is clearly observed between the calculated and experimental values for both limestone contents, 10% and 20%. Here, both the calculated and the experimental values decrease with a reduction in the limestone particle size. This trend is opposed to the one presented in Fig. 3,



Fig. 6. Packing density, γ_i of the calculated results by the modified model and experimental results (Table 1) of blended cements with 10% and 20% limestone and various particle sizes.

where the predicted values of the small-sized particle systems increased, rather than decreased, relative to those of the medium-sized particle systems.

It may be concluded that the models of Stovall and de Larrard can accurately simulate the packing density of the blended cements investigated in our work in cases where only small and simple modification is required. The modified model developed in this work provides an accurate tool for predicting the packing density of blended cements containing any limestone particle size (larger, similar or smaller to that of the cement) for any required limestone content.

3.3. Blended cement with limestone having combined particle sizes

The packing density of blended cement is highly dependent on its particle size, as was clearly shown in the previous sections and our earlier works [11,13]. Combining different-sized limestone particles within the same blended cement creates a higher packing density. An increase in packing density may be obtained by achieving optimal proportions between the different-sized particles in the blended cement [8]; this result is preferable to the one attained for blended cement with single-sized limestone particle additives [11,13]. As such, the packing density of blended cements with combined limestone particle sizes was further explored. The original cement was partially replaced by limestone powder containing a combination of particle sizes, both larger and smaller than the original cement particles.

We examined the ability of the above-developed model to predict the packing density of those newly blended cements with the combined limestone particle sizes. To this end, we partially replaced the original cement with 'combined' limestone powders (having large-sized particles, CC70 μ m, and small particles, CC3 μ m), replacing 5%, 20% and 35% of the cement (as permitted according to EN-197/3). The various, examined blended and combined cements are presented in Table 2. For each different blended cement combination, the amount of water required in order to attain a normal consistency was measured (according to EN-196/3) and the experimental packing density was calculated by Eq. (9), as presented in Table 2.

The packing density, γ_i , of the blended cements with the combined particle sizes was predicted by Eq. (1) using the modified loosening and wall effect calculations described above in Section 3.2 (by Eqs. (10) and (11), where a = 0.306 and b = 0.800, respectively). The real packing density, ϕ , was calculated according to Eq. (8) (K = 4.8). Fig. 7 presents the predicted values, as compared to the experimental results for each of the three limestone quantities (5%, 20%, and 35%). Note that only the combinations that provided the greatest packing density for each quantity are presented. In these limestone systems due to the combination of several particle sizes the packing density is better than that of the original cement made of single size particles only. This means that by replacing the single size cement with the improved packed limestone powder the

Table 2

Experimental results for the packing densities of blended cements with limestone with large and small particle combinations in various quantities.

Limestone content (% by mass)	Combination of limestone sizes	Packing density, Ø volume solid/total volume
5	3.5% CC70 μm + 1.5% CC3 μm 3.0% CC70 μm + 2.0% CC3 μm	0.5391 0.5329
20	17.0% CC70 μm + 3.0% CC3 μm 16.0% CC70 μm + 4.0% CC3 μm	0.5623 0.5571
35	30.0% CC70 μm + 5.0% CC3 μm 28.0% CC70 μm + 7.0% CC3 μm	0.5776 0.5748



Fig. 7. Calculated by the modified model and experimental packing density of blended cements with 5%, 20% and 35% limestone and combined particle sizes.

packing density of the entire blended cement is greater, therefore as the content of the limestone increases the packing density of the blended cement is increased accordingly (Fig. 7). The same trend – increased packing density with increased limestone content – was obtained for both the calculated and the experimental values; however, there are some differences in the packing density values between these two effects. Based on these results, the modified model cannot fully simulate the packing density of blended cements with combined particle sizes and some further modification is required.

Following Lecomate et al. [19], who found small differences in the K index for various tested cements, the compaction index K (Eq. (8)) for the combined particle size cements was slightly changed. Lecomate et al. [19] showed, for example, that the best fit between the real and the simulated packing density of blended cement with fly-ash was obtained by K = 5.0. Several values of the K index were examined and the best fit was obtained with K = 5.4. The calculated packing density values (Eq. (1)) using K = 5.4 (Eq. (8)) for blended cements with combined particle sizes are presented in Fig. 8; they are compared to the experimental packing density values (Table 2) relative to the size combinations for 5%, 20% and 35% limestone contents. Good prediction of the packing density is observed for all the studied, blended and combined (limestone) cements using K = 5.4. Note, however, that the high (35%) limestone cement showed a slight deviation between the calculated and the experimental data, which is most significant in cases of very fine particle cements containing only 3 µm particles. According to Kwan [7], when fine powders are used, strong inter-particle forces exist, causing agglomeration and the loosening of particle packing in cementitious materials. This being the case may make the measurement of the packing density by means of the water requirement difficult and somewhat problematic in those blended cements having a very high content of fine powder; such a situation may yield a slight deviation between the calculated and the experimental values (Fig. 8c).

Fig. 8 emphasizes several trends that can be predicted by our modified model (based on the Stovall and de Larrard models):

i. The packing densities of blended cements with larger limestone particles than those of the original cement are greater than that of the original cement, mainly for blended cements with high quantities of limestone (20% and 35%; Fig. 8b and c) and vice versa; the packing densities of blended cements with smaller limestone particles than those of the original cement are lower than that of the original cement with a high limestone content (35%).



Fig. 8. Calculated packing density (K = 5.4) and the experimental packing density of the blended cements with different limestone contents (a) 5%, (b) 20%, (c) 35%.

- ii. Optimal packing densities are obtained by means of specific particle-size combinations for each different quantity (5%, 20%, 35%) of cement replaced by the limestone, as also reported by Gallias et al. [8] Optimal powder packing was obtained at a ratio of 0.2 between the small and the large particles (CC3/CC70) in high limestone-content cements (20% and 35%). For the low-content blended cements, this ratio is much higher, 0.4. That is to say, that in blended cements with small quantities of limestone additives, a greater amount of fine particles is required to promote their dense packing, whereas in high-content limestone blended cements, fewer fine particles are needed to pack the powder well.
- iii. A high content of small limestone particles greatly decreases the packing density of blended cements, especially with 20% and 35% additives. This means that too much fine powder,

relative to the larger particles, stymies the creation of a dense powder arrangement.

4. Conclusions

Packing densities of blended cements with limestone were calculated on the basis of models developed by Stovall and de Larrard. Blended cements with single-sized limestone particles, larger than, similar to and smaller than the original cement particles were examined, as well as blended cements with various combinations of large and small particles. The calculated packing density values of the different blended cements were correlated with experimental data obtained by measuring the amount of water required to reach a normal consistency.

After our aforementioned investigations and minor modifications, it may be concluded that the Stovall and de Larrard models accurately simulate the packing density of blended cements with different-sized additive particles in various combinations, mainly those related to the interaction functions accounting for loosening l(r) and wall w(r) effects and the compaction index K.

The loosening and wall effects developed by de Larrard can accurately predict the packing densities of blended cements having similar- or larger-sized particles than the original cement; however, the de Larrard model does not fit accurately enough to predict the packing densities of blended cements with fine-sized particle, smaller than those of the original cement. The modification of the interaction functions, e.g., loosening and wall effects, was mainly obtained by adjusting the coefficient values. Values of 0.306 and 0.800 for *a* and *b* in Eqs. (10) and (11), respectively, were found to best fit the packing density calculations for blended cements with the various particle sizes investigated in this work.

The compaction index, K (Eq. (8)), varied slightly between single-sized particle blended cements and combined-sized particle blended cements; the K values of 4.8 (following de Larrard) and 5.4 (following Lecomate et al.) were employed, respectively.

The modified model developed in this work provides an accurate tool for predicting the packing densities of blended cements containing any limestone particle size (larger than, similar to or smaller than that of the original cement) in all the various quantities and combinations. Simulating such kind of blended cements made with limestone of different particle sizes (not only one size) will enable to design more sustainable blended cements with low clinker content and improved properties.

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