Evaluation by discrete element method (DEM) of gap-graded packing potentialities for green concrete design

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ABSTRACT: Partial replacement of Portland cement by pozzolanic mineral admixtures exerts direct positive effects on CO_2 emissions. The green character is reinforced by making use of incinerated vegetable waste, such as rice husk ash (RHA). Gap-grading leads to improved particle packing density with RHA as the fine component, so that high strength concrete can be produced. Characteristics of the capillary pores developed in the hydrating binder have impact on the transport-based durability properties. Yet, their assessment constitutes a complicated problem, especially in experimental approaches. This paper, therefore, presents a new economic and reliable approach to conduct such investigation by DEM. Application demonstrates the positive effects of gap-graded blending.

1 INTRODUCTION

Portland cement (PC) production contributes by about 6% to global emissions of CO2. One of the obvious contributions to reducing detrimental effects of Portland cement production on global warming as a result of CO_2 emissions is to reduce PC content significantly (Stroeven *et al.*, 2002). Use of an admixture of vegetable origin such as RHA will additionally contribute to waste management and energy conservation (Stroeven *et al.*, 1995). Experimental research with Vietnamese participation performed at Delft University of Technology (DUT) during the last decades of the previous century also exploited the gap-grading particle packing principle successfully in designing aggregate (very fine sand and coarse aggregate) as well as binders (Portland cement (PC) blended by rice husk ash, diatomite earth or metakaolin) (Bui, 2001; Vu, 2002). Detwiler and Mehta (1989) and Goldman and Bentur (1993) showed PC binders blended with an inert admixture also to lead to proper strength levels provided the blend was gap-graded, revealing the crucial importance of particle packing. A new series of Vietnamese PhD students advanced this topic at DUT (Le and Stroeven, 2012; Nguyen, 2011a, b).

Particle packing is a problem receiving major attention in physics and mathematics. Recog-nition that it has impact on concrete is also going back to the beginning of previous century. This may be exemplified by an old simple compression test on cement paste specimens. Test-loaded specimens were after crushing and completely grinding down recovered upon compression of the particulate material in a mold. Test-loading demonstrated compressive strength level recovered. This is the particle packing effect in *optima forma*; strength is due to van der Waals forces. Nowadays, separate gap-graded grain fractions are used in the design of Super High Performance Concretes and of Engineered Cementitious Composites.

Experimental research is time-consuming, labor-intensive and thus expensive. As a result, doing research on virtual material is also gaining in popularity in concrete technology. The Discrete Element Method (DEM) definitely offers the most reliable approach, despite various random-generator-based systems in vogue in the concrete technology field (Stroeven *et al.*, 2009). DEM incorporates particle interference, a phenomenon characteristic for the dense randomly packed aggregate and binder particles in the High Performance Concrete (HPC) range. At DUT, we have executed studies with Chinese participation on meso - as well as micro-level of the virtual material using the DEM systems SPACE and HADES

(Chen *et al.*, 2006; He, 2010; Hu, 2004). The favorable effect of gap-grading on strength found in the afore-mentioned Vietnamese-Dutch experimental research was confirmed (Bui *et al.*, 2005).

Recent DEM studies of (blended) PC have focused on the pore network structure as affected by gap-grading. The interpretation of the first test series with SPACE seemed conforming to the concept of Vogel and Roth (2001) for soil materials (Chen *et al.*, 2006). For concrete, this would imply continuous pores around aggregate grains that could mutually connect in (partly) percolated Interfacial Transition Zones (ITZs). The inkbottle effects that are hypothesized for pore structures in concrete could be associated with incomplete connections due to small ITZ overlap. The most recent ongoing investigations by HADES (Stroeven *et al.*, 2012a,b) also reveal the pore trees particularly concentrated in ITZs, however with branching pores that form connections outside ITZs in the so called bulk regions that form the major part of the ma-tured paste in concrete. So, the peak value in the connected pore fraction near the aggregate grain surfaces was concluded not dominating the global value of connected porosity of con-crete. Hence, the degree of ITZ percolation cannot be expected to dominantly govern chloride diffusion (Maghsoodi and Ramezanianpour, 2012a,b). The favorable effects on the pore net-work structure by gap-grading the blend were convincingly revealed (Stroeven *et al.*, 2012a,b) and will be outlined herein. This will have consequences for designing "green" concretes.

2 GAP-GRADED BLENDING EFFECTS

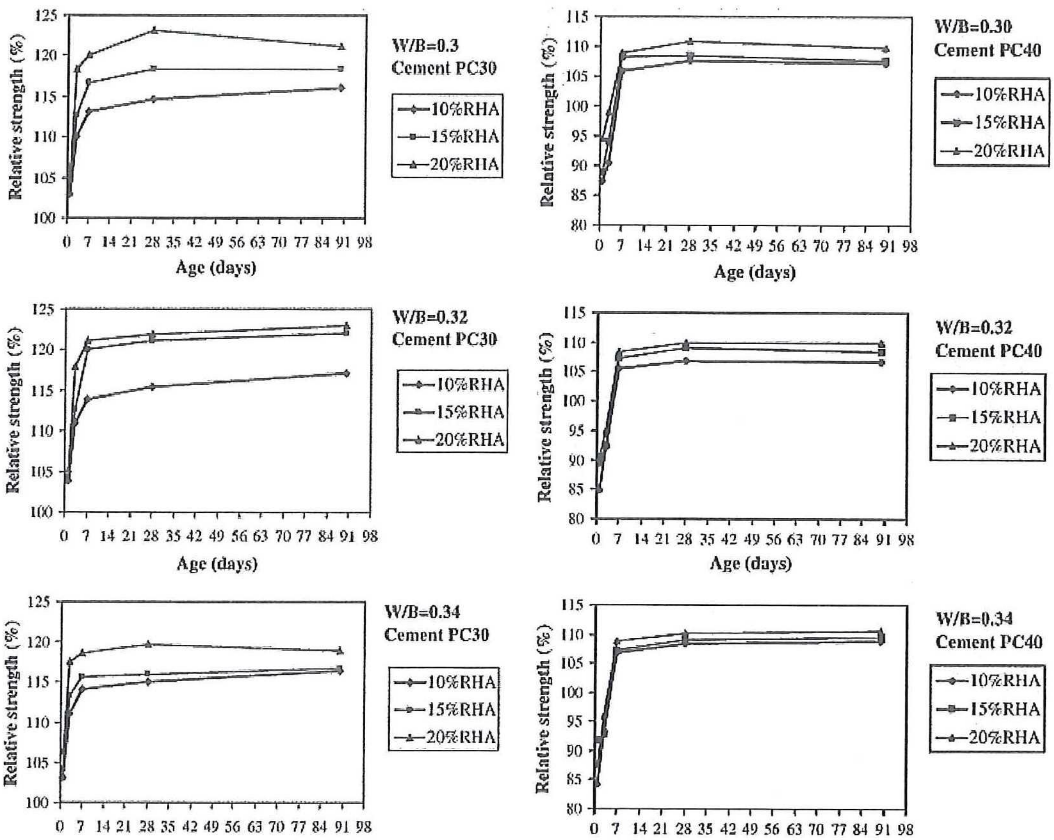


Figure 1. Relative compressive strength values of gap-graded aggregate RHA-blended concrete

The RHA is produced in the traditional way from Vietnamese rice husks and grinded until its internal po-rous structure collapsed, significantly reducing water demand (Bui, 2001). Mean particle size was 5μm. For additional details, see Bui (2001) and Bui *et al.* (2005). 70% crushed basalt and 30% of fine sand of fluvial origin constituted a gap-graded aggregate mixture. 500 to 550 kg/m^3 Portland cement of two quali-ties were used. Three water/binder ratios (w/b) were investigated and replacement percentages of 10, 20 and 30 were envisaged. Naphthalene-based superplasticizer additions were used to get cohesive mixtures with high slump values. 100mm cubes were used for compressive strength testing at different stages of maturation. Detailed test results have been published in the aforementioned publications. However, Fig. 2 reveals the blending efficiency resulting from gap-grading that is only realized when RHA is combined with the coarser cement (PC30). Since strength is improved due to increase in PC quality, the results in Fig. 1 are presented in relative terms.

Fig. 2 presents computer simulation data, revealing gradient structures of λ^{-3} values, whereby λ stands for the mean free spacing. This is supposedly proportional to physical (van der Waals) strength. The normalized values for the *coarsest* cement demonstrate the far more efficient packing in the gap-graded case for 10% cement replacement. At higher dosage this effect of optimized packing is absent, however. Nevertheless, green concrete could be produced with high dosage of fine-grained RHA without strength loss. Moreover, energy is produced during RHA incineration; another "green element" involved in the blending concept. The gap-grading effect is not depending on pozzolanity. Goldman and Bentur (1993) demonstrated that even blending with carbon black (inert!) can give rise to proper results since the physical strength contributions can compensate for the loss of chemical strength. Also in our research we found blending by incinerated diatomite earth not to underscore meta-kaolin-blended mixtures due to higher fineness and thus better packing of the cement grains (Stroeven and Vu, 1998).

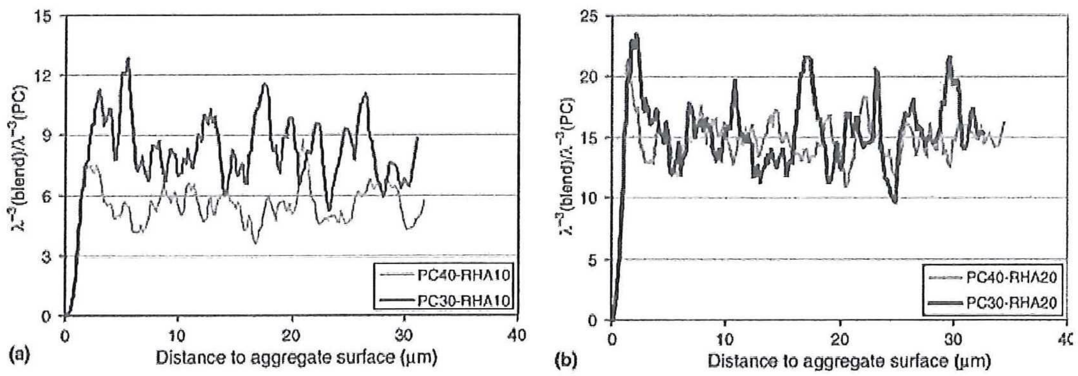


Figure 2. RHA blending reveals increased density in virtual concrete when gap-graded with the coarser PC. λ^{-3} is supposedly proportional to global van der Waals bond, whereby λ is the mean free spacing.

3 PRODUCTION OF CEMENTITIOUS MATERIALS

3.1 Simulation of fresh cement particles by HADES

To obtain matured virtual cement paste, firstly, fresh cement particles need to be generated. In this research packing of fresh cement particles is simulated by HADES (HAbanera's Discrete Element Simulator). HADES is an advanced dynamic force-based DEM system for making realistic packing simulations of arbitrarily shaped particles. This could be the aggregate on meso-level or the binder on micro-level.

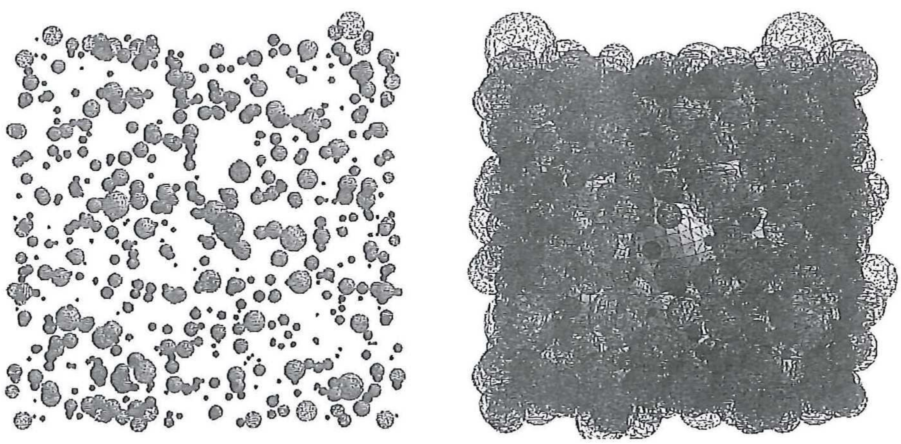


Figure 3. Spherical particles dynamically compacted from loose (left) into dense random state (right) by the DEM system HADES

Mechanical interaction in HADES is based on a contact mechanism algorithm that evaluates the interaction forces exerted between segments of tessellated surfaces of neighboring parti-cles. The contact forces are functions of distances and of areas of the segments. Several forces can be applied in this way on a particle such as spring force, cohesion force, damping force and friction force. HADES renders possible implementing particle packing in containers with periodic boundaries, simulating an infinite space, with rigid boundaries, simulating aggregate's surfaces, or with mixed conditions. Gradual reduction of container size while particles move makes it possible achieving higher packing densities as met in practice. This is illustrated Fig. 3.

3.2 *Simulation of hydration process*

In this research, a new numerical multi-phase model for simulating hydration of (pozzolanic blended) cement is utilized. Herein, the hydrating grains are simulated by spherical integrated particles based on the so called 'integrated particle kinetics model' (IKPM), coupling a fresh core of material and its hydration product (CSH) as a shell coating this core (Le and Stroeven, 2012; Stroeven and Stroeven, 1997). Nonetheless, different from IKPM model that is used for only single phase material (C_3S), each fresh spherical core also incorporates information of its components, i.e. percentages of phases in this model. So, the model is referred to as 'extended integrated particle kinetics model' (XIPKM).

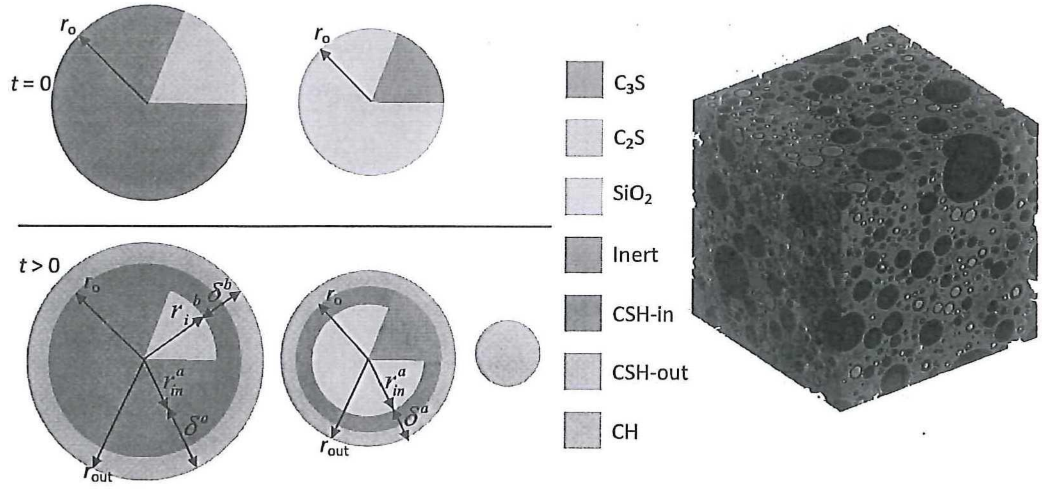
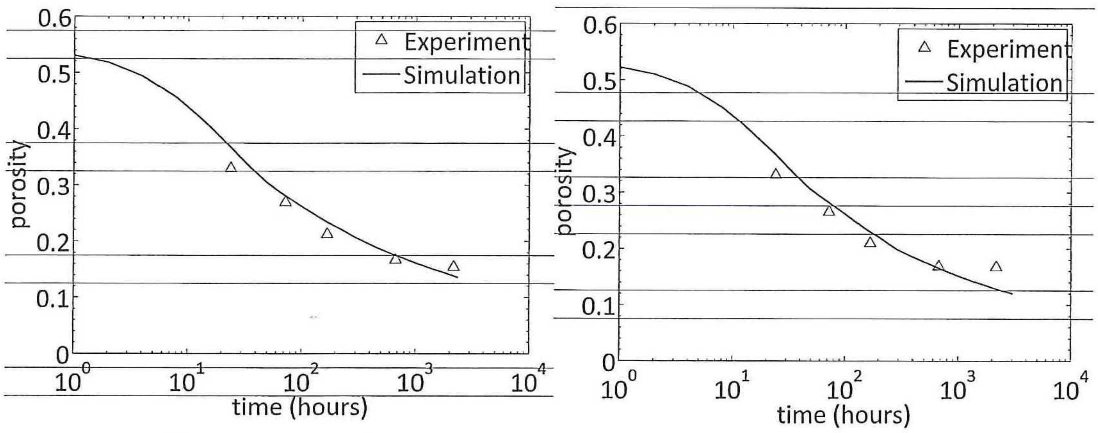


Figure 4. Particle models of cement, pozzolanic admixture and hydration product (at the left) and visualized microstructure (at the right)



(a) 10% cement replacement

(b) 20% cement replacement

Figure 5. Porosity of the RHA-blended PC samples (the experimental results are derived from testing of Nguyen (2011b) on RHA-blended PC samples with $w/b = 0.4$ and 10-20% replacement).

The present model takes into account the two major phases of PC, i.e., tri-calcium silicate (C_3S) and di-calcium silicate (C_2S). Yet, it could be expanded to cover more phase components. Fig 4 is an illustration of the hydration model with three types of particles. Beside the two compo-site/integrated PC particles (left and middle ones in Fig. 4), another hydration product (CH) is modeled as single spherical particles (right one in Fig. 4). Researches have demonstrated that the CH product diffuses and nucleates randomly either in the pore space or precipitates on the surface of the existing CH grains. The quality of the new hydration concept is verified on the basis of experimental data in Fig. 5. Much better correspondence is found than can be expected from popular random sequential systems, as argued by Williams and Philipse (2003).

4 POROSIMETRY BY DRAMUTS

The DraMuTS method has been introduced in Stroeven *et al.* (2012a,b). Basically, nodes are distributed at random or seeded at selective places in the virtual material. A path planning algorithm is designed so that the nodes are connected by straight lines resulting in the formation of a "tree". This can be achieved starting from multiple sources, leading to multiple tree structuring. When a straight line between neighboring nodes is obstructed, a more nearby point is selected preventing iterations. Trees can ultimately merge when similar nodes are involved. The result is in general a pore structure delineated by continuous zigzag lines inside the pores. This renders possible studying (dis)continuity in the pore system, so that pores connecting external surfaces of the simulated cube specimens (trunks) can be distinguished from dead-end pores connected to such trunks (the branches) and from isolated pores. The number of trees is a reflection of pore fractionation: it presents the number of transport routes through the specimen. The expansion of the trees by DraMuTS is illustrated by Fig. 6.

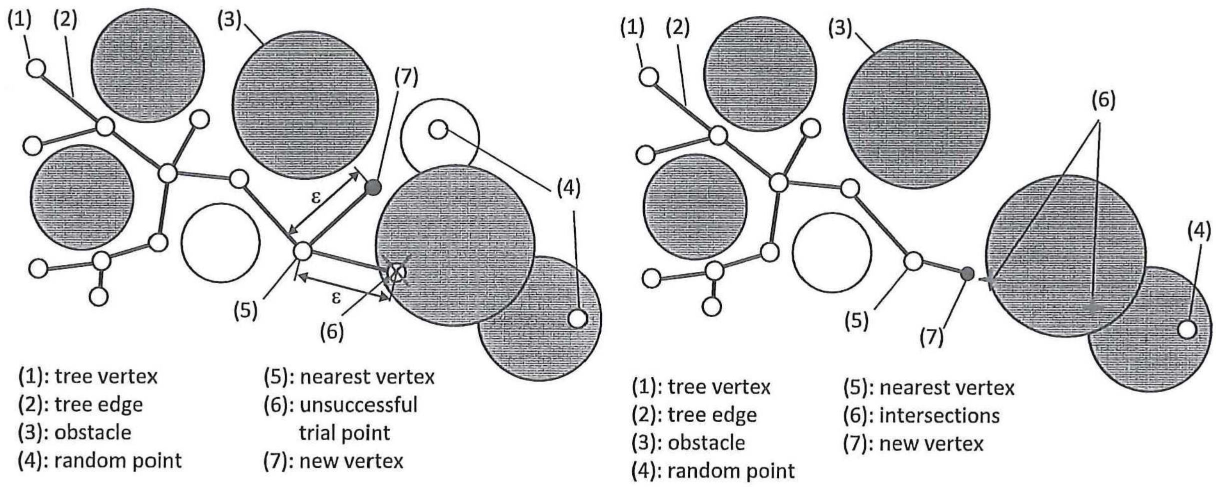


Figure 6. Path finding in robotics (RRT), at the left, and in DraMuTS, at the right.
Note the different locations of (7) after failure in (6).

A second uniform random point system is thereupon generated (note "Double Random" in the name of the method). The node fraction inside the pores directly governs porosity because point fraction is an unbiased estimator of volume fraction. All points inside the pores are thereupon provided with pikes in systematically arranged orientations. The pikes connect the relevant node with the pore surface in the given direction (length l_i). By averaging all cubed values of pike length in a point and thereupon taking the 3rd root, a measure is obtained for local pore radius. This is the so called star volume method that can also be instrumental for obtaining this 3D information from 2D sections (Stroeven *et al.*, 2012a,b) combining all these local pore size measures, a volume-based pore size distribution function (PoSD) is straightforwardly obtained.

5 EXAMPLES

Details of virtual blending experiments are plotted in Table 1. The delineated pore network structures at high and reduced sensitivity are displayed in Fig. 7. The size of relevant pores has to be assessed in the stage of durability estimation. This governs the required sensitivity, because a larger number of dispersed sampling points lead to increased fineness of the pores that will be detected. Fig. 8 reveals the influence of blending with a fine-grained mineral admixture as RHA: the extension of the ITZ is reduced, and its internal porosity refined.

Table 1 Hydration simulation input

Sample	W40Pc	W40Po20	W25Pc	W25Po20
w/b	0.40	0.40	0.25	0.25
% Replacement	0	20	0	20
Number of model cement particles	2091	1601	2644	2002
Number of model RHA particles	-	3457	-	4323

PC: CEM I (PC30) (van Eijk, R.J. (2001)); Blaine: 286 (m^2/kg)
Cement composition: 66 % C_3S, 16 % C_2S, 7 % C_3A and 11% C_4AF
RHA: $RHA_{(18+)}$ (Bui, 2001); BET: $58m^2/g$
Model PC: Rosin Rammler PSD with n = 1.052, b = 0.040 and size range of 3~40 (μm)
Model RHA: PSD function of $RHA_{(18+)}$ (Bui, 2001) and size range of 3~13 (μm)

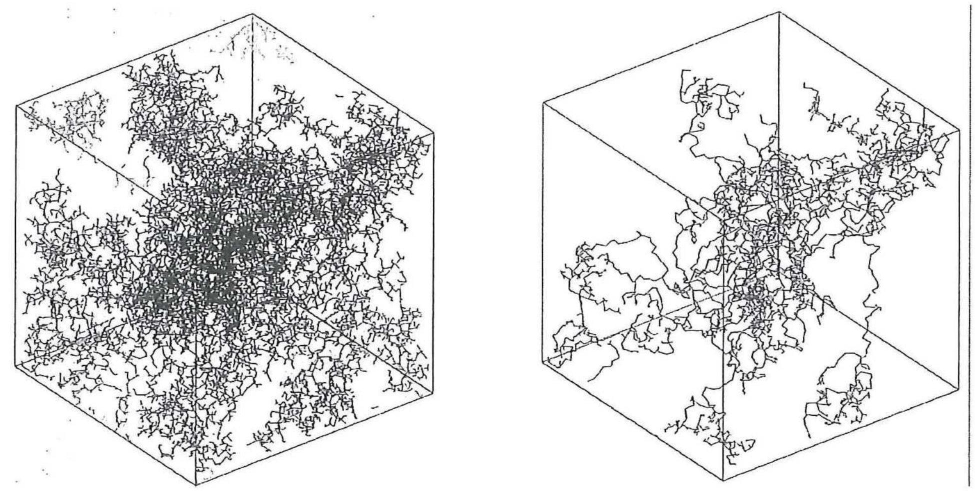


Figure 7. Pore delineation in 100μm cubes for gap-graded blended-PC. All capillary pores are shown (left) as well as the continuous trunks only (right). Visually similar results were obtained for plain PC

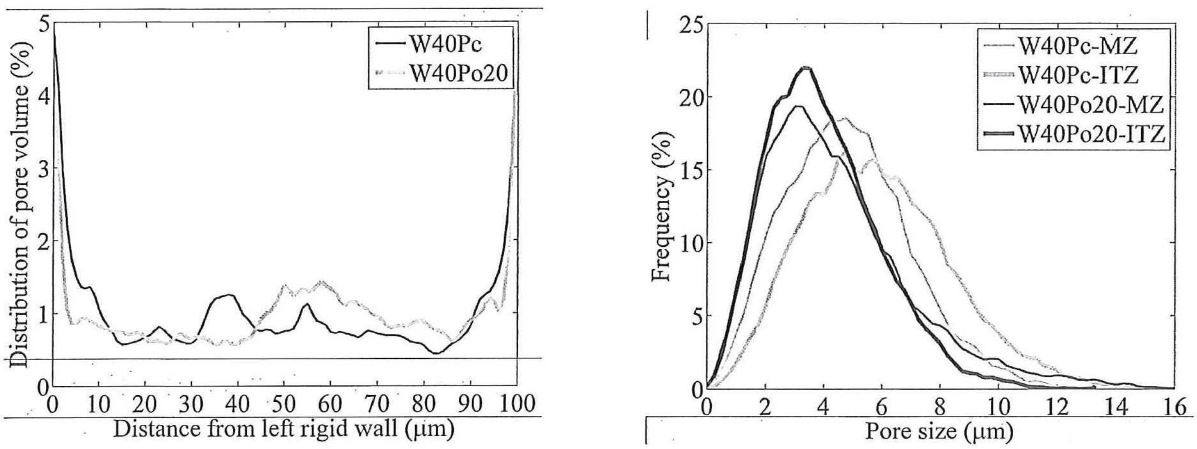


Figure 8. Effect of blending on ITZ extension (left) and on volume-based pore size distribution in and outside the ITZ zone (= Middle Zone).

Whereas the pores in the plain PC are largest in the ITZ, this tendency is reversed for the blended PC! This will have favorable impact on transport-based durability issues. The reader is referred to Stroeven et al (2012a) for another example on blending effects.

6 CONCLUSIONS

The approach by HADES and DraMuTS introduced herein constitutes an economic and reliable way of investigating structural problems. Particularly, topology and geometry of the complex and tortuous pore network structure that are at the basis of transport-based durability properties are readily obtained. For that purpose, the fresh simulated particle structure is hydrated by XIPKM, which is able considering the two major compounds of the PC as well as particles of a blending mineral admixture. Application to RHA-blended PC has demonstrated the favorable effects on packing density, which is underlying strength, and on pore refinement in an ITZ of reduced extension. This will have positive impact on durability.

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