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Using particle packing technology for sustainable concrete mixture design

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The annual production of Portland cement, estimated at 3.4 billion tons in 2011, is responsible for about 7% of the total worldwide CO_2-emission. To reduce this environmental impact it is important to use innovative technologies for the design of concrete structures and mixtures. In this paper, it is shown how particle packing technology can be used to reduce the amount of cement in concrete by concrete mixture optimization, resulting in more sustainable concrete. First, three different methods to determine the particle distribution of a mixture are presented; optimization curves, particle packing models and discrete element modelling. The advantage of using analytical particle packing models is presented based on relations between packing density, water demand and strength. Experiments on ecological concrete demonstrate how effective particle packing technology can be used to reduce the cement content in concrete. Three concrete mixtures with low cement content were developed and the compressive strength, tensile strength, modulus of elasticity, shrinkage, creep and electrical resistance was determined. By using particle packing technology in concrete mixture optimization, it is possible to design concrete in which the cement content is reduced by more than 50% and the CO_2-emission of concrete is reduced by 25%.

Key words: Aggregate, cement spacing, concrete, flowability, particle packing, optimization

1 Introduction

Nowadays, sustainability is one of the main focuses of attention in concrete industry. Though concrete is a structural material of which the total environmental impact per cubic meter is limited, compared to similar types of building materials, the CO_2-emission resulting from cement production is large, because of the vast amount of cement and concrete produced yearly. To produce Portland cement, calcination of limestone is required and during this chemical process about 0.5 kg CO_2 per kg of cement is released,

not yet taking into account the CO_2-emission from the fuel burning required for this process. The annual production of Portland cement, estimated at 3.4 billion tons in 2011 [minerals.usgs.gov], is responsible for about 7% of the total worldwide CO_2-emission [Mehta, 2001; ecosmartconcrete.com; pbl.nl]. Taking environmental measures would be good, especially when it is realised the worldwide production of Portland cement has doubled over the past 10 years from an estimated 1.7 billion tons in 2001 to 3.4 billion ton in 2011 [minerals.usgs.gov]. Replacing concrete by other building materials would not solve this environmental issue. Therefore, the solution should be found in reducing the environmental impact of concrete itself. In some applications, where strength of concrete is governing, this can be achieved by using a stronger type of concrete so less material is required [Gorkum, 2010]. For other applications, where strength is not the governing design criterion, concrete mixtures that contain less cement can be used. In all cases it is important to design concrete structures and mixtures in such way that the environmental impact is minimized. Therefore, in the literature review part of this paper it is presented in which ways particle packing technology can be used to support concrete mixture optimization. Furthermore, the paper shows that particle packing can be used to increase the strength of concrete while reducing the amount of cement, leading to considerable CO_2 reduction.

2   Particle optimization methods: a literature review

Particle packing optimization in concrete mixture design covers the selection of the right sizes and amounts of various particles. The particles should be selected to fill up the voids between large particles with smaller particles and so on, in order to increase the particle packing density α_t. The definition of particle packing density α_t is the solid volume of particles in a unit volume. In the history of concrete, the concept of packing of aggregates already received interest in the 19th century [Féret, 1892], but especially the last decades, particle size optimization has gained new interest with the introduction of new types of concrete, such as high performance concrete, self-compacting concrete, fiber reinforced concrete and ecological concrete. Particle optimization methods can be divided into three groups:

•   *Optimization curves*. Groups of particles, with a specific particle size distribution, are combined in such a way that the total particle size distribution of the mixture is closest to an optimum curve. (Subsection 2.1)

- *Particle packing models*. These models are analytical models that calculate the overall packing density of a mixture based on the geometry of the combined particle groups. (Subsection 2.2)
- *Discrete element models*. With numerical models a 'virtual' particle structure from a given particle size distribution is generated. (Subsection 2.3)

2.1 *Optimization curves*

After Féret stated in 1892 that the choice of aggregates influences concrete strength [Féret, 1892], many researchers tried to find the ideal grading curve. In this area the most well-known researcher is Fuller with his famous Fuller curve [Fuller and Thompson, 1907]. Mix design calculations based on his curve are still used today. The Fuller curve is described by Equation 1 with q = 0.5 [Talbot and Richart, 1923]. See also Figure 1. The curve should represented the grading with the greatest density, based on their conclusions that the gradation that gives the greatest density of the aggregates alone may not necessarily give the greatest density when combined with water and cement because of the way the cement particles fit into smaller pores.

Some researchers tried to improve this curve, like [Andreasen and Andersen, 1930]. They proposed the use of an exponent q in the range of 0.33 – 0.50. This adjustment factor q had to be determined experimentally and therefore can differ depending on the characteristics of the particles. With angular coarse particles the ideal curve would be best prescribed with a lower value for q, since more fine particles should be added to fill the voids between the coarse particles [Kumar and Santhanam, 2003].

$$P(d) = \left(\frac{d}{d_{max}} \right)^q \qquad (1)$$

$P(d)$	size cumulative distribution function [-]
d	particle diameter being considered [m]
d_{max}	maximum particle diameter in the mixture [m]
q	parameter (0.33-0.5) which adjusts the curve for fineness or coarseness [-]

In 1980 Funk and Dinger recognized that any real size distribution must have a finite lower size limit d_{min} [Funk and Dinger, 1980]. So, in contrast to the Fuller curve their ideal size distribution not only pays attention to the largest grain but also to the smallest used grain.
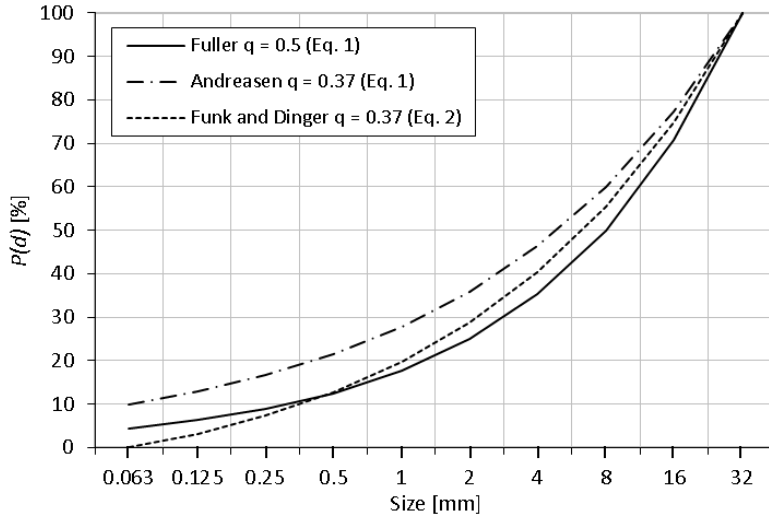
Figure 1: Ideal packing curves according to Fuller, Andreasen and Funk and Dinger for a maximum particle diameter of 32 mm and a minimum particle diameter of 63 μm

As a result, the range of the fine particles is better represented. Andreasen's equation was modified into in Equation 2. They proposed an adjustment factor of q = 0.37 for optimum packing.

$$P(d) = \frac{d^q - d_{min}{}^q}{d_{max}{}^q - d_{min}{}^q}$$

(2)

d_{min} minimum particle diameter in the mixture [m]

These curves (Equation 1 and 2) should lead to the mixture with the highest packing density, by combining optimal amounts of differently sized particles. Packing density is defined as the solid volume of particles in a unit volume, or as one minus the porosity. According to the optimization curves a wider range of the particle size distribution results in a higher packing density, irrespective of the particle shape. However, particle shape greatly influences the packing density, especially, when particles of several size classes, with varying particle characteristics, are used [Walker, 2003; Zheng et al., 1990]. For that reason, Zheng et al. tried to include particle shape by determining q as the average of all q -values for each size class with varying particle shape. Peronius and Sweeting [1985] presented an equation for calculating the porosity of mixtures, depending on the,

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roundness of the particles and the deviation from the Fuller curve. Other researchers, such as [Brouwers and Radix, 2005; Garas and Kurtis, 2008; Hunger, 2010; Kumar and Santhanam, 2003; Vogt and Lagerblad, 2006] used the model by Funk and Dinger to optimize their concrete mixtures by adjusting the q -value based on their experimental results or the required workability.

Adjusting mixture composition to a fixed optimization curve is relatively easy since it requires only a limited amount of input parameters. When the q factor is fixed, only the particle size distributions of the available materials are necessary to optimize a concrete mixture. Commercial computer programs such as *EMMA* [concrete.elkem.com] are available. However, particle characteristics like shape or packing density are not taken into account. The output of the model is an optimized particle size distribution, which not inevitably leads to a mixture with the highest packing density. This is because optimization curves are limited with respect to taking into account differences in particle shape and particle packing of different size groups. Furthermore, Palm and Wolter [2009] and Stroeven et al. [2003] show that the application of gap graded mixtures can lead to higher packing densities.

Optimization curves are continuous particle size distributions based on geometrical considerations. Andreasen and Andersen [1930] started with the assumption of a similarity condition that has to be fulfilled as the particles and their environments increase in size. In this way, a change of the size scale should not affect the packing density of the system. Funk and Dinger [1994] demonstrated that the packing density of continuous particle size distributions (based on this similarity principle) corresponds closely to the packing density of multiple monosized particle groups as described by Furnas [1931]. However, they were not convinced that the packing of discrete (subsection 2.2) and continuous (subsection 2.1) size distributions are actually (mathematically) related [Funk and Dinger, 1994, pp 76]. In 2006 Brouwers demonstrated that the theories on discrete and continuous packings are related mathematically and are actually complementary for packings consisting of multiple particle groups with the same monosized packing density [Brouwers, 2006].

2.2 *Particle packing models*

The purpose of analytical particle packing models is to calculate the theoretical packing density of a mixture. The calculation is based on the particle size distribution and the

packing density of the different particle groups that are present in the mixture. Mathematical equations are used to determine how particles of different sizes interact geometrically. The two basic mathematical equations of almost all particle packing models are the same and purely based on the geometry of the particles, Equations 3 and 4.

$$\alpha_t = \frac{\alpha_1}{1-r_2} = \frac{\alpha_1}{r_1} \qquad \text{large particles (1) are dominant} \tag{3}$$

$$\alpha_t = \frac{1}{r_1 + (r_2/\alpha_2)} \qquad \text{small particles (2) are dominant} \tag{4}$$

α_t calculated packing density of a mixture [-]

α_1 packing density of the large size class 1 [-]

α_2 packing density of the small size class 2 [-]

r_1 volume fraction of size class 1 [-]

r_2 volume fraction of size class 2 [-]. For two size classes, by definition $r_1 + r_2 = 1$

The equations prescribing the packing density were first introduced by Furnas [1929]. They are valid for two monosized groups of particles without interaction between the particles. Either the amount of large particles is dominating the particle structure and small particles fit in their voids, or the amount of small particles is dominating and large particles are embedded in a matrix of small particles.

Since the equations 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.

In 1930, Westman and Hugill developed an algorithm that used discrete theory of packing, which could already be used for multiple particle groups without interaction [Westman and Hugill, 1930]. They acknowledged the existence of geometrical particle interaction, but the model did not yet take into account this interaction between particle groups.

In 1931 Furnas published a method to calculate maximum packing density of multiple particle groups as well as an equation to describe the effect of geometrical interaction between two different size classes on the maximum packing density [Furnas, 1931].

Ben Aïm and Le Goff [1967] implemented this effect of geometrical interaction into the Furnas model, by taking into account the influence of the presence large particles on the packing density of small particles (wall effect). In that same period, Schwanda [1966] published a model which already incorporates both interaction of large particles on small particles (wall effect) and interaction of small particles on large particles (loosening effect) [Reschke, 2000].

The next step in improving particle packing models was the extension from two particle groups to multiple particle groups. Toufar et al. [1976], Stovall et al. [1986] and Yu and Standish [1987] combined the basic equations from Furnas, while Dewar [1999] came up with a stepwise approach where smaller particles are packed in the voids of larger particles and so on.

Nowadays, analytical particle packing models can calculate the packing density of an entire concrete mixture based on the particle size distributions and packing density of the materials used in that mixture . The input parameters are the packing density and particle size distribution of the particle groups, possibly combined with the compaction energy at which the packing density is measured. The output of an analytical packing model is the theoretical packing density of the mixture. For mixture optimization, the packing density of several mixture compositions has to be determined until the maximum packing density is found. Several models are available for this, but each of them differs in how particle interaction such as wall effect, loosening effect and/or compaction energy is implemented in the mathematical equations of the models. Since explaining the differences between the mathematical equations of these models is not in the scope of this paper, reference is made to overviews by [Fennis, 2011; Funk and Dinger, 1994; Goltermann et al., 1997; Johansen and Andersen, 1991; Kumar and Santhanam, 2003]. For readers further interested in using one of the currently available models, reference is made to Europack [Idorn, 1995; Toufar et al., 1976], MixSim [Dewar, 1999; Jones et al., 2002; mixsim.net], *4C-Packing* [dti.dk; Glavind et al., 1999], Compressible Packing Model [Larrard, 1999; *RENE LCPC* or *BETONLABPRO* via lcpc.fr], the Schwanda model [Geisenhanslucke, 2008; Reschke, 2000; Schwanda, 1966], Linear-Mixture Packing Model [Yu and Standish, 1987; 1996] and the Compaction-Interaction Packing Model [Fennis, 2011].

2.3 *Discrete element models*

Discrete element models generate a 'virtual' particle structure from a given particle size distribution. In the earliest models, once a particle was placed, its position would not

change anymore. In these static simulations usually particles are randomly positioned in a defined space, starting with the largest particles. The result is a three dimensional space filled with particles of different sizes, which usually do not have contact with each other. Because of this, the result is officially not a packing or packing structure. Examples of models using static simulations are the hymostruc model [citg.tudelft.nl] or the model used by Zheng and Stroeven [1999] with which e.g. the distribution shown in Figure 2, left hand side, was generated.
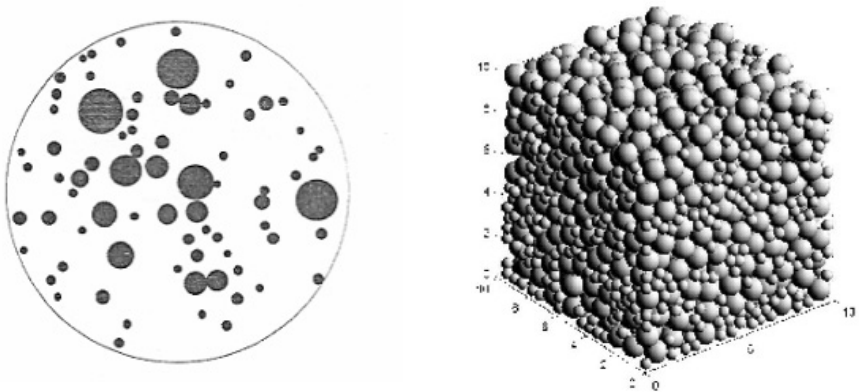
Figure 2: Discrete element models creating a particle structure without particles touching each other (left hand side [Zheng and Stroeven, 1999]) or a stable particle structure (right hand side [Fu and Dekelbab, 2003])

With increasing computational speed, the models evolved to dynamic models in which all particles can move. Particles are generated and subsequently move because of forces acting on the particles. For instance, particles can experience gravity and can collide or remain situated. In this way, the resulting packing corresponds to a random loose packing. A disadvantage of this simulation type is, that the resulting packing structure does not have the highest possible packing density that can be achieved with the given particle size distribution [Fu and Dekelbab, 2003]. To solve this, some models generate particles in a container followed by a stepwise decrease of the container volume [Stroeven and Stroeven, 1999]. Other models allow particles to overlap initially and then rearrange the particles while enlarging the container size until no particles overlap [Kolonko et al., 2008].

The result of each simulation is a virtual particle structure in which the size, the shape and the location of all particles are known, see also Figure 2 right hand side. In this way, the virtual structure can be used to assess random particle packing. Packing density can be

calculated from the total occupied volume in the container; however, the particle structure contains much more information. It can be used to evaluate for instance the resistance to external loads [Snoeijer et al., 2003] or the number of contacts between particles. Also it is possible to simulate flowing concrete such as in a slump flow measurement [Gram and Silfwerbrand, 2007; Roussel et al., 2007].

To find the mixture composition with the highest packing density, several mixture compositions should be simulated, which is very time consuming. Especially with broader particle size distributions, computational time increases with hours, because of the high amount of small particles in the mixture. Some researchers like [Kolonko et al., 2008] solve this problem by making use of a stepwise approach in which small particles are packed and then serve as a matrix between larger particles. However, this leads to an increase of input parameters, which already consist of particle size distribution, container size and/or the amount of particles, but should now include several model parameters such as gravity, density, damping, elasticity, shear, friction and particle contact.

For additional information on discrete element modeling and applications, the authors refer to other papers in this issue of *Heron*.

2.4   *Concluding remarks*

The ideal particle size distribution depends on the particle characteristics and therefore it varies with each type of concrete. For instance, when rounded sand is combined with coarse recycled aggregates, the optimal particle size distribution will differ from one of a mixture with angular sand and rounded coarse aggregates. Therefore, one single ideal optimization curve does not inevitably lead to the mixture with the highest packing density. In concrete mixture design high packing density is important to reduce the water content, especially for ultra-high performance concrete and ecological concrete. Analytical particle packing models and discrete element models can calculate the maximum packing density of concrete mixtures. However, due to limitations in computational speed discrete element models are not ideal for concrete mixture optimization, since numerous mixtures have to be evaluated to find the optimal composition. Consequently, at this moment analytical particle packing models provide the best solution for concrete mixture optimization based on particle packing density.

3   Particle packing and its influence on concrete properties

Optimizing the particle packing density of concrete mixtures has several advantages for concrete properties in the wet as well as in the hardened state. Adding fine particles to a particle structure helps filling up the voids in the particle structure leaving only minimum space for water. In this way adding fine particles will reduce the water requirement [Kronlöf, 1997; Larrard, 1999; Wong and Kwan, 2008]. Increased packing density of the aggregates, improves strength as long as all voids between the aggregates are filled with cement matrix [Reschke, 2000]. A strong aggregate structure with a high packing density will restrain the amount of shrinkage and creep that can actually be realized. Furthermore, a lower water/cement ratio reduces shrinkage, because of the reduced amount of evaporable water in the cement paste [Neville, 1995]. Similarly, Kwan and Mora [2001] reason that a higher packing density leads to a smaller void ratio and thus a smaller amount of cement paste is needed. The heat of hydration and the drying shrinkage are reduced, since both are roughly proportional to the volume of cement paste in the concrete [Kwan and Mora, 2001]. Dhir et al. [2005] demonstrated that changes in the performance of other engineering properties when using particle packing techniques, different cement types and lower water and cement contents, are proportional to the changes in cube compressive strength.

In this section, the advantage of high particle packing density on concrete properties is explained based on the underlying physical relationships. Subsections 3.1 and 3.2 present how particle packing density and particle structure are related to water demand and strength. In subsection 3.3 it is shown how these relations can be used in concrete mixture design.

3.1   *Particle packing density and water demand*

The particle packing density of a concrete mixture has an important influence on its water demand. The definition of particle packing density α_t is the solid volume of particles in a unit volume. However, distinction should be made between the packing density of a stable particle structure and the volume of particles in a real concrete mixture. In a stable particle structure all particles are in contact with each other and packed with certain packing density α_t. In a real concrete mixture, the partial volume of all the particles in a unit volume, φ_{mix} , is lower. This is shown in Figure 3, where the same amount of particles in a stable particle structure is packed closer (Fig. 3, middle) than in a real mixture (Fig. 3, left). In a real concrete mixture part of the water is used to fill the voids between the particles,

while the rest of the water is regarded as excess water. This excess water provides the flowability of the mixture. Flowability increases with a higher excess of water in the mixture. In that case, the solid content of the mixture φ_{mix} decreases.
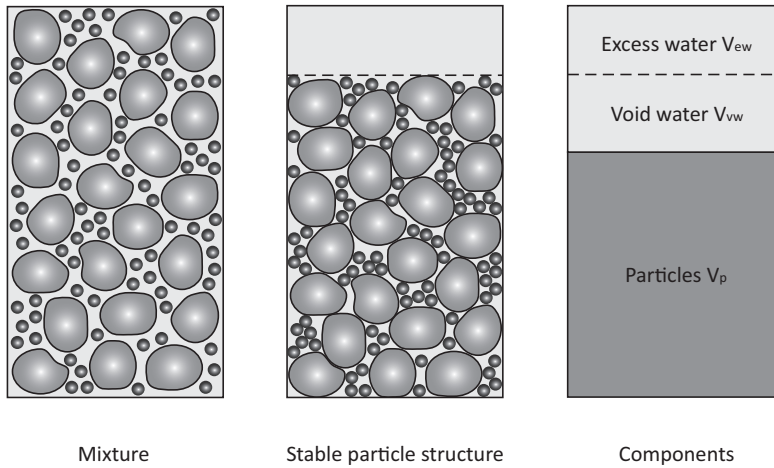

Figure 3: The volume of a flowable mixture compared to the volume occupied by a stable particle structure containing the same particles

If the particle composition of a concrete mixture is optimized in such a way that the maximum particle packing density increases, less void water is necessary. Because of this effect high packing density improves the workability of a mixture. When part of the water that first filled up the voids between the particles, becomes available as excess water, it will provide more flowability. Clearly, the resulting increase in flowability is very useful in the design of concrete mixtures.

In Figure 4 the relation between flow value and packing density for more than 60 tested mortar mixtures is shown [Fennis, 2011]. The results are presented as a function of the ratio between solid volume in the real mixtures φ_{mix} and the maximum calculated packing density α_t.

The relation between packing density and water demand can be used to predict the required amount of water of a mixture. An increased packing density lowers the required amount of void water. Therefore, concrete mixtures with the same workability can be designed with a lower water requirement. In this way, increasing the packing density enables the design of high strength concrete with low water/cement ratio or ecological concrete with a constant water/cement ratio but a lower amount of cement.
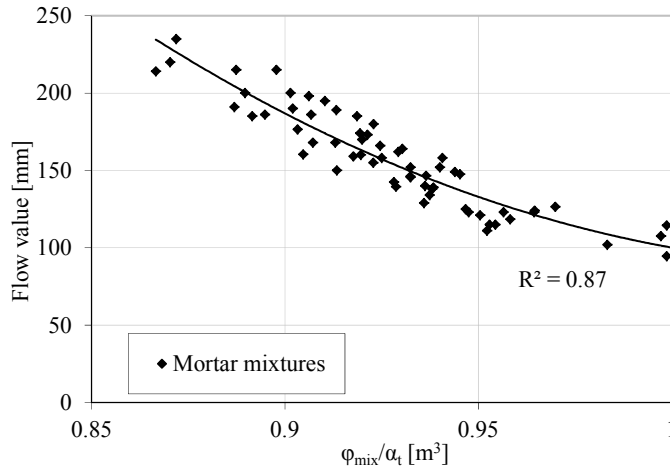
Figure 4: The flow value as a function of φ_{mix}/α_t for mortar mixtures [Fennis, 2011]

3.2   *Packing density and cement spacing*

Optimizing the packing density is not only beneficial for the water demand, but it can also decrease the space between the cement particles. This space between the cement particles depends on the amount of water in a mixture as well as the particle structure of the mixture. With a high packing density in the mixture, cement particles and other particles are close to each other, reducing the space that needs to be filled by hydration products. This leads to high strengths. The other way around, with higher amounts of water and higher water/cement ratios, the cement particles are further apart. Therefore, during the hydration process, the hydration products of the cement particles need to bridge a larger distance, eventually leading to lower strengths.

Traditionally, water/ cement ratio (or a derived parameter) is the basis of most equations for predicting the strength of concrete [Abrams, 1919; Bolomey, 1935; Féret, 1897; Larrard, 1999; Mechling et al., 2007; Mikulić et al., 2008; Neville, 1995; Popovics, 1998; Powers, 1968; Souwerbren, 1998]. However, by using particle packing technology the cement spacing can be better predicted than by using water/cement ratio and volume fractions alone. By making use of the entire particle structure as prescribed by a discrete element model or in this case an analytical particle packing model, it is possible to calculate the space available for cement particles for strength predictions or even hydration simulations. Incorporating the entire particle structure by using particle packing models provides more advanced information for mixture design, because also the size of fillers and aggregates can be taken into account.

The space between the cement particles can be described by the Cement Spacing Factor *CSF* (Equation 5). This Cement Spacing Factor consists of two parts: the space between the particles in a stable particle structure and a volume factor to account for the water in the mixture.

$$CSF = \frac{\varphi_{cem}}{\varphi_{cem}^* \dfrac{\alpha_t}{\varphi_{mix}}} = \frac{\varphi_{cem}}{\varphi_{cem}^*} \frac{\varphi_{mix}}{\alpha_t} \tag{5}$$

CSF	cement spacing factor [-]
φ_{cem}	partial volume occupied by the cement in a stable particle structure [-]
φ_{cem}^*	maximum partial volume that the cement may occupy given the presence of other particles [-]
φ_{mix}	partial volume of all the particles in a mixture in a unit volume [-]
α_t	calculated packing density of a mixture [-]

As a measure of the space between the cement particles in a stable particle structure, the value $\varphi_{cem}/\varphi_{cem}^*$ is used. In this relation φ_{cem} is the volume in the mixture which is occupied by the cement. φ_{cem}^* is the maximum volume that cement may occupy given the presence of the other particles. As such, $\varphi_{cem}/\varphi_{cem}^*$ represents the free space surrounding the cement particles on a volume basis. In principle, more cement particles in a mixture (higher φ_{cem}) will lead to a closer cement spacing. Addition of fine fillers creates higher packing density (leading to a smaller φ_{cem}^*) and thus closer cement spacing.

In a real mixture, not only the voids in the stable particle structure are filled with water, but some excess water is always present to allow flowability, see Figure 3. A stable particle structure is defined as a structure of particles in which all particles are in contact with one or more particles in such a way that the packing structure is stable under the influence of gravity. By adding the water, all particles move away from each other by the factor α_t/φ_{mix}. Therefore adding more water leads to larger distances between the cement particles and thus to lower strength.

Figure 5 shows the relation of Cement Spacing Factor to the compressive strength of 48 mortar mixtures [Fennis, 2011]. In Figure 6, an example is presented of how particle packing density of mixtures can influence the spacing between the cement particles. When cement is replaced by coarse fillers, with a similar size as cement, the spacing between the cement particles increases. This can be explained by considering a container filled by cement particles. If 20% of the cement is replaced by a filler of similar size, the packing

density remains constant. In total the same volume percentage of the container is still occupied by the particles, however, now only 80% of the original cement particles are present in that container. Thus the cement particles are relatively further away from each other (Figure 6 left hand side). However, when cement is replaced by finer particles (for instance M600, see also Section 4) the packing density increases. In that case, the 80% cement particles will fit in a smaller container (Figure 6, right hand side) and therefore the space between the cement particles is smaller than with the coarse filler [Fennis, 2012].
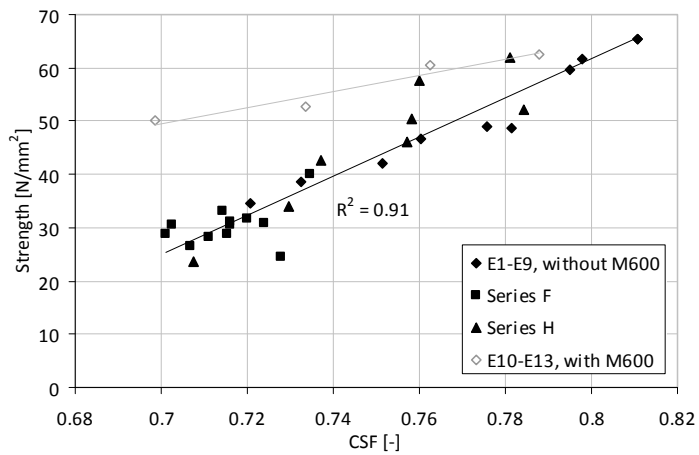
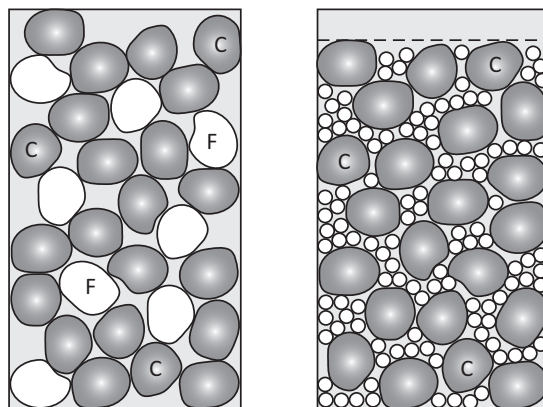*Figure 5: Cube compressive strength of mortars in relation to Cement Spacing Factor CSF [Fennis, 2011]*

Figure 6: The volume occupied by a stable particle structure with coarse filler (F) compared to the volume occupied by a stable particle structure with fine filler (C=Cement)

3.3 **Cyclic design procedure for ecological concrete**

The relations between packing density and water demand, and between particle structure and cement spacing can be used in the design of concrete mixtures. To use these relations in concrete mixture design for ecological concrete, a three step cyclic design procedure has been developed as presented in Figure 7. The design procedure usually starts by calculating the packing density of a new mixture (top corner), but can also start with a mixture adjustment of a reference mixture (bottom, right corner).
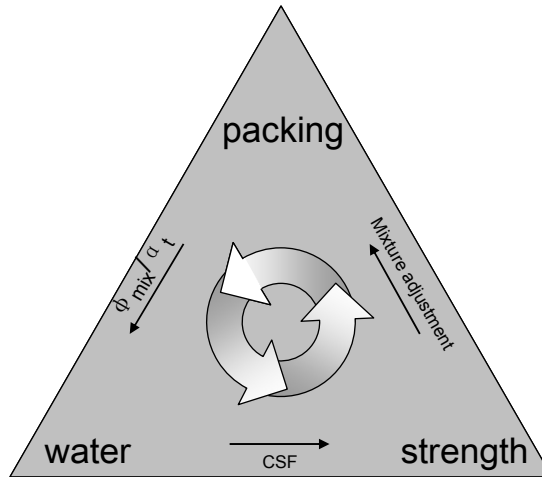

Figure 7: Cyclic design procedure for ecological concrete

For the particle packing optimization (top corner) it is convenient to start from an existing mixture and use an analytical particle packing model such as the compaction-interaction packing model [Fennis, 2011] to find a mixture composition with a higher packing density. Also mixture optimization based on other models (Section 2) could be used as long as the model provides a good calculation of the particle packing density of all the solid constituents in the mixture. The output of the particle packing model in this step will be a mixture composition with its corresponding packing density a_t.

In the next step the water demand of the mixture is determined by using φ_{mix}/a_t, see Subsection 3.1. This value is directly related to the flowability of a mixture, so φ_{mix} and thus the volume of water can be adjusted to the required consistency. When water absorption of the materials and predicted air content are available, the amount of water to be dosed during the mixing process can be adjusted here.

In the following step the strength is predicted on the basis of this mixture composition, its packing density and its particle structure. The strength prediction is based on the

assumption that in low strength ecological concrete all aggregates are stronger than the produced concrete. In that case, the cement glues the aggregates together and the strength depends on the spacings to be bridged to connect all aggregates. With a high packing density in the mixture, cement particles and other particles are close to each other, reducing the space that needs to be filled by hydration products, which leads to higher strengths. With higher amounts of water and higher water/cement ratios, the cement particles are further apart, eventually leading to lower strengths. With this concept, expressed by the Cement Spacing Factor (CSF), the strength of a mixture can be predicted [Fennis, 2011].

In the last step of the cycle, the requirements of the user are taken into account to adjust the mixture composition. In the research project presented in this paper, the requirement is the strength of the concrete. If the predicted strength in the last step is higher than the desired strength, the cement content can be lowered in the next step. Furthermore, additional user defined requirements can be used such as a minimum amount of powders or a fixed proportion of cement to powders. Since the mixture composition is adjusted to the requirements in this step, the particle size distribution might change again.

Cyclic design is necessary as long as one of the material fractions change in the last design step. For instance, when the cement content is lowered due to a high predicted strength value, this changes the overall particle size distribution and thus the packing density. In that case the cyclic design procedure is repeated until the mixture composition does not change anymore in the last design step.

4   Experimental program ecological concrete

As a demonstration, the design method prescribed in Subsection 3.3 is used to design three ecological concrete mixtures. The mixtures were designed for strength class C20/25 with minimum amounts of cement CEM I 42.5 N (ENCI). Three types of cement replacing materials were used: fly ash because of its ball bearing effect and high packing density, quartz powder because of its fine and narrow particle size distribution, and ground incinerator bottom ash as residual product with low environmental impact. The materials used in the mixtures were: Portland cement CEM I 42.5 N (ENCI Maastricht), blast furnace slag cement CEM III/B 42.5 N (ENCI IJmuiden), Fly ash (Vliegasunie), bottom ash (INASHCO), quartz powder M600: d_{50}=3 µm (Sibelco), river sand and coarse aggregates (Filcom, Papendrecht) and Glenium 51 con 35 (BASF).

The mixture compositions of the three ecological mixtures and their reference mixture are presented in Table 1 as series A. The particle size distributions of the mixtures are presented in Figure 8. The reference mixture A1 was designed with 260 kg cement per m^3 concrete, which is the minimum cement content according to the Dutch standards. Since the mixture was designed for strength class C20/25 with design strength 33 N/mm^2, a relatively high amount of water had to be dosed. Mixture A2 and A3 were optimized on packing density. Using the design cycle as presented in Section 2 the cement content was minimized to 110 kg/m^3, in order to reach the estimated design strength of 33 N/mm^2. Mixture A2 contained a combination of fly ash and quartz powder M600. In mixture A3 also blast furnace slag cement was used to increase the packing density even further and create a more ecological mixture containing less Portland clinker. Mixture A4 was designed with wet ground municipal solid waste incinerator bottom ash from INASHCO.
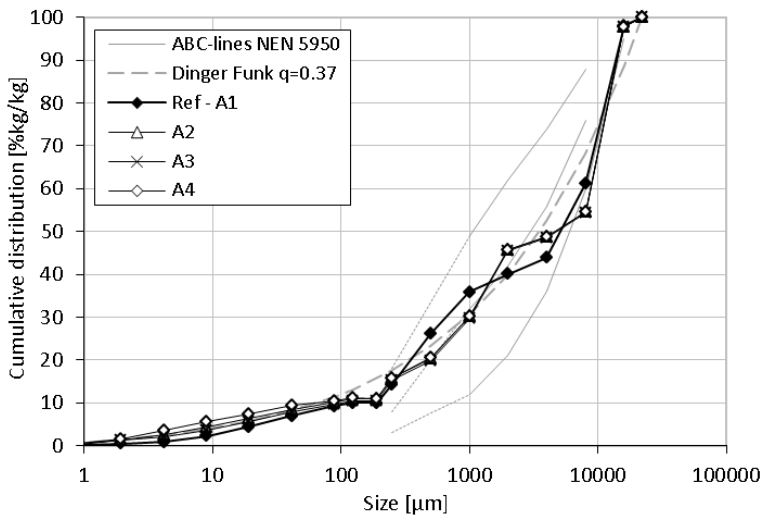

Figure 8: Particle size distribtuions of concrete mixtures series A combined with workability recommendations of [NEN-EN 5950: 1995 nl] and Dinger and Funk optimization curve for q =0.37

To determine the strength development in time, cube compressive strength and tensile splitting strength were measured according to [NEN-EN 12390:2009] on 150 mm cubes after 2, 7, 28, 56 and 90 days. At every point in time three specimens for compressive strength and three specimens for splitting tensile strength were tested. Furthermore, prism compressive strength and modulus of elasticity were determined after 28 days. The experiment was executed by loading three 100×100×400 mm prisms with a constant

deformation speed of 10^{-3} mm/s. The longitudinal displacements were recorded continuously by means of four LVDT's per specimen, which were placed in longitudinal direction in the center of the long sides with 136 mm measuring length.

Drying shrinkage and creep of the ecological mixtures were measured on six 100×100×400 mm concrete prisms in the longitudinal direction over a distance of 200 mm. Shrinkage testing started after 7 days of hardening at 20^0C, 95% RH. Creep measurement started after 28 days of hardening, of which 7 days at 20^0C, 95% RH and 21 days at 20^0C, 50% RH. For the creep measurement a force of 0.33 times the prism compressive strength was applied. The prism compressive strength was determined on specimens of the same size, which hardened under the same conditions as the creep specimens. All shrinkage and creep tests were conducted for 90 days at 20^0C and 50% RH.

The electrical resistance is measured at a frequency of 120 Hz on a saturated 150×150×150 mm concrete cube tightened between two steel plates. Measurements were taken on surface dried, saturated cubes at 20 ± 2^0C and 65 ± 5% relative humidity. Other durability aspects such as chloride penetration and microstructural development were evaluated in a parallel research project on low cement content concrete and will be presented in [Valcke et al., 2013].

5 Experimental results ecological concrete

Evaluation of the fresh state of the ecological concrete mixtures showed that all mixtures in series A were homogeneous and stable. Results for slump [NEN-EN 12350-2:2009], air content [NEN-EN 12350-7:2009] and density [NEN-EN 12350-6:2009] are presented in Table 1. All the mixtures had sufficient workability for casting and vibration. Mixture A3 was judged as harsh compared to the other mixtures, which was especially noted when filling the moulds. To increase the ability to cast in practice, it is recommended to increase the total powder content. This will also improve the cohesion of the mixtures which was evaluated as low for mixtures A3 and A4, leading to easy collapse of the slump. These two mixtures had similar workability as mixture A2, consistency class C1 earth moist. Very low segregation and little bleeding of the mixtures was observed, by virtue of the dense particle structure.

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Table 1: Mixture compositions [kg/m³] and the measured material properties of series A

Composition		Mixtures			
		A1	A2	A3	A4
CEM I 42.5 N	[kg/m³]	260	110	44	125
CEM III/B 42.5	[kg/m³]	-	-	66	-
Fly ash SMZ Maasvlakte	[kg/m³]	-	88	65	75
Quartz powder M600	[kg/m³]	-	62	85	-
Ground IBA	[kg/m³]	-	-	-	50
Aggregates 4-16	[kg/m³]	1193	1162	1160	1157
Sand 0-4	[kg/m³]	718	867	866	864
Glenium 51 [% kg/kg of powders]	[%]	0.8	0.8	1.2	1.2
Effective amount of water	[kg/m³]	162	103	103	112
Water/cement ratio	[-]	0.62	0.94	0.94	0.90
Water/powder ratio	[-]	0.62	0.40	0.40	0.45
Estimated density (1% air)	[kg/m³]	2351	2409	2408	2402
Packing density CIPM	[-]	0.886	0.897	0.898	0.890
Rheological properties					
Slump	[cm]	20	3	*12*	*14*
Air content	[%]	0.9	1.8	0.9	0.9
Density	[kg/m³]	2366	2406	2424	2456
Mechanical properties					
2-day cube compressive strength	[N/mm²]	13.9	15.2	7.2	12.9
7-day cube compressive strength	[N/mm²]	24.2	25.2	17.6	22.9
28-day cube compressive strength	[N/mm²]	32.1	39.6	33.5	37.9
56-day cube compressive strength	[N/mm²]	36.4	39.9	35.3	48.0
90-day cube compressive strength	[N/mm²]	36.4	53.1	39.1	55.1
7-day tensile splitting strength	[N/mm²]	2.0	2.1	1.4	2.0
28-day tensile splitting strength	[N/mm²]	2.5	2.7	2.5	3.0
56-day tensile splitting strength	[N/mm²]	2.6	2.7	3.0	3.3
90-day tensile splitting strength	[N/mm²]	2.7	3.7	2.6	4.0
28-day prism compressive strength	[N/mm²]	29.5	26.6	20.0	29.1
28-day modulus of elasticity	[N/mm²]	30500	32500	30500	30500

The cube compressive strength development of series A is presented in Table 1. All mixtures reach the design strength of 33 N/mm² for strength class C20/25 within 28 days. The contribution of the fly ash and quartz powder to decrease the water demand is substantial. However, neither the conventional water/cement ratio nor the water/binder ratio of mixtures A2 and A3 can explain the strengths attained by these mixtures. Mixture A2 has a water/cement ratio of 0.94 and a water/binder ratio of 0.83 [NEN-EN 206-1:2001]. Mixture A3 has a water/cement ratio of 0.94 and a water/binder ratio of 0.89. Based on the water/binder ratio the 28-day strength of these mixtures should have been between 20 and 25 N/mm². The additional strength gain up to 39.6 N/mm² for mixture A2 and up to 33.5 N/mm² for mixture A3 is explained by the high particle packing density of the powders and possible additional pozzolanic effects of the fly ash and quartz powder both improving the microstructure of the concrete. This is also confirmed by the electrical resistance measurements. Mixture A4 contained ground incinerator bottom ash as cement replacing material and reaches a 28-day cube compressive strength of 37.9 N/mm².

The relationship between the average cube compressive strength and the tensile splitting strength of the ecological mixtures is the same as for normal concrete. Also the measured moduli of elasticity of the ecological mixtures comply with the relation for compressive strength versus modulus of elasticity as described in Eurocode 2 for normal concrete. Only mixture A4 has a relatively low modulus of elasticity compared to its cube compressive strength. The correspondence of the ecological mixtures to the standard relations proves the possibility to design for cube compressive strength in the cyclic design procedure.
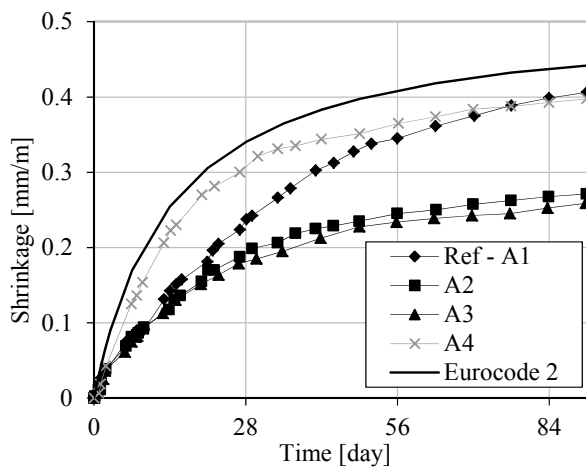

Figure 9: Shrinkage of mixtures series A compared to Eurocode 2

The measured drying shrinkage and creep of the ecological mixtures A2 and A3 were relatively low compared to normal concrete with the same compressive strength. The results can be explained by the relatively low cement content, the normal modulus of elasticity and the high density of the particle structure, which provides resistance against deformation of the cement matrix. In Figures 9 and 10 the shrinkage and creep are compared with corresponding formulations in Eurocode 2 [NEN-EN 1992-1-1:2005] for concrete in strength class C20/25. Mixture A4 has a relatively higher shrinkage than mixtures A2 and A3. In the first 77 days this shrinkage is even larger than the shrinkage of the reference mixture A1. The shrinkage of both reference mixture A1 and mixture A4 is still lower than the shrinkage estimation according to Eurocode 2. The creep of reference mixture A1 is larger than was expected based on its strength class and Eurocode 2.

The variation in the shrinkage and creep measurements of the different mixtures shows that shrinkage and creep very much depend on the types of fillers and binders which are used in the ecological concrete. When for a certain application shrinkage or creep is the most important mixture design criterion, performance-based design will lead to optimal ecological concrete.
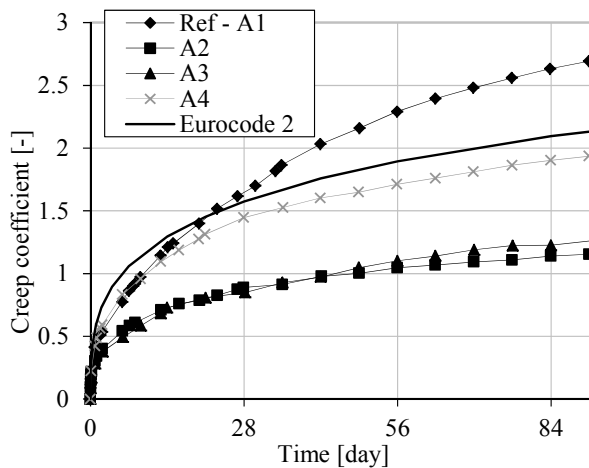

Figure 10: Creep of mixtures series A compared to Eurocode 2

An indication of the durability of ecological concrete can be provided by measuring the permeability of concrete, since concrete with lower permeability is less sensitive for transport mechanisms such as chloride ingress. The two electrode method (TEM) is used to

determine the electrical resistance of concrete and provides an indirect measurement of the amount of uninterrupted water in a fully saturated specimen. This amount of water is related to the quantity of continuous pores in the concrete and thus also to the permeability of the concrete. From theoretical and experimental work there appears to be a relationship between resistivity of and chloride diffusion in a particular concrete composition [Andrade et al., 1993; Polder, 1995; Polder, 2000]. Furthermore, Smith [2006] reports that electrical resistance measurements are related to chloride penetration resistance as measured by the rapid chloride permeability test.

The resistivity of the three ecological mixtures is higher than the resistivity of reference mixture A1, Figure 11, which is positive to prevent ion ingress. Mixtures containing blast furnace slag cement often show high resistivities, caused by differences in microstructure and pore solution conductivity. This explains the differences in the resistivity of A2 compared to A3. The resistivity was not related to the water/cement ratio or water/binder ratio. In fact, mixture A2 with a water/cement ratio of 0.94 and a water/binder ratio of 0.83 has a much better resistivity than A1 with a water/cement ratio of 0.62.

The electrical resistance measurements show that predicting concrete properties related to durability by using the water/cement ratio, the water/binder ratio and/or the cement content is not reliable. To evaluate the durability, the total composition of the mixture specified by the particle size distribution, water content, cement content and water/powder ratio, should be taken into account. For durability aspects, extensive research on performance-based design is recommended.
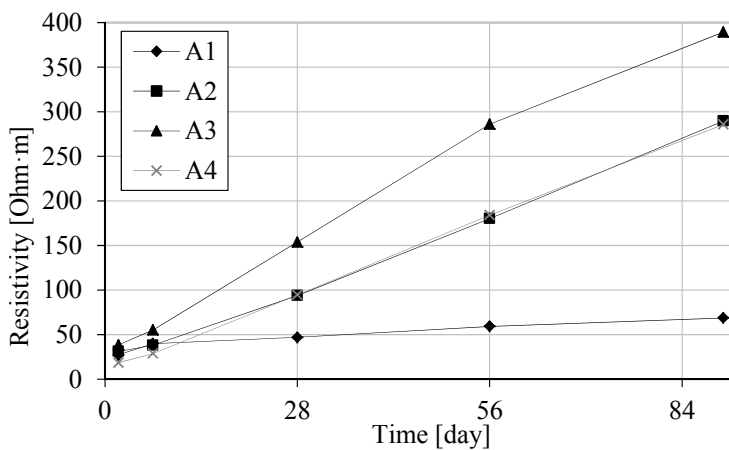

Figure 11: Results of resistivity measurements (TEM) of series A

6   Conclusions

The packing density of a particle mixture by itself is a powerful parameter in the design of concrete mixtures. It helps to determine the suitability of aggregates and fillers for the use in concrete mixtures. In fact, particles should contribute to a higher packing density and to a reduced water demand. Whether adding particles to a concrete mixture indeed has this desired effect, can be determined by particle packing models. In this paper three different methods to determine the particle distribution of a mixture are presented; optimization curves, particle packing models and discrete element modelling.

Higher packing densities leave less space for voids to be filled with water, which reduces the water demand and increases the strength of concrete mixtures. Via the packing structure, the amount of water in a real mixture and the Cement Spacing Factor CSF it is possible to predict the strength of concrete. These relations can be used in a cyclic design procedure to optimize mixture compositions of, for instance, ecological concrete.

Based on experiments on ecological concrete it was demonstrated how particle packing technology can be effective to lower cement contents, without changing concrete properties in a negative way. Three ecological concrete mixtures were designed containing fly ash, quartz powder and ground incinerator bottom ash, thereby saving up to 57% of Portland cement and reducing the CO_2-emission of concrete with 25% [Fennis, 2011]. By making use of the cyclic design method all ecological mixtures reached at least their predicted design strength of 33 N/mm². The mixtures were tested on compressive strength, tensile strength, modulus of elasticity, shrinkage, creep and electrical resistance. The results confirmed that relationships between cube compressive strength, tensile splitting strength and modulus of elasticity correspond to those for normal concrete. Furthermore, the particle packing optimization resulted in a stiff and strong particle structure, which had a positive influence on concrete properties such as shrinkage and creep. However, concrete properties such as shrinkage, creep and electrical resistance can significantly be influenced by the mineralogical composition of the filler and binders. The electrical resistance tests on these ecological mixtures showed that the increased packing density and reduced water demand eventually lead to a denser microstructure of the concrete. The experimental program showed that it is possible to design ecological concrete in which 50% of the cement is saved by using particle packing technology in concrete mixture optimization.

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