The use of particle packing models to design ecological concrete

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Ecological concrete can be designed by replacing cement with fillers. With low amounts of cement it becomes increasingly important to control the water demand of concrete mixtures. In this paper a cyclic design method based on particle packing is presented and evaluated on the basis of experiments on cement pastes combined with quartz powder. The packing density of sixteen pastes is measured using the mixing energy test. Furthermore, the cement pastes were tested on compressive strength at 7 and 28 days. Adding quartz powder M600 can increase the packing density by more than 10%. This means that fine quartz powders can improve the packing density to such extent that the cement content can be decreased while simultaneously the water cement ratio is decreased. This occurs for pastes with a packing density higher than the completely saturated packing density of cement. Additional strength tests were performed on two mixtures with constant water cement ratio and showed a 15% strength increase when 20% of the cement was replaced by quartz powder M600. In the design procedure for ecological concrete, increased strength efficiency can be balanced by lowering the amount of cement.

Key words: Cement paste, ecological concrete, filler, particle packing, water demand

1 Introduction

In concrete production, Portland cement is the component with the highest environmental impact. This is because the production of Portland cement requires high amounts of energy and CO_2 is released when limestone is transformed to calcium oxide during the burning process. Energy consumption and CO_2 emissions of concrete can be reduced when by-products from other industries, like fly ash, are applied as cement replacing materials or fillers. By this strategy not only the CO_2 emissions are reduced but residual products from other industries are reused and therefore less material is dumped as landfill and more natural resources are saved. Many by-products from other industries, like silica fume, fly ash or blast furnace slag, have characteristics which can positively influence the concrete properties. However, regulations limit the use of large amounts of cement replacing materials, requiring a minimum amount of cement in concrete (e.g. 260 kg/m^3 in the Netherlands) to make sure that concrete properties such as strength and durability are at a sufficient level. Nevertheless, it is believed that by making use of the particle packing optimization techniques, nowadays used in the production of high strength concrete, it is possible to optimize the particle packing in order to lower the cement content in concrete without changing concrete properties in a negative way [1]. This low cement content concrete or so called ecological concrete can be designed by optimizing the concrete composition in such a way that the highest packing density is achieved. The optimization results in a stiff and strong particle structure, which has a positive influence on the mechanical properties such as shrinkage and creep. Furthermore, the high density of the particle structure leaves less space for voids to be filled with water, which reduces the water demand and increases the strength of concrete. The water demand of the particle structure is very important since a slight increase in water demand in mixtures with low cement content results in higher water cement ratios. A higher water cement ratio will decrease strength and durability. Therefore, it is important to have a design method which can control the water demand of an ecological concrete mixture at a certain desired workability. In this paper such a design method, based on particle packing and the workability of cement pastes, is presented and evaluated on the basis of experiments on cement pastes combined with quartz powder.

2 Particle packing theory

The subject of optimizing the concrete composition by selecting the right amounts of various particles has already aroused interest for more than a century. To optimize the particle packing density of concrete, the particles should be selected to fill up the voids between large particles with smaller particles and so on, in order to obtain a dense and stiff particle structure. Most of the early researchers, working on the packing of aggregates, proposed methods to design an ideal particle size distribution, like Fuller in 1907. Optimizing concrete to a predefined 'ideal' particle size distribution is still popular today, as it is the method that is most used in practice and applied in most national standards. However, the ideal particle size distribution depends on the particle type, thus varying for each type of concrete. For instance, when rounded sand is combined with coarse recycled aggregates, the optimal particle size distribution will differ from the one of a mixture with sand from crushed rock and rounded coarse aggregates. The solution to find the ideal grading curve for each combination of different aggregates is to include the geometry of the particles by making use of a particle packing model.

The basic mathematical formulae of almost all particle packing models are the same and purely based on the geometry of the particles. The formulae prescribing the packing density were first introduced by Furnas in 1929 [2]. They are valid for two monosized groups of particles without interaction between the particles. The volume of each of these monosized particle groups can be expressed in its partial volume φ_i, which is the volume occupied by size class i in a unit volume. Furthermore, the relative volume of each size class can be expressed as its volume fraction r_i. By definition $\sum_{i=1}^{n} r_i = 1$ and $r_i = \varphi_i \Big/ \sum_{i=1}^{n} \varphi_i$. (A list of symbols is presented at the end of this paper.)

When there is only one size class present ($r_1 = 1$), the partial volume of this size class (φ_1) is equal to the total occupied volume or total packing density α_t . When there are two size classes present, the following two cases can be distinguished.

Case 1: The volume fraction r of the large particles is much larger than the volume fraction of the small particles ($r_1 >> r_2$).

In this case small particles (diameter d_2) can be added to a container filled with large particles (diameter d_1). By adding the small particles into the voids between the large particles, the voids are filled and thus the total occupied volume and the packing density increase. The total volume occupied by particles in a container [3,4] is expressed by equation 1.

$$\alpha_t = \varphi_1 + \varphi_2 = \alpha_1 + \varphi_2 \quad \rightarrow \quad \alpha_t = \frac{\alpha_1}{1 - r_2} = \frac{\alpha_1}{r_1} \tag{1}$$

Summarizing: The total packing density equals the volume of the large particles (which is restricted by the maximum packing density of the large particles) plus the volume of the small particles, in a unit volume.

Case 2: The volume fraction of the small particles is much larger than the volume fraction of the large particles ($r_2 >> r_1$).

In this case large particles can be added to a container filled with a matrix of small particles. By adding some large particles into a matrix of small particles, the large particles fill up the volume they occupy by 100%. Their contribution to the packing density is therefore equal to their partial volume: φ_1. The small particles can fill up the rest of the unit volume ($1-\varphi_1$) with their maximum packing density:

$$\alpha_t = \varphi_1 + \varphi_2 = \varphi_1 + \alpha_2 (1 - \varphi_1) \quad \rightarrow \quad \alpha_t = \frac{1}{r_1 + (r_2/\alpha_2)} \tag{2}$$

Summarizing: The total packing density equals the volume of the large particles plus the remaining volume filled with the maximum amount of small particles (which is restricted by the maximum packing density of the small particles), in a unit volume.

The packing model proposed by Furnas can be extended to *n* particle groups by making use of the geometrical and physical relations. It is physically not possible to have a large volume fraction of small particles ($r_2 >> r_1$) and simultaneously fit all the small particles within the voids between the large particles. Therefore, in this situation automatically case 2 becomes operative and particle group 2 becomes the dominating particle group. Thus, this physical relation comprehends that the packing density is always the minimum value of formula 1 and 2. The same 'minimum'-relation is true for *n* particle groups. With this additional relation the particle packing model for multi component mixtures, with dominant particle group *i* and remaining particle groups *j*, becomes [4]:

$$\alpha_t = \underset{i=1}{\overset{n}{Minimum}} \left\{ \alpha_i + (1 - \alpha_i) \sum_{j=1}^{i-1} \varphi_j + \sum_{j=i+1}^{n} \varphi_j \right\} \tag{3}$$

Since these formulae depend on the particle packing and amount of particles of the monosized groups, they are valid for any type of particle and automatically include particle characteristics such as shape and texture, as long as the particles preserve their shape during packing. Unfortunately, these formulae are only valid to calculate the particle packing of particle groups without any interaction. However, in reality there will always be interaction between the particles:

Wall effect: The effect of large particles on the packing density of small particles. This effect appears close to a wall or close to a large particle, where smaller particles can not be packed as densely as their maximum packing density.

Loosening effect: The effect of small particles on the packing of large particles. This effect appears when the small particles are too large to fit in between the voids between the large particles. The disturbance of the packing of the large particles by the smaller particles is called the loosening effect.

The effectiveness of the particle packing model therefore depends entirely on how these interaction effects are applied in the model. De Larrard [4] implemented these effects quite effectively for various types of rounded and crushed aggregates by introducing interaction parameters a_{ij} and b_{ij}, Equation 4.

$$\alpha_t = \underset{i=1}{\overset{n}{Minimum}} \left\{ \frac{\alpha_i}{1 - \sum_{j=1}^{i-1} \left[1 - \alpha_i + b_{ij} \alpha_i (1 - 1/\alpha_j) \right] r_j - \sum_{j=i+1}^{n} \left[1 - a_{ij}\, \alpha_i / \alpha_j \right] r_j} \right\} \tag{4}$$

3 Theoretical design method for cement pastes

Particle packing optimization techniques, such as the model presented above, help us to design an optimal particle structure of concrete. However, to make concrete flowable a certain amount of water is required. This is the amount of water necessary to fill the voids between the particles plus an extra amount of water that creates a small water layer surrounding the particles. With an increased particle packing density the total volume of the voids is decreased, and the amount of water can be reduced. Furthermore, previous experiments [4, 5] have shown how the amount of water in the mixture ($1-\varphi_{mix}$) can be related to the viscosity of the mixture and the maximum particle packing density. Figure 1 and 2 present the relation between the relative amount of water, expressed as φ_{mix}/α_t, to the viscosity of cement pastes and the flow value of mortars.

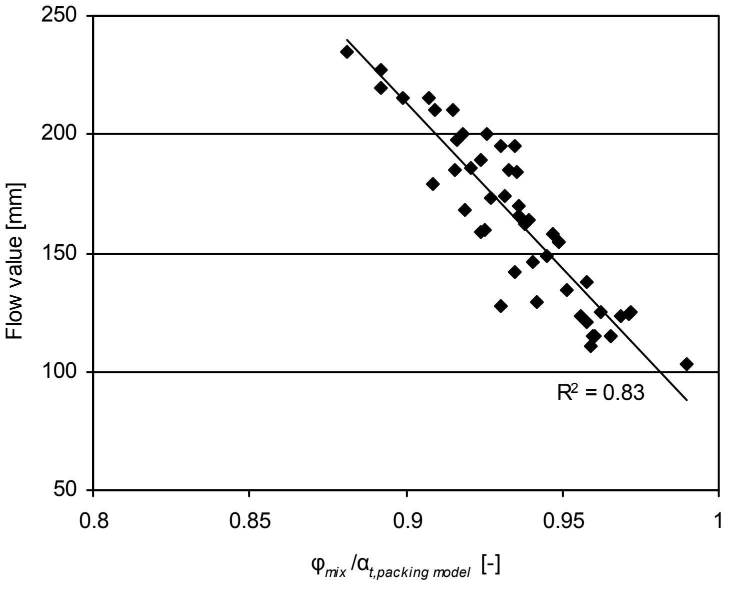

Figure 1: Flow value related to the ratio of solid volume of a mortar and the maximum packing density of that mortar as calculated by particle packing calculations (CPM [4])

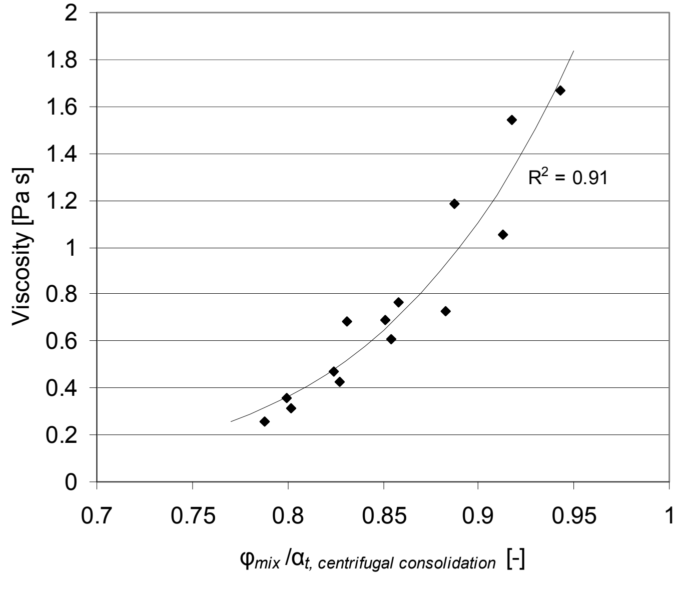

Figure 2: Viscosity related to the ratio of solid volume of a cement paste and the maximum packing density of that paste as determined by centrifugal consolidation [5]

The relation between φ_{mix} / α_t and the flowability can be used to optimize ecological concrete at a fixed workability. At a certain viscosity, φ_{mix} / α_t is constant. If α_t is maximized by particle packing optimization techniques, the total amount of particles in the concrete mixture (φ_{mix}) can increase and less water is required (1- φ_{mix}). The result is a lower water cement ratio of the mixture. A low amount of water in the mixture will result in either an increase of the strength at a constant cement content or the same strength at a reduced cement volume.

From the previous section it follows that the solution to an ecological concrete design lies in improving α_t. This can be done by, for instance, adding fillers to the concrete. (For convenience the term 'filler' is used for small particles in the remaining part of this paper; however, for the design of ecological concrete, the filler can just as well be a binder.) The influence of the addition of the small particles is explained on the basis of a two component mixture consisting of cement and filler. Starting from a mixture with 100% cement particles and the voids completely filled with water, but no excess of water, small particles can be added to the mixture. For each ratio of cement and filler a theoretical packing density can be calculated for which the voids remain exactly filled with the available amount of water. The mixture remains to be completely saturated, as presented in Figure 4 by the $\alpha_{saturation}$ line. The real packing density of a mixture containing cement combined with filler can also be higher or lower than this packing density, resulting in the following three cases (see also Figure 3):

Case 1: Adding small particles results in a higher packing density than the theoretical packing density necessary for a completely saturated mixture.

In this case small particles 'fit' in the voids between large particles. Because the small particles are positioned in the voids between the large particles, less water is needed to fill these voids. The water, which was already present in the mixture before adding the filler, will not only fill the voids but there will also be an excess amount of water in the mixture. For concrete design this result provides the possibility to either decrease the amount of water in the mixture (design of high performance concrete) or decrease the amount of water and binder (design of ecological concrete). This case can only exist for low ratios of d_2 and d_1 with little particle interaction.

Case 2: Adding small particles results in the theoretical packing density necessary for a completely saturated mixture.

By definition in this case all voids are exactly filled with water, so the mixture is completely saturated. Since the total volume of the particles in the mixture increases by adding the filler, also the particle packing of the mixture needs to increase to keep a constant void volume, Equation 5.

$$\alpha_{saturation} = \frac{V_{cement} + V_{filler}}{V_{cement} + V_{filler} + V_{water}} = \frac{V_{cement} + V_{filler}}{V_{cement} + V_{filler} + \left(\frac{V_{cement}}{\alpha_{cement}} - V_{cement} \right)} \tag{5}$$

Case 3: Adding small particles results in a lower packing density than the theoretical packing density necessary for a completely saturated mixture.

In this case particle interaction is high and small particles do not fit in the voids between large particles. The small particles will push the larger particles away from each other (loosening effect) to such an extent that extra water is needed to fill the voids. The water, which was already present in the mixture before adding the filler, will not be able to fill up the voids completely. For concrete design this case is undesirable. The workability of the mixture can only be guaranteed when the water cement ratio is increased.

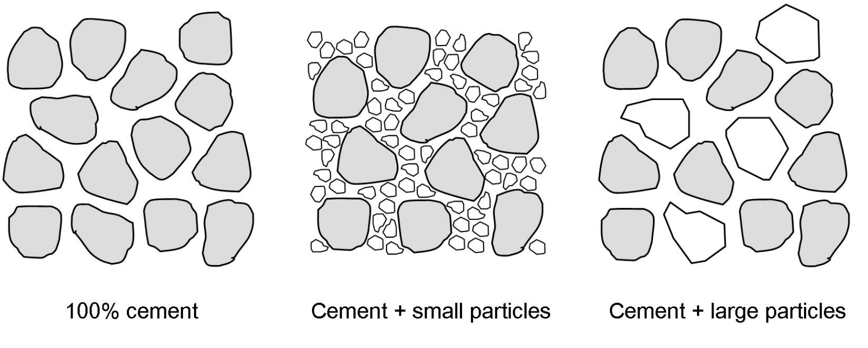

Figure 3: Schematic particle packing configurations. Small particles fit in the voids between the cement particles, thus increasing maximum packing density (case 1). Large particles do not fit in the voids between the cement particles. At constant packing density, large particles increase the average distance between cement particles (case 3).

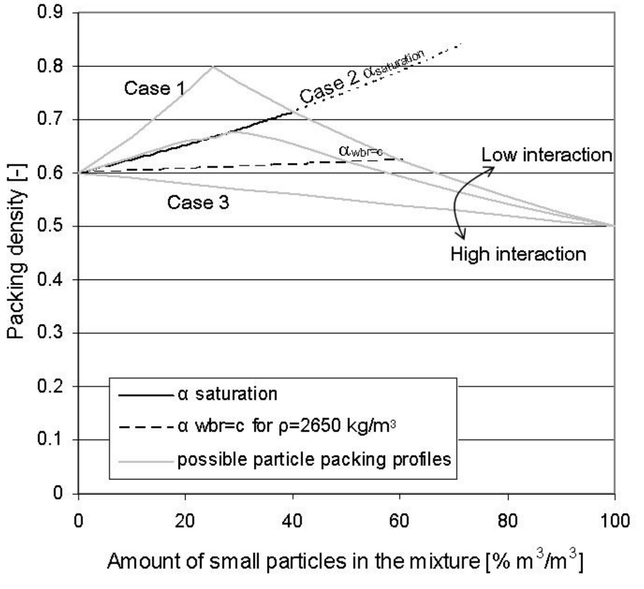

Figure 4: Particle packing profiles with high, low and intermediate interaction in relation to the theoretical design cases for completely saturated mixtures and mixtures with a constant water binder ratio

Figure 4 shows how these three cases can be used to design ecological concrete for cement with a packing density of 0.6. Case 1 is the only desired case for ecological concrete design solely based on packing density. The packing density of the mixture should at least be higher than $\alpha_{saturation}$, Equation 5.

A special case might be considered when the reactivity of the new binder (cement and filler) remains constant though cement is replaced by filler. In that situation packing densities can be lower than $\alpha_{saturation}$ but the water binder ratio needs to remain constant or decrease to ensure at least the same strength. This means that the packing density of the ecological mixture needs to be higher than the packing density at constant water binder ratio ($\alpha_{wbr=c}$), Equation 6 and Figure 4. Additional experiments such as strength tests or isothermal conduction calorimetry are required to determine the reactivity of the new binder (cement and filler).

$$\alpha_{wbr=c} = \frac{V_{cement} + V_{filler}}{V_{cement} + V_{filler} + V_{water}} = \frac{V_{cement} + V_{filler}}{V_{cement} + V_{filler} + \left(\frac{V_{water}\rho_{water}}{V_{cement}\rho_{cement}} \left(V_{cement} + V_{filler} \right) \right)} \tag{6}$$

4 Experiments

In practice it might not be possible to reach the theoretical saturation packing density by adding filler to cement. This is because cement and filler are no monosized fractions, which results in increased geometrical interaction between the fractions. Furthermore, inter-particle forces such as van der Waals forces and electrical forces will influence the packing density. To investigate whether packing densities above the saturation packing density can be expected in practice, the water demand of sixteen pastes is tested. Additionally, the cement pastes are tested on compressive strength.

4.1 Set up for packing density measurements of fine powders

The maximum packing density of dry particles can be determined according to NEN-EN 1097-3 [6] for loose bulk density. The method can be extended to determine the maximum packing density at a certain compaction level, by applying external loads such as vibration or top-weight. However, with fine particles (< 1 mm), the inter-particle forces become increasingly important. These inter-particle forces can cause, for instance, agglomeration of particles, thus lowering the packing density. Since the inter-particle forces depend on the conditions (dry, wet, use of superplasticizer etc.) of the packing structure, also packing density is influenced by this. Therefore it is important to measure the maximum packing density of the particles under the same conditions as under which the particles would be used in concrete.

The maximum packing density of wet particles is determined by mixing energy measurements [7,8]. In this method water is added slowly to a powder mixture. When the water is added it condenses on the dry particles to form capillary bridges (pedular bonds) localized at the particle contacts. In this way, agglomerates of particles are formed. The strength of the pendular bond increases with the liquid-vapor surface energy and depends inversely on the square of the particle diameter. At less than complete saturation, the strength of the agglomerates increases with the amount of liquid and the surface energy of the liquid. When enough water is added to fill all the voids, the absence of internal liquid-vapor surfaces at 100% saturation causes the strength to suddenly decrease at this point [9]. The mixing energy method is based on the idea that the differences in internal pendular bond strength can be measured by recording the mixing energy during mixing of the powders. The mixing procedure started by mixing the dry powders while adding (an estimated) 95% of the water and the superplasticizer in a three litre Hobart mixer.

After mixing for one minute the mix rested for two minutes. If necessary, powder adhering to the wall was scraped from the bowl during this resting period. Then, mixing was continued with a constant water supply of ± 0.3 ml/s in order to reach the saturation point in about 2 minutes. During mixing, the voltage, electricity consumption and the phase shift between the voltage and the electricity consumption of the mixer are registered to determine power use, Figure 5. The saturation point is recorded as the water to powder ratio at which maximum power use is measured. The test is repeated after which the average volume percentage of powders in the mixture at the saturation point over two tests is recorded as the maximum packing density. The accuracy of the test method expressed as packing density is estimated at ± 0.001 [10].

4.2 Materials and mixture compositions

The maximum packing density of sixteen pastes was tested by the mixing energy method. The mixtures consisted of cement (CEM I 42.5 N ENCI Maastricht), quartz powder (M6, M300, M600 Sibelco) or a combination of both. The addition of quartz powder was 13, 25, 51 or 79% [kg/kg] of the total cement volume. Quartz powder from three different size classes was used, as presented in Figure 6. To minimize the differences in mixing energy between the mixtures, all mixtures consisted of 1500 grams of powder to which the water

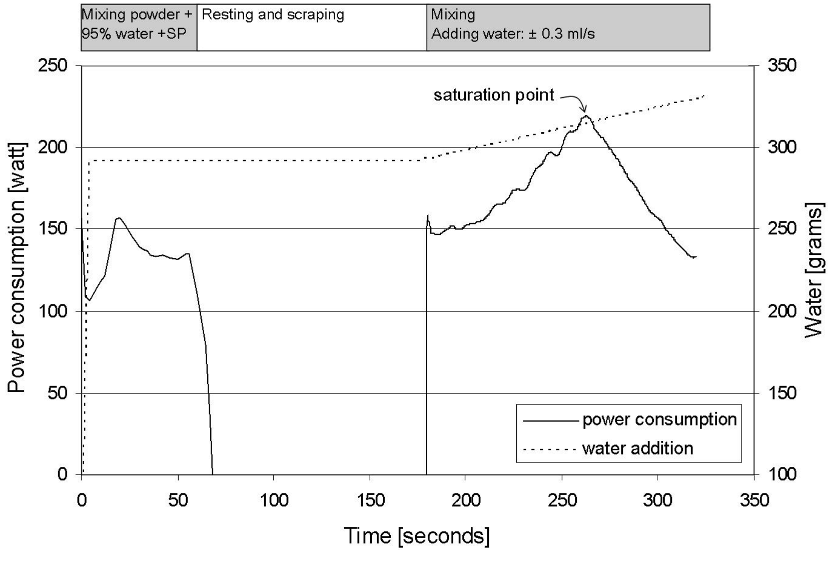

Figure 5: Power consumption and water supply during the mixing process of mixture A5

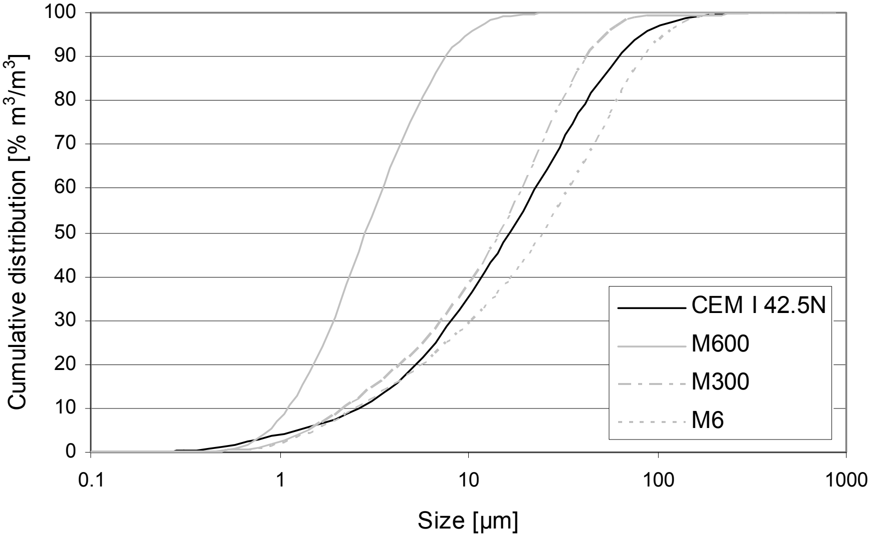

Figure 6: Particle size distributions of cement and quartz powder in ethanol, laser diffraction measurements

and superplasticizer (Glenium 51 BASF con.35%) were added according to the procedure described in the set up. For mixture compositions see Tabel 1.
For the strength tests the same powder compositions were tested according to NEN-EN 196-1 [11] including the same amount of superplasticizer (1.2% per gram cement) and with an adjusted water content to ensure stability of the mixtures. The mixture compositions are presented in Table 2. The prisms were tested for cube compressive strength at 7 and 28 days.

4.3 Results

Figure 5 shows the result of one of the mixing energy tests of mixture A5. Power consumption and added amount of water are expressed over time. The total amount of water (including the water from the superplasticizer) is determined at the point of maximum power consumption. Packing density is calculated as the volume percentage of powders in the mixture at this point and presented in Table 1, as average over two tests. Adding the coarse filler M6 slightly improves the packing density. When M300 to the cement is added, α_t is decreased a little. The addition of M600 shows a significant improvement of the packing density. Adding 25% or 51% of M600 can increase the packing density of cement by more than 10%.

Strength tests show the highest 28-day strength for the reference mixture B1 with 100% of cement. Adding quartz powder M6, M300, M600 only decreases the 28-day strength of the mortar specimens. According to these results a higher reactivity of the new types of binder (consisting of cement and quartz powder) is not presumable.

5 Discussion

5.1 Experimental results M6 and M300

In Figure 7 the measured packing densities from Table 1 are presented in relation to the completely saturated packing density for a cement paste with maximum packing density of 0.605 (CEM I 42.5 N). Also the packing densities for cement pastes combined with quartz powder with a constant water binder ratio are presented as $\alpha_{wbr=c}$. The graph

Table 1: Mixture compositions and averaged results of mixing energy tests

Mix	Cement	Quartz powder			Glenium 51	α_t
		M6	M300	M600		
	[g]	[g]	[g]	[g]	[g]	[-]
A1	1500				18	0.605
A2	1350	150			18	0.605
A3	1200	300			18	0.608
A4	1050	450			18	0.614
A5	900	600			18	0.612
A6	1350		150		18	0.602
A7	1200		300		18	0.604
A8	1050		450		18	0.600
A9	900		600		18	0.602
A10	1350			150	18	0.649
A11	1200			300	18	0.668
A12	1050			450	18	0.668
A13	900			600	18	0.664
A14		1500			18	0.583
A15			1500		18	0.542
A16				1500	18	- *

** Required mixing energy is too high for correct measurement* shows that adding quartz powder M6 and M300 does not increase the packing density enough to retain a constant water binder ratio. Basically, adding this type of particles to a mixture (or replacing cement with these particles) will lead to mixtures requiring a higher water binder ratio. Adding relatively more water to the mixtures will lead to lower strengths. Furthermore, the strength tests according to NEN-EN 196-1 of these mixtures do not show to have a constant (or increased) reactivity of the binder. Since M6 and M300 are in the same size range as CEM I 42.5 N these particles do not act as fillers, but more as small aggregates placed between the cement particles. This configuration of particles increases the distance between the cement particles, Figure 3. For that reason higher (or constant) reactivity of the binder is not expected. To verify this, reactivity tests performed on cement with low amounts of filler and sufficient amount of superplasticizer are recommended.

To design ecological concrete, cement particles are replaced by fillers. Replacing cement by M6 or M300 will lead to an increased water binder ratio, a decreased reactivity of the binder and a particle configuration in which cement particles are pushed away from each

Table 2: Mixture compositions and results of strength tests

Mix	Cem I	Quartz powder			Water	SP	Sand	Compressive strength	
	42.5 N	M6	M300	M600		1.2%		7 days	28 days
	[g]	[g]	[g]	[g]	[g]	[g]	[g]	[MPa]	[MPa]
B1	900				326	10.8	2700	50.1	64.5
B2	810	90			326	10.8	2700	42.8	59.6
B3	720	180			326	10.8	2700	36.8	49
B4	630	270			326	10.8	2700	33.6	42
B5	540	360			326	10.8	2700	24.1	34.7
B6	810		90		326	10.8	2700	47.6	61.6
B7	720		180		326	10.8	2700	37.7	48.6
B8	630		270		326	10.8	2700	35.6	46.6
B9	540		360		326	10.8	2700	27.8	38.5
B10	810			90	326	10.8	2700	47.8	62.6
B11	720			180	326	10.8	2700	47	60.4
B12	630			270	326	10.8	2700	38.9	52.6
B13	540			360	326	10.8	2700	35.9	50

other. These effects will lead to lower strength and a decreased density of the microstructure of the concrete. For that reason replacing cement with these types of powders is not recommended.

5.2 ***Experimental results M600***

When cement is replaced by particles which are much smaller than cement and/or have a higher characteristic packing density of their own, packing density profiles will be higher. For a fine filler such as M600 the packing density profile is even above $\alpha_{saturation}$ for mixtures containing up to 24% [m^3/m^3] of M600 of the total mixture. This means that when cement is replaced by small amounts of M600, mixtures will have a lower water demand and can be designed with a decreased water cement ratio. In this case small particles fit well between the coarser cement particles, as long as they are properly dispersed with the help of a superplasticizer, Figure 3. The strength tests of these mixtures do not show to have a constant (or increased) reactivity of the binder. However, these tests were performed with a constant water binder ratio, so no advantage was taken of the increased packing density. To show which influence the increased packing density can have on concrete strength, two additional strength tests were performed with a constant water cement ratio. Since the packing density of the cement paste of these mixtures is close

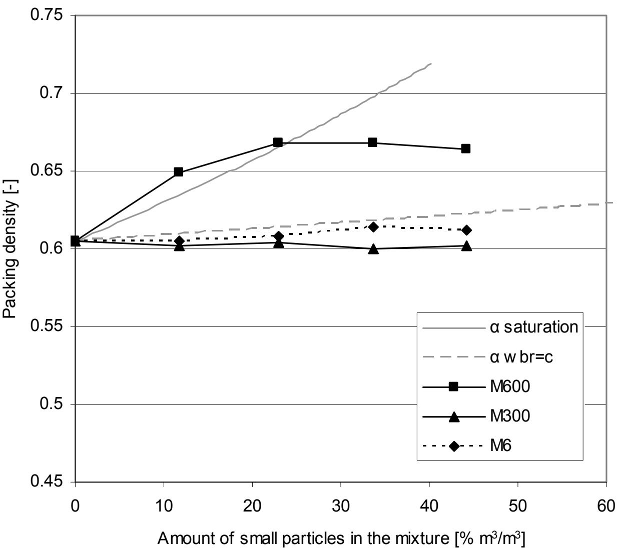

Figure 7: Particle packing profiles of cement combined with quartz powder M6, M300 and M600 as measured by the mixing energy test, in relation to the completely saturated packing density and the packing density for cement-quartz powder mixtures with a constant water binder ratio.

to $\alpha_{saturation} = \alpha_{wcr=c}$ these two mixtures had almost the same workability as the reference mixture B1. Mixture B15 with 20% [kg/kg] M600 in Table 3 shows a 15% strength increase compared to the reference mixture.

In ecological concrete design, replacing cement by M600 will lead to a decreased water binder ratio, an increased binder strength and a dense particle configuration with small particles in the voids between the cement particles. These effects will eventually lead to an increased density of the microstructure of concrete.

Table 3: Mixture compositions and results of strength tests with superplasticizer

Mix	Cement	Quartz powder	Water	Glenium 51	Sand	Compressive strength	
	I 42.5 N	M600	wcr=0.37	1.2%		7 days	28 days
	[g]	[g]	[g]	[g]	[g]	[MPa]	[MPa]
B1	900		326	10.8	2700	50.1	64.5
B14	810	90	293	10.8	2700	54.9	68
B15	720	180	259	10.8	2700	54.6	74.4

5.3 Cyclic design of ecological concrete

In the design method as presented in section 3, fillers are added to cement pastes. As long as cement pastes are regarded, the addition of filler will result in the same mixture composition as the replacement of cement by filler with simultaneously lowering the amount of water in the paste. So both approaches will lead to the same result: a more environmental friendly mixture with a lower cement content. However, for the design of ecological concrete adding fillers to the concrete will lead to a different mixture composition than replacing cement by filler. Since the design cases for adding fillers to pastes as presented in section 3 can also be formulated for concrete, cyclic design with a computer model is recommended. The first step will be to virtually add filler to an existing mixture. The new packing density of the mixture is calculated. If the filler is suitable, the water demand of the mixture can be lowered. This would normally lead to a stronger mixture, but in ecological concrete design this increased strength efficiency is balanced by lowering the amount of cement. Then a second design cycle starts with the new mixture composition including the filler and the reduced amount of cement. A new optimal packing density is calculated by the particle packing model. When the water demand and strength comply with the desired strength class and flowability the cyclic design process is finished.

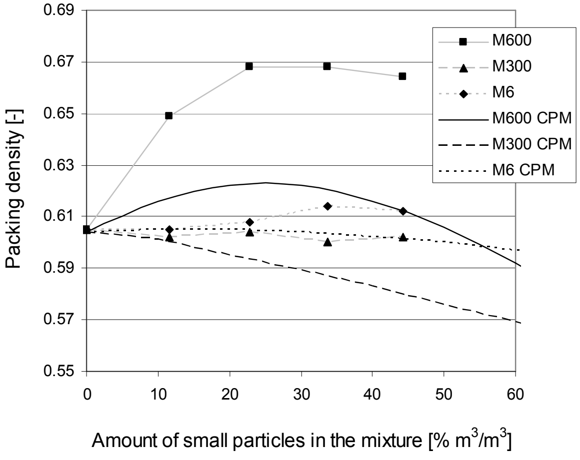

Figure 8: Particle packing profiles of cement combined with quartz powder M6, M300 and M600 as measured by the mixing energy test, in relation to the calculated packing densities by CPM

5.4 *Future work*

The computer model for the cyclic design of ecological concrete comprehends a particle packing model to calculate the maximum packing density of each mixture composition. At this moment, the particle packing model implemented in the design cycle is the compressible packing model (CPM) by de Larrard [4]. However, for very small particles particle packing predictions by CPM deviate considerably from experiments, Figure 8. For M600 the difference between the calculated and the measured packing density can be up to 7%. A deviation in the calculated packing density of this order leads to a predicted minimum water binder ratio of 0.21 in stead of the measured 0.17 for cement with 44% of M600. With small particles, the packing density is influenced by interaction caused by surface forces such as Van der Waals forces, electrical charges and steric forces. To improve the computer model, these interaction forces are to be implemented in the particle packing model.

6 Conclusions

For the design of ecological concrete a cyclic design procedure, based on particle packing and water demand, is presented. The procedure is explained on the basis of cement paste combined with various amounts of quartz powder added as filler.

- Experimental results show that water demand of a cement paste can be lowered by adding small quartz powder particles.

- For packing density profiles above $\alpha_{saturation}$ adding filler can simultaneously lead to a decreased water cement ratio, a decreased amount of cement and possibly to a higher reactivity of the cement due to the packing configuration;
  - replacing 20% [kg/kg] cement by M600 can lead to a 15% strength increase at a constant water cement ratio;
  - even higher strengths can be expected when the increased packing density is used to design mixtures with a decreased water cement ratio;
  - ecological pastes and concretes can be designed when the increased strength efficiency is balanced by lowering the amount of cement.
- For packing density profiles below $\alpha_{saturation}$ adding filler will simultaneously lead to an increased water cement ratio at a decreased amount of cement and probably a lower reactivity of the cement due to the packing configuration;
  - replacing cement by M300 or M6 leads to strength decrease at a constant water binder ratio of 0.5;
  - ecological pastes and concretes can only be designed with fillers which increase the reactivity of cement.
- A cyclic design procedure, including a particle packing model, assesses the suitability of fillers to be included in ecological concrete mixtures on the basis of packing density profiles in relation to $\alpha_{saturation}$ and $\alpha_{wbr=c}$.
- To improve the accuracy of the particle packing model, it should include particle interaction caused by surface forces.
- A recommendation is made to include the reactivity of cement combined with filler, as measured by isothermal conduction calorimetry in the cyclic design procedure for each type of filler.

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List of symbols

r_i	Volume fraction of particle group i. By definition $r_i = \varphi_i \Big/ \sum_{i=1}^{n} \varphi_i$ and $\sum_{i=1}^{n} r_i = 1$.
d_i	Diameter of particle group i. i = 1 for the largest diameter.
φ_i	Partial volume: the volume occupied by size class i in a unit volume.
φ_{mix}	Partial volume of all the particles of a mixture in a unit volume.
α_i	Packing density of particle group i.
α_t	Packing density of a mixture
$\alpha_{saturation}$	Packing density for which a mixture remains to be completely saturated.
$\alpha_{wcr=c}$	Packing density at constant water cement ratio
$\alpha_{wbr=c}$	Packing density at constant water binder ratio
a_{ij}	Parameter which describes the loosening effect caused by the particles in class j on the packing of the particles in class i.
b_{ij}	Parameter which describes the wall effect caused by the particles in class j on the packing of the particles in class i.
V	Volume
ρ	Particle density

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