Development of Eco-concretes

H.J.H. Brouwers

Eindhoven University of Technology, Eindhoven, The Netherlands

Introduction

The building sector, comprising both buildings and infrastructure, is the largest consumer of energy and materials [1, 2]. Table 1 shows the global production of the most important man-made materials in 2008.

Table 1: Global production of materials in million tons (2011).

Timber	4000	Quicklime	130
Plastics and rubber	300	Glass	120
Steel	1400	Cement	3500
Gypsum	250	Concrete	21000

Some of these materials are exclusively used in the building sector; others such as steel and timber are also used in other industries. The numbers illustrate that building materials are globally of the utmost importance, both economically and environmentally, and their production may involve contamination and the depletion of finite resources. Contamination concerns emissions to water, air and soil, and the production of waste, stench, noise, and other forms of nuisances. This takes place during production, but also during the entire life-cycle (and beyond) of a material and object. In contemporary societies the aspect of 'contamination' has largely been solved in the past 40 years. The challenge we are now facing is a more efficient use of resources.

Many of the building materials contain irregularly shaped particles, of various sizes (polydisperse), mostly randomly packed as is the case in concrete. The particles need to be combined in a specific way to obtain the desired properties of the building material. In other words, there is a need for recipes according to which the raw materials are combined and processed into the desired materials or products. These products not only have to fulfill a broad range of technical demands, but must also meet requirements with respect to maintenance, repair, recycling, sensory qualities etc. The raw materials and ingredients need to be available in sufficient quantities of constant quality, and the product should be manageable at the building site. A material that often fulfills these requirements is concrete.

Concrete is a spectacular material, not only because of its enormous production volume. A conventional concrete consists of cement,

aggregates and water, and comprises grains with a size ranging from 300 nm to 32 mm, in massive constructions (e.g. dams) even to 64 or 128 mm. There is no man-made material in which such a broad range of particle sizes, comprising five decades, are combined. In fresh state it is fluid and enables a high degree of design freedom, while in the hardened state concrete is durable and almost maintenance-free, with a great architectural potential.

It has been seen that the cement industry is reducing the environmental impact of cement, for example by fuel and material substitution. The substitution of clinker is regulated by European cement standard EN 197-1. EN 197-1 defines granulated BFS, pozzolans, fly ash, burned slate, limestone and microsilica as substituents. In The Netherlands the use of CEM I (solely clinker based) is already reduced to about 30% of the total cement market, i.e. blended cements are dominating.

The building industry, and the concrete industry in particular, is also interested in the addition of by-products in concrete themselves. This offers them more freedom in regard to mix design, and the use of by-products that are not (yet) included in EN 197-1. The European concrete standards EN 206-1 and EN 15167-1 allow for the use of fly ash, silica fume and granulated BFS in a concrete mix.

One of the performance criteria that is gaining importance is the environmental footprint of the building material used. For concrete, these developments imply smart use of cement, which is the most energy- and cost-intensive component of concrete. A second, equally important motivation is cost reduction, as cement is the most expensive component in a concrete mix. For a sound concrete recipe, all the ingredients need to fit both physically and chemically: this is achieved by 'particle size engineering' and 'mineral oxide engineering'. In the following methods for designing alternative concretes are presented. Furthermore, the Dutch regulations for using "nonstandard" are discussed.

Particle size engineering

Particles of various sizes are combined to produce the concrete mix. The combination of all individual particle distributions results in an overall particle size distribution of the mix. This overall grading of the mix, containing particles from 300 nm to 32 mm, determines the mix properties in fresh state: the flow properties and workability. But the properties of the hardened concrete, such as strength and durability, are also determined by the overall particle size distribution. A mix design tool has been developed based on the insight that superior properties of a granular mix are achieved when a so-called geometric particle size distribution is obtained considering all solids in the mix (so not aggregates only), an idea already put forward in 1950 [3]. Geometric particle arrangements had been proposed much earlier [4], in their study of aggregate packing more than 100 years ago.

Using linear optimization, a fit of a granular blend containing all the solids used, is made according to the defined distribution function [5, 6]. Figure 1 shows the cumulative particle size distribution or PSD (close to lognormal distribution) of a number of ingredients (micropowder, cement, sands, gravel etc.), which are combined in a way that their mix best approaches the geometric packing represented by the 'target function'. The logarithmic scale in Figure 1 again illustrates the enormous particle size range in this typical concrete mix, namely five decades.

The concretes designed with the presented approach show excellent properties in both fresh and hardened states.



Figure 1: The cumulative particle size distribution (PSD) of all ingredients in a mix (measured with a laser granulometry and sieves).

Compressive conventionally vibrate concrete (CVC) and selfcompacting concrete (SCC) strengths of 30 to 60 N/mm² have been achieved with a total binder (cement clinker and cementitious byproducts) content of 200-270 kg/m³ [5]. Earth-moist concrete (EMC) with a compressive strength of 100 N/mm² has been produced with 325 kg/m³ OPC as sole binder. More importantly, it was seen that the flexural strength hardly decreased when the OPC content was reduced to 175 kg/m3 [6]. Much better workability and higher strength were obtained compared with concretes with the same cement contents. The developed design method allows for a more performance-based mix design. Many practical mixes of companies have been optimized

with regard to efficient cement and admixture use. In many cases this has resulted in the incorporation of alternative ingredients, such as

aggregates, inert powders (fine aggregate) and cementitious byproducts. For use as binder in mortar, concrete or stabilized waste, insight into the hardening reactions of cement clinker, cementitious byproducts and contaminants (if present) is required, which is the rationale for mineral oxide engineering.

Mineral oxide engineering

The hydration of the mineral oxides appearing in OPC, the most abundant binder, was first studied. Based on the water retention data provided by Powers and Brownyard [7], the hydration reactions of the four major clinker phases (C_3S , C_2S , C_3A , C_4AF) and their hydration products, such as C-S-H and CH etc., were quantified [8]. Next, reaction models for alkali-activated slag and slag-blended cement were established based on stoichiometric calculations [9]. The models correlate the mineral compositions of slag and Portland cement clinker, and their blending proportions, with the quantities and compositions of the hydration products formed, including their rate (reaction speed) of formation.

Blast furnace slags typically possess C/S ratios of about unity, and part of the CH produced by the hydration of C_3S and C_2S in the clinker is available to increase the C/S ratio of C-S-H formed from the slag. Chen [9] proposed that the fraction of consumed CH is proportional to the difference in C/S ratio of the slag and the C/S ratio of C-S-H produced by the clinker, namely 1.7, a concept that is compatible with reality (Figure 2).



Figure 2: Predicted and measured C/S ratio in C-S-H versus slag proportions in blended cement [9].

For the numerical simulation of the hydration reactions and the pore water composition, in the author's group a 3-D simulation model (CEMHYD3D) from NIST [10] was adopted and extended [9, 11, 12].

Performing cellular-automata like rules on the matrix of voxels simulates the hydration (Figure 3).



Figure 3: Development of OPC microstructure by CEMHYD3D [11].

Van Eijk [11] and Chen [9] modeled the pore water composition (e.g. pH), and incorporated the reactions of fly ash and slag. As well as the chemical extensions, the kinetics of the model has also been modified. In the original model the system resolution had considerable effect on the simulation results. The simulations performed with different system resolutions (from 0.5 to 2 μ m) demonstrated the robustness of the improved model [9]. Currently, CEMHYD3D has been extended with several additional possibilities which enable the hydration of particles as small as 0.2 μ m, designated as 'multi-scale' feature [12].

Conclusions

Changing environmental regulations and technical standards enable performance-based recipes rather than prescriptive-based recipes of cement-based construction materials, driving product innovation in civil engineering. The scientifically and technically driven design and production methodology will be continued to obtain more sustainable, durable and functional concrete. There are still plenty of practical problems to be solved, scientific questions to be answered, new raw materials that enter the market, and conceivable product innovations. Furthermore, the present approach can also be used to develop architectural building products/components such as cladding materials. The sensory properties are then of special interest, and this is an endless source of new ideas for our research.

Bibliographical references

- /1/ P. Graham: "Building ecology", Blackwell, Oxford, U.K. (2003).
- /2/ B. Berge: "The ecology of building materials" (2nd ed.), Elsevier, Amsterdam, The Netherlands (2009).
- /3/ N.M. Plum: "The predetermination of water requirement and optimum grading of concrete under various conditions", Building Research Studies No. 3/Statens Byggeforskningsinstitut Studie Nr. 3, The Danish National Institute of Building Research, Copenhagen, Denmark (1950).
- /4/ W.B. Fuller and S.E. Thompson: "The laws of proportioning concrete", Trans. Am. Soc. Civ. Eng. Vol. 33 (1907) pp. 222-298.
- /5/ M. Hunger: "Integral design of ecological self-compacting concrete", PhD Thesis, Eindhoven University of Technology, Eindhoven, The Netherlands (2010).
- /6/ G. Hüsken: "A multifunctional design approach for sustainable concrete, with application to concrete mass products", PhD Thesis, Eindhoven University of Technology, Eindhoven, The Netherlands (2010).
- 17/ T.C. Powers and T.L. Brownyard: "Studies of the physical properties of hardened Portland cement paste", Bull. 22, Res. Lab. of Portland Cement Association, Skokie, IL, U.S (1948), reprinted from J. Am. Concrete Inst. (Proc.) Vol. 43 (1947) pp. 101-132, pp. 249-336, pp. 469-505, pp. 549-602, pp. 669-712, pp. 845-880, pp. 933-992.
- /8/ H.J.H. Brouwers: "A hydration model of Portland cement using the work of Powers and Brownyard", Eindhoven University of Technology/Portland Cement Association, Skokie, Illinois, U.S., http://www.cement.org/ (2011).
- /9/ W. Chen: "Hydration of slag cement, theory, modeling and application", PhD Thesis, University of Twente, Enschede, The Netherlands (2007).
- /10/ D.P. Bentz: "Three-Dimensional computer simulation of Portland cement hydration and microstructure development", J. Am. Ceram. Soc. Vol. 80 (1997) pp. 3-21.
- /11/ R.J. van Eijk: "Hydration of cement mixtures containing contaminants, design and application of the solidified products", PhD Thesis, University of Twente, Enschede, The Netherlands (2001).
- /12/ A.C.J. de Korte: "Hydration and thermal decomposition of cement/calcium-sulphate based materials", PhD Thesis, in progress.