Measuring the packing density to lower the cement content in concrete

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ABSTRACT: Geometrically based particle packing models can help to predict the water demand of concrete, and thus the material properties. In this paper it is described how centrifugal consolidation can be used to determine the packing density of powders. The method is assessed based on experimental data, calculations and polarization and fluorescence microscopy of the samples. Results show that an average maximum packing density can be measured, which depends on the initial water powder ratio, the use of superplasticizer, the mixing procedure of the paste and the applied compaction energy. Viscosity measurements show the influence of the particle packing density on water demand and how concrete mixtures can be designed to lower the cement content in concrete.

1 INTRODUCTION

Since the revolutionary development of new types of concrete in the last decade, questions arise on how to design tailor made concrete. Therefore, mixture design methods, which can predict the material properties of concrete based on the cement content, become more and more important. Geometrically based particle packing models such as the Compressible Packing Model (Larrard 1999) can help to predict the water demand of concrete, and thus (through water binder ratio and packing structure) the material properties. However, these models do not adopt micro-particles accurately, while exactly these powders have a very large influence on concrete properties. A very important aspect to implement micro-particles into geometrically based particle packing models, is the availability of a simple and accurate method to determine the maximum particle packing density of a powder. The packing density of coarse particles is usually measured dry according to NEN-EN 1097-3. For fine powders, it is important to measure the packing density in the same fluid (water and superplasticizer) as used in the concrete, because of the strong surface forces influencing the packing density. In this paper, it is described how centrifugal consolidation (Miller et al. 1996, Kjeldsen et al. 2006) can be used to determine the packing density of cement pastes and powder mixtures in water combined with superplasticizer. The method is assessed based on experimental data, calculations and polarization and fluorescence microscopy (PFM) of the samples. An investigation on cement pastes shows the influence of the particle packing density on water demand and viscosity and how concrete mixtures can be designed to lower the cement content in tailor made concrete.

2 METHOD

2.1 Centrifugal consolidation of cement paste

The particle packing density of cement pastes was determined by centrifugal consolidation according to the following procedure: A paste, with a known composition, was mixed in a three-liter Hobart mixer. First, the dry powders were mixed for ten seconds after which the water and superplasticizer were added. Mixing was continued for one minute after which the paste rested for one minute. Paste adhering to the wall was scraped from the bowl. After this, mixing was continued for one more minute. Subsequently, the paste was poured into 90 mm long test-tubes with an internal diameter of 22 mm. By determining the mass of the paste in the test-tube, the amounts of powder and water in the test-tube at the beginning of the test were known. The test-tube was then centrifuged for ten minutes at 4000 rounds per minute in a Dumee Jouan E82N centrifuge with an internal diameter of ± 300 mm. By centrifuging the test-tube, the particles in the paste are compacted and less amount of water is necessary to fill the voids in between the compacted particle matrix. Therefore, the total sample will possess an excess amount of water, which will occur as a water layer on top of the (compacted) paste. This water layer can be removed with a pipette, after centrifuging. By determining the amount of removed water, the amount of
water and particles in the compacted sample are known and thus the packing density of the powder can be calculated at the applied compaction energy and under the assumption that the particles absorb no water.

2.2 Mixtures

The cementitious materials used are Portland cement CEM I 32.5 R and CEM I 52.5 R (ENC I Maastricht, $\rho = 3150$ kg/m$^3$), blast furnace slag cement CEM III/B 42.5 N (ENC I Jmuiden, $\rho = 2950$ kg/m$^3$) and fly ash (Vliegasunie, SMZ Maasvlakte, $\rho = 2286$ kg/m$^3$). Particle size distributions are shown in Figure 1. Eleven mixtures were composed with various amounts of cement and fly ash, Table 1. Mixtures 1 to 4 were composed to investigate the influence of the water powder ratio on the packing density. Mixtures 5 to 11 were designed to show how packing density and packing structure vary with different amounts of fly ash. A superplasticizer, Cugla SL01, was added to the pastes with a water cement ratio below 0.5 for the sole purpose of plasticizing the pastes to enable filling the test-tubes. It should be noted; however, that the use of the superplasticizer also influences the resulting particle packing density.

2.3 Polarization and fluorescence microscopy

To obtain a better understanding of the resulting packing structure and the packing density after centrifugal consolidation, polarization and fluorescence microscopy (PFM) was used to characterize the samples. To produce the thin sections, a specimen was first dried at 40°C. Next, this specimen was impregnated with UV-fluorescent resin under a vacuum. After hardening of the resin, the specimen was sawn in the longitudinal direction. Then, the swn surface was polished and glued onto a glass plate. Subsequently, the specimen was sawn parallel to the glass plate, resulting in a 1 mm thick block. This block was ground and polished until it was circa 25 µm thick. Finally, the thin section was covered with a cover glass.

3 RESULTS

3.1 Packing density

The composition of the pastes is presented in Table 1, together with the results of the centrifugal consolidation test. Mixture 2 reaches a higher packing density than Mixture 1, 0.559 compared to 0.515. Furthermore, the water powder ratio of Mixture 1 diminishes more, from 0.5 to 0.299, than the water powder ratio of Mixture 2 (0.350 to 0.250). The differences in the diminution of the water powder ratio during the centrifugal consolidation test in combination with the obtained water powder ratio after the test suggest that the packing structure might change during the test, thus influencing maximum packing density. Note that the packing density of a segregated powder will always be lower than the packing density of a well mixed powder with the same particle size distribution.

Figure 1. Particle size distributions cementitious materials.

Table 1. Mixture compositions in % by mass and packing density measurements.

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Powder Type</th>
<th>Water powder ratio</th>
<th>Cugla SL-01 con 35% %</th>
<th>Average packing density after consolidation</th>
<th>Average water powder ratio after consolidation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CEM I 32.5 R</td>
<td>100</td>
<td>0.500</td>
<td>0.515</td>
<td>0.299</td>
</tr>
<tr>
<td>2</td>
<td>CEM I 32.5 R</td>
<td>100</td>
<td>0.350</td>
<td>1</td>
<td>0.559</td>
</tr>
<tr>
<td>3</td>
<td>CEM III B 42.5 N</td>
<td>100</td>
<td>0.350</td>
<td>1</td>
<td>0.539</td>
</tr>
<tr>
<td>4</td>
<td>CEM I 52.5 R</td>
<td>100</td>
<td>0.420</td>
<td>1</td>
<td>0.489</td>
</tr>
<tr>
<td>5</td>
<td>CEM I 32.5 R</td>
<td>70 Fly ash 30</td>
<td>0.350</td>
<td>1</td>
<td>0.559</td>
</tr>
<tr>
<td>6</td>
<td>CEM I 32.5 R</td>
<td>50 Fly ash 50</td>
<td>0.350</td>
<td>1</td>
<td>0.566</td>
</tr>
<tr>
<td>7</td>
<td>CEM I 32.5 R</td>
<td>30 Fly ash 70</td>
<td>0.350</td>
<td>1</td>
<td>0.579</td>
</tr>
<tr>
<td>8</td>
<td>CEM III B 42.5 N</td>
<td>70</td>
<td>0.350</td>
<td>1</td>
<td>0.555</td>
</tr>
<tr>
<td>9</td>
<td>CEM III B 42.5 N</td>
<td>50</td>
<td>0.350</td>
<td>1</td>
<td>0.563</td>
</tr>
<tr>
<td>10</td>
<td>CEM III B 42.5 N</td>
<td>30</td>
<td>0.350</td>
<td>1</td>
<td>0.573</td>
</tr>
<tr>
<td>11</td>
<td>Fly ash</td>
<td>100</td>
<td>0.350</td>
<td>1</td>
<td>0.590</td>
</tr>
</tbody>
</table>
The packing density of cement combined with fly ash is presented in Figure 2 for Mixtures 1 and 5-11. Combining of CEM III B 42.5 N with fly ash does not result in an increased packing density of the mixture compared to the weighed average packing density of the two basic components. Combining CEM I 32.5 R with fly ash even results in a decreased packing density compared to the weighed average. However, replacing the cement by fly ash in percentages of 30, 50 and 70% will result in a higher absolute packing density of the paste.

3.2 Polarization and fluorescence microscopy
The PFM study especially aimed at evaluating packing structure of each sample, the differences in the packing structure over the height of the sample and the differences in packing structure caused by the starting water powder ratio of the CEM I 32.5 R-paste. For this reason, all thin sections are evaluated at seven specified depths: the top surface, and 1, 2, 5, 10, 25 and 45 mm below the top surface. At these depths, the range of the particle size distribution, the median of the distribution, the largest particle and the water cement ratio are assessed. Furthermore, specific details were recorded like agglomeration, hydration or bad compaction.

Six pastes (Mixture 1, 2, 3, 5, 8 and 11) were chosen for evaluation by combined centrifugal consolidation and PFM research.

The specimen of Mixture 1 shows a quite porous top layer, which structure differs considerably from the deeper parts of the specimen. Directly below this layer, a transition zone exists, which consists of particles ranging from 5–15 µm (Fig. 3), but also contains agglomerates varying from 20 to 60 µm with a maximum size of about 100 µm. Also between 2 and 3.5 mm from the top surface, the specimen is much more porous than the deeper parts of the specimen. At about 5 mm from the top surface, particle size distribution and composition are about the same. In the zone around 25 mm, particles are substantially coarser and the water cement ratio is lower compared to the upper part of the specimen. Only a few particles range from 5 to 15 µm. At 45 mm, particle structure is about the same as at 25 mm depth. However, a few ‘mega particles’ with a maximum particle size of about 130 µm are present, which could not be found at 25 µm, Figure 4. Also the hydration is high and water cement ratio is very low, which is shown by the low UV-fluorescence.

Specimen M2 (Mixture 2) also consists of CEM I 32.5 R; however, it was originally produced with a
Figure 5. M2, 1 mm from top of specimen. Hydration products surrounding some of the particles are observed.

Figure 6. M2, 45 mm from top of specimen. Almost no hydration products are observed.

Figure 7. M5, 6 mm from top surface. Fly ash and cement are well dispersed. Voids with a horizontal orientation are observed.

The specimen of Mixture 3 was originally produced with CEM III/B 42.5 N with a water cement ratio of 0.35. From PFM analysis, it followed that segregation in this specimen is minimal. In the top part, fine particles as well as coarse particles are present with a median size of 25 \( \mu m \) (Fig. 5). The cement matrix is somewhat porous. Hydration products surround the particles. Deeper in the specimen, between 5 and 25 mm, almost no hydration products are visible. At 45 mm (Fig. 6) hardly any hydration took place, which is also confirmed by the absence of calcium hydroxide crystals. Some parts of the specimen are poorly compacted; however, on average compaction is high and not much water is present. The water cement ratio decreases with increasing depth (UV-fluorescent light), which corresponds to the observed hydration of the particles.

The specimen of Mixture 3 was originally produced with CEM III/B 42.5 N with a water cement ratio of 0.35. The specimen contained an average effective water cement ratio of 0.29 after consolidation. From 1 mm down to 25 mm, microstructures are similar. Particles of all compositions and sizes are present at all depths and particle size distribution is constant over this 25 mm. At the top surface, particles are slightly smaller and at 45 mm more large particles are present, but overall segregations is low.

Mixtures 5, 8 and 11 were used to investigate the packing structure of fly ash combined with cement. The specimens showed that the fly ash is always well dispersed and forms no clusters or agglomerates. Also no size segregation over the height of the specimens is visible. In specimen M8, where fly ash is combined with slag cement, no segregation or systematic clustering occurs. In all three specimens voids can be found with a horizontal orientation. Figure 7.

4 DISCUSSION

Two main effects, influencing the average packing density in a specimen, were observed during the evaluation of the thin sections:

- Size segregation of a particle mixture with a wide particle size distribution and a high water cement ratio.
- Varying water cement ratio over the depth of the specimen after consolidation.

4.1 Size segregation

If two particles of the same material and size are exposed to the same force, they will be pushed to the bottom of the sample at the same rate. However, the force working on a large particle is about four times larger than the force working on a particle which is half its size. This means that with a wider particle size
distribution, size segregation is more likely to occur. In CEM I 32.5 R, with particle sizes ranging from 5 µm up to 130 µm, the forces working on the particles vary by a factor of more than 600. If particles can move freely through the cement paste during consolidation, large particles end up at the bottom of the specimen and small particles at the top part of the specimen (M1). However, M2 shows only minimal segregation. This might be caused by the lower initial water cement ratio. If the original cement paste already forms a packed structure, particles will hook to their neighbouring particles thus preventing free movement of the particles and size segregation (particle interlock). The horizontal voids found in specimens M5 and M8 comply with this last theory. A water ‘bubble’ from the original paste can not be released during centrifugal consolidation because of the stiffness of the particle packing above.

4.2 Varying water cement ratio

If cement paste is compacted by vibration, the basic factors influencing compaction are gravitational forces, shear forces, inter-particle forces, mixing energy and vibration energy. Due to the mixing and vibration, shear forces and inter-particle forces are partly neutralized thus enabling the particles to relocate and form a denser packing. Relocation can also occur when particles move due to gravity. If inter-particle forces and/or shear forces are not neutralized, fine particles will stick together and form an agglomerate. Therefore, in this description ‘relocation’ can refer to a particle or to a group of particles forming an agglomerate. During the centrifugal consolidation test, only three forces influencing relocation have to be considered: the rotational force, the shear forces and the inter-particle forces. In this case, variation in the achieved packing density after compaction is mainly influenced by the compaction force, because shear forces and inter-particle forces are fixed by the mixture composition, which is constant over the sample at the beginning of the consolidation test.

The rotational force working on a particle in the test-tube during the consolidation test can be described by formula 1 (Alonso & Finn 1996).

\[
F = m \cdot \alpha = m \cdot \omega^2 R
\]  

\( F \) = force  
\( m \) = mass  
\( \alpha \) = acceleration  
\( \omega \) = rotation speed  
\( R \) = distance from particle to rotation centre

Figure 8. Packing density variation over the height of M2 for the centrifugal force (Fcentrifuge) and for a constant force (Fgravity) at the same compaction energy. C can range from 46 to 200, the worst and best case scenarios are shown.

\[
\sigma = \text{total stress}  
\]

\[
p = \text{water pressure}  
\]

\[
\sigma' = \text{contact stress between the particles}  
\]

\[
\rho_w = \text{volume density of water}  
\]

\[
\rho_n = \text{wet volume density of the powder}  
\]

\[
a = \text{acceleration}  
\]

\[
h = \text{height of the sample}  
\]

Normally, \( a \) would be the gravitational force, which would be constant over the total height of the sample. In the centrifugal consolidation test, \( a \) changes over the height depending on the distance to the rotational centre.

With Formulas 1 and 2, the contact stress between the particles can be calculated. Then Terzaghi’s logarithmic compression law (Verruijt 1994) is applied to calculate the shortening of the sample:

\[
\varepsilon = -\frac{1}{C} \ln \left( \frac{\sigma_i + \Delta \sigma}{\sigma_i} \right)  
\]

\( \varepsilon \) = strain  
\( C \) = compression constant  
\( \sigma_i \) = stress in the sample  
\( \Delta \sigma \) = stress increase over (part of) the sample

Formula 3 is used to calculate the packing density differences over the height of specimen M2. The main results are presented in Figure 8. It shows that the variation in \( F \) over the height of the specimen during centrifuging has no significant influence on the measured average packing density and that the compression constant (material factor) is the main factor influencing the variation of packing density over the height. The calculated variation profiles comply with the investigation on MgO suspensions by Kjeldsen (Kjeldsen et al. 2006).
4.3 Packing density and water demand

The PFM-research and the accompanying calculations show that centrifugal consolidation can be used to determine the average packing density of a powder, under the condition that the water powder ratio of the paste is low to avoid size segregation of the particles. However, it should be noted that this average packing density after consolidation is influenced by the initial water powder ratio, the use of superplasticizer, the mixing procedure of the paste and the applied compaction energy. This means that only packing densities determined according to a standardized test procedure can be compared to each other.

The results from particle packing measurements on powders can be used as input parameters for particle packing models such as Compressible Packing Model by de Larrard (Larrard 1999). By combining cement and fillers in a smart way, packing density of a concrete mixture can be increased. For instance, by replacing cement with fly ash as was shown in Figure 2. The increased packing density can be used to lower the water demand of concrete (Fennis et al. 2007).

Figure 9 shows how the maximum packing density as measured by the centrifugal consolidation test can be linked to water demand of a mixture. Viscosity measurements (viscometer: PAAR Physica MC1, Bingham model) from 15 mixtures of 100% CEM I 32.5 R, 100% CEM I 52.5 R or 50% of both cements with various water cement ratios are presented and plotted against the ratio of solid volume of the cement paste and the maximum packing density determined by centrifugal consolidation. If two mixtures are designed to have the same viscosity (0.36 Pa.s), but one has an increased maximum packing density from 0.55 to 0.60, the water cement ratio can be decreased from 0.40 to 0.34. In this way, particle packing density measurements can be used to lower the water demand and/or cement content of concrete.

5 CONCLUSIONS

From the tests and calculations performed to assess the centrifugal consolidation test, it can be concluded:

- In general, mixtures with a smaller particle size distribution are less likely to segregate, since the mass difference between the particles is smaller.
- Mixtures with a low water content are less likely to segregate, because of particle interlock.
- A high water-powder ratio in a mixture causes segregation, which influences the measured average packing density.
- After centrifugal consolidation the packing density/water powder ratio varies over the height of the sample as was observed in the PFM-research and confirmed by calculations.
- The centrifugal consolidation test can be used to measure an average maximum packing density. A standardized test procedure is recommended because the results depend on the initial water powder ratio, the use of superplasticizer, the mixing procedure of the paste and the applied compaction energy.
- Particle packing density measurements and particle packing models can help decrease the water demand and/or the cement content of concrete at a constant viscosity.

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