Recipes for porous building materials

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Download date: 11. Feb. 2019
Inaugural lecture
prof.dr.ir. H.J.H. Brouwers
2 July 2010

Recipes for porous building materials

Where innovation starts
Recipes for porous building materials, more with less

Presented on 2 July 2010
at the Eindhoven University of Technology
Mr. Rector,
Members of the Executive Board,
Ladies and Gentlemen,

The title of this inaugural lecture reads ‘Recipes for porous building materials, more with less’. This title will be elucidated backwards.

1. More with less
The theme ‘more with less’ has recently been used frequently, in particular with respect to sustainability1. ‘More with less’ is for instance the title of a recent national program for energy saving in existing housing stock. The ability to do ‘more with less’ was coined by Buckminster Fuller (1895-1983) as ‘Ephemeralization’: ‘more and more with less and less until eventually you can do everything with nothing’ (Wikipedia (2010)). Fuller’s vision was that ephemeralization will result in ever-increasing standards of living for an ever-growing population despite finite resources, and may be seen as a positive reply to doom scenarios which are predicted every now and then, such as by the ‘Club of Rome’. As well as being an author, inventor and futurist, Buckminster Fuller was also an architect, known for instance for his geodesic domes. An interesting spin-off of Fuller’s dome design conceptualization was the Buckminster Ball, which was the official FIFA-approved design for footballs from their introduction at the 1970 World Cup until recently. Also the first fullerene discovered was buckminsterfullerene C60, made in 1985, the name being a tribute to Buckminster Fuller, whose geodesic domes it resembles. Fullerenes are molecules composed entirely of carbon, in the form of a hollow sphere, ellipsoid or tube. Spherical fullerenes are also called ‘buckyballs’, and cylindrical ones are called ‘buckytubes’ or carbon nanotubes. This nanotechnology is considered as the next step in science, integrating engineering with physics, biology and chemistry. Fields of application include structural and skincare products, ICT (such as the already mentioned nanotubes), biotechnology, instrumentation and the environment,

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1 A similar theme reads ‘less is more’ from architect Mies van der Rohe, a precept for minimalist design, a trend in design and architecture where in the subject is reduced to its necessary elements.
and the estimated total annual production in 2010 is less than 10,000 tons (Pitkethly (2004)). By far the largest increase in use is predicted in the building materials sector.

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

<table>
<thead>
<tr>
<th>Material</th>
<th>Production (Mton)</th>
<th>Material</th>
<th>Production (Mton)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Timber</td>
<td>4000</td>
<td>Quicklime</td>
<td>130</td>
</tr>
<tr>
<td>Plastics and rubber</td>
<td>250</td>
<td>Glass</td>
<td>120</td>
</tr>
<tr>
<td>Steel</td>
<td>1400</td>
<td>Cement</td>
<td>2500</td>
</tr>
<tr>
<td>Gypsum</td>
<td>250</td>
<td>Concrete</td>
<td>15000</td>
</tr>
</tbody>
</table>

Table 1

Some of these materials are exclusively used in the building sector; others such as steel and timber are also used in other industries. As well as the huge amount of raw materials involved, enormous amounts of energy are also used for the production and transport of raw materials, building materials and products (Graham (2003), Berge (2009)). 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, or ‘more with less’.

3. Porous
Most of the materials listed in Table 1 are porous, which means they contain void spaces. Even materials that are normally non-porous, such as steel, glass and plastics, are also available in porous form, for example as foams. Porosity can be distributed regularly or randomly, and determines the mechanical, physical and chemical properties of materials. Regular porosity is found in regularly stacked stones, blocks etc. Void space is also found in crystal lattices, such as the cubic fcc, bcc and sc lattices, which are modeled as regularly stacked spheres. Kepler
hypothesized that fcc and hcp lattices, having a packing fraction of about 0.74 (and hence a void fraction of 0.26), constitute the densest possible packing of spheres. This so-called 'Kepler Conjecture' has recently been claimed to have been proved mathematically (Aste and Weaire (2000)).

It has been known for about 50 years that randomly stacked spheres also have a typical close packing fraction, of about 0.64 (Scott (1960), Scott and Kilgour (1969)). Also randomly packed non-spherical particles have their characteristic packing fraction (Figure 1). Building materials contain irregularly shaped particles, of various sizes (polydisperse), mostly randomly packed as is the case in concrete.

4. Recipes
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 porous material that often fulfills these requirements is concrete. Attention is focused on this material, but gypsum- and limestone-based materials are chemically and physically very similar, and will also be discussed briefly in this lecture.
Concrete

Among all porous building materials, this material is chemically and physically the most complex, scientifically the most interesting, and its worldwide production is larger than all other man-made materials combined (Table 1). Concrete is a spectacular material, not only because of its enormous production volume. A conventional concrete consists of cement, aggregates and water (Figure 2), 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. The maximum grain size is often limited by the maximum spacing between the reinforcing bars.

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
Recipes for porous building materials

almost maintenance-free, with a great architectural potential. The heaviest and tallest object ever moved by mankind to another position is the Troll A gas platform, a concrete construction with a total mass of 656,000 tons and a height of 472 m.

Aggregates include sand and gravel, which are extracted from rivers and the sea and are weathered rock material, or intentionally crushed stone material. Secondary sources of aggregates are slags and recycled (crushed) concrete. Due to the shortage of dredging and excavation concessions, more and more crushed stone such as limestone and granite is entering the Dutch building materials market. Crushing also produces inert powders (fine aggregates), i.e. particles of the size of cement. The quarrying of dimension stone (slabs, blocks etc.) also generates powders (stone flour) and aggregates as by-products.

Cement is the finest ingredient in traditional concrete. Reactive powders, such as fly ash and granulated slag, have similar fineness, as well as inert (non-reactive) powders such as the stone flour already referred to.

Cement is the most energy-intensive and costly ingredient of concrete. Cement is made by heating limestone (calcium carbonate), with small quantities of other materials (such as clay) to 1450°C in a kiln. The resulting hard substance is called ‘clinker’, which is then ground with a small amount of gypsum into a powder. This powder contains grains with a typical size of 300 nanometer (300 \(10^{-9}\) m) to 100 micrometer (100 \(10^{-6}\) m), and is called ‘Ordinary Portland Cement’, the most commonly used type of cement and usually referred to as OPC. Limestone is a sedimentary rock composed largely of the mineral calcite (calcium carbonate: \(\text{CaCO}_3\)). It is usually quarried from deposits made of skeletal fragments of marine organisms, formed millions of years ago. Clay consists mainly of fine particles of

![Figure 3](image-url)

Classic cement kiln (left) and modern rotary kilns (right).
the mineral oxides $\text{Al}_2\text{O}_3$, $\text{Fe}_2\text{O}_3$, $\text{SiO}_2$ (quartz), $\text{CaO}$, $\text{MgO}$, $\text{K}_2\text{O}$ and $\text{Na}_2\text{O}$, which are formed by the gradual weathering of rock material. As quartz is the hardest and most inert mineral in rocks, the largest chunks (sand and gravel) consist of quartz, and are used as aggregate.

The manufacture of cement clinker in the rotary kiln is an energy-intensive process. In recent decades the efficiency of these kilns has increased. Thermodynamically about 3.0 GJ/ton cement clinker is needed, the most modern rotary kilns can achieve 3.6 GJ/ton, and a typical average value for the already operating kilns in developed countries is 5.0 GJ/ton. The most modern kilns produce 10,000 tons of clinker per day (Figure 3)!

Energy efficiency is improved by replacing conventional fossil fuel with waste as a low-cost secondary fuel, e.g. ground car tires, bone meal, sewage sludge, paper sludge etc. (Figure 4). When using alternative fuels, sintering temperatures lower than 1450°C can be seen, which is also beneficial for energy saving (Engelsen (2007)).

Savings and optimization are also possible on the materials side. By-products from other industries can be used as a substitute for the feedstock. If these materials have cementitious properties, they can even substitute the end-product, clinker. The main motivation for substitution to prevent landfill and saving energy,
and to a lesser extent to reduce the depletion of raw materials (clay, limestone, gypsum stone). These raw materials are among the most abundant in the earth’s crust, and are still being formed by weathering of rock and by marine organisms\(^2\). Figure 5 shows an overview of the A-S-C content of binders (quicklime, OPC) and by-products that can substitute feedstock and cement clinker\(^3\).

![Figure 5](image)

**Figure 5**

Ternary phase diagram of binders and by-products.

Blast furnace slag (BFS) is a by-product from the steel industry and is a common substitute for clinker. The use of so-called slag cement results in very durable concrete and comprises about half of the Dutch cement market (Chen (2007)). Powder coal fly ash (class C and F) has a very similar composition to clay, and class F fly ash is similar to OPC. These fly ashes are produced in huge quantities (Table 2).

<table>
<thead>
<tr>
<th>By-product</th>
<th>Quantity (million tons)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal ashes</td>
<td>1000</td>
</tr>
<tr>
<td>Blast furnace slag</td>
<td>120</td>
</tr>
<tr>
<td>Steel slag</td>
<td>140</td>
</tr>
<tr>
<td>Flue gas desulfurization (FGD) gypsum</td>
<td>50</td>
</tr>
</tbody>
</table>

*Table 2*


\(^2\) The total annual quantity of limestone formed by marine organisms is about 8 bln tons (Morse and Mackenzie (1990)), much greater than that used for the production of clinker.

\(^3\) Cement chemical notation is used here: \(C = CaO\), \(S = SiO_2\), \(A = Al_2O_3\), \(F = Fe_2O_3\) and \(H = H_2O\).
Fly ash can also substitute clinker, and a substitution rate of 30% is already common (Figure 4). A recent study presented a substitution level of 50% (Baert (2009)). As well as fly ash, the sulfur dioxide is also removed from the flue gases of coal-fired power plants using limestone as reactant, yielding flue gas desulfurization (FGD) gypsum. This gypsum accounts for about 20% of the global gypsum production (Tables 1 and 2).

Gypsum is used in combination with cement clinker, but is also used as raw material for the production of the binder hemi-hydrate (‘Plaster of Paris’). When gypsum (calcium sulfate dihydrate) is heated to 165°C, hemi-hydrate is formed. Adding water to this hemi-hydrate results in rapid hardening (the reverse reaction) to gypsum. As this gypsum stone is slightly water-soluble, gypsum is mainly used indoor as plaster or in the form of plasterboard (‘drywall’). These gypsum plasterboards have excellent acoustic, thermal and esthetic properties (Figure 6).

The binder quicklime (‘burnt lime’) is produced by calcination of limestone (Figure 4), through which CO₂ is released. When this quicklime is mixed with water, it hydrates to slaked or hydraulic lime, CH or Ca(OH)₂. Plain hydraulic lime is used for plastering, but it can also be mixed with a medium-size aggregate (e.g. sand) to produce a lime mortar. In the presence of air, this hydraulic lime hardens back to limestone again, which can be used indoor and outdoor.

Mortars of quicklime, water and sand are also used for the production of sand-lime bricks and autoclaved aerated ‘concrete’. To activate the reaction of slaked lime with the crystalline silica (quartz) of the sand, the mortar is autoclaved in compressed steam at 190°C, through which a hydration product C-S-H (calcium
silicate hydrate) is formed. Sand-lime bricks and aerated concrete are fire-resistant, sound-insulating, load-bearing, water-resistant and lightweight building materials, and are used in the form of bricks, blocks and panels. It is worth pointing out that Roman concrete (‘Opus caementicium’) contained quicklime and volcanic ash (as source of amorphous mineral oxides) as binders, which hydrate to predominantly C-S-H, and also contained medium-size and coarse aggregates, making it a true concrete.

To summarize, most porous building materials are of mineral origin, and can be used in their plain form, e.g. in the form of blocks, slabs etc., or as crushed stone, yielding aggregates. On the other hand, they can be thermally treated to produce a binder, the most important being cement. Cement is mixed with water and subsequently hydrates (reacts with water) to a hardened product. This paste (binder plus water) is often mixed with aggregates, yielding a mortar when only medium-size aggregates are added, or concrete when medium and coarse aggregates are added (Figure 7). Fresh paste, mortar and concrete are cast in a formwork, which determines the shape of the hardened product. This shape can be basic (block etc.), but complex shapes are also possible. In fresh state, concrete is a concentrated slurry. The solid content in the mix is about 85%; the remaining volume fraction is taken by water and air.

It has been seen that the cement industry is reducing the environmental impact of cement, for example by fuel and material substitution. The building industry, and the concrete industry in particular, is also interested in a further reduction of cement clinker and even cement, for example by adding inert powders (i.e. fine aggregate) and reactive powders to the concrete mix. This demand for cement reduction is driven by various current trends in the building industry, namely the introduction of new procurement and contract forms such as Private Finance Initiatives (PFI) and Public Private Partnerships (PPP), and by more performance-oriented clients (both public and private).

At the same time, construction firms have changed their strategic focus from cost-efficiency to adding value for money for the client, resulting in new contract forms such as Design & Construct (D&C), Building, Operate & Transfer (BOT) and variants of these, which focus on the total cost of ownership rather than the investment costs alone. A positive development, associated with the procurement shift just described is that all EU member states have developed a CE mark for the building industry. The idea behind this CE mark is to facilitate cross-border trading. Within the building industry this implies that materials and products are judged more on functional demands and less on product specifications - the so-called

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4 European Community conformity marking.
‘defined performance design’. For the concrete industry in particular, this development is being driven by the *Equal Concrete Performance Concept*, clause 5.2.5.3 in the EN 206-1 standard. This creates competition and hence a tremendous demand for innovation by the construction industry, including the cement and concrete industry. For the gypsum- and limestone-based building materials industry, this performance-based mode of working is already common practice.

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’.

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**Figure 7**

Ingredients of a number of building materials, including their particle size and specific surface area, based on a graph by Prof. Dr. Dr. H. Pöllmann, Martin-Luther-University of Halle-Wittenberg, Germany.
Particle size engineering

Particles of various sizes are combined to produce the concrete mix. As discussed earlier, randomly close-packed monosized spheres can achieve a packing fraction of 0.64, and higher packing fractions are obtained by combining different sizes. This principle also holds for the concrete ingredients, consisting of irregularly shaped and continuously graded particles. 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.

In the author’s group, 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 by Plum (1950). Geometric particle arrangements had been proposed much earlier by Fuller and Thompson (1907), in their study of aggregate packing more than 100 years ago.

To apply this design method, we need to be able to characterize powders with respect to their granulometric properties, and to avoid their agglomeration. Two technological developments, which became available many years after Plum (1950), have enabled this development:

i. The particle size characterization of powders (particles from 100 nm to 100 µm).

ii. The introduction of contemporary superplasticizers. Contemporary laser diffraction technologies allow powders to be characterized, and with 3rd generation (polycarboxylate) superplasticizers they can effectively be dispersed.

With these considerations in mind, a design model based on the packing model referred to above was established. Using linear optimization, a fit of a granular blend containing all the solids used, is made according to the defined distribution function (Hüsken and Brouwers (2008)), Hunger (2010) and Hüsken (2010)).
Figure 8 shows the cumulative particle size distribution or PSD (close to lognormal distribution) of a number of ingredients (micro-powder, 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 8 again illustrates the enormous particle size range in this typical concrete mix, namely five decades.

The method has been successfully applied to the design of new concrete mixes, which are currently in production by a number of companies. Based on the present design method, recipes for self-compacting concrete (SCC) have been developed. This type of concrete is one of the most important recent developments in the building industry. Also recipes for conventional vibrated concrete (CVC) and earth-moist concrete (EMC) have been designed. Earth-moist (or ‘zero-slump’) concrete is used in the cast concrete products industry (such as paving stones, kerbstones and concrete pipes). These concrete mixes are rammed and vibrated in the rigid mould, and demoulding can take place almost immediately, so that short processing times with high production quantities are achieved. In contrast to CVC and SCC, the voids in these concretes are partly saturated with water, while the remaining void fraction is filled with air. Capillary forces between the finer particles combined with the inner friction of the mix provide the required so-called green (early) strength.

The cumulative particle size distribution (PSD) of all materials used in a mix (measured with Malvern 2000 and a √2 sieve set) and the PSD of the mix (dashed line) composed with the help of the mix design tool. The target function (solid line) is also shown (Hunger (2010)).
The concretes designed with the presented approach show excellent properties in both fresh and hardened states. Within the framework of the EU Integrated Project ‘I-Stone’, SCCs have been designed and produced with a D\text{max} of 32 mm, with recycled aggregates, with microencapsulated phase changing materials (PCMs), with photocatalytic TiO\textsubscript{2}, with dimension stone ‘waste’ etc. Concretes with a specific density of 1100 kg/m\textsuperscript{3}, a thermal conductivity of 0.27 W/mK and a 28-day compressive strength of 37 N/mm\textsuperscript{2} have also been developed using lightweight aggregates ((Hunger (2010), Hüsken (2010)). A spin-off project, with the province of Overijssel, the municipality of Hengelo and Struyk Verwo Groep, concerns a street paved with 1000 m\textsuperscript{2} TiO\textsubscript{2}-containing photocatalytic concrete, a patented technique (Murata et al. (1999)). This street is currently being monitored and modeled using CFD by Dr. Milagros Ballari, and so far an NO\textsubscript{x} reduction of 25-45% has been observed (three separate measurements during spring 2010).

Compressive CVC and SCC strengths of 30 to 60 N/mm\textsuperscript{2} have been achieved with a total binder (cement clinker and cementitious by-products) content of 200-270 kg/m\textsuperscript{3} (Hunger (2010)). EMC with a compressive strength of 100 N/mm\textsuperscript{2} has been produced with 325 kg/m\textsuperscript{3} OPC as sole binder. More importantly, it was seen that the flexural strength hardly decreased when the OPC content was reduced to 175 kg/m\textsuperscript{3} (Hüsken (2010)). Much better workability and higher strength were obtained compared with concretes with the same cement contents. In this regard, the property cement efficiency was introduced, defined as compressive strength (N/mm\textsuperscript{2}) per unit of cement content in a concrete mix (kg/m\textsuperscript{3}). Equivalently, it has become possible to design equally performing concretes with less superplasticizer (and without viscosity modifying admixtures) and high cement efficiencies, in other words, ‘more with less’.

Within the framework of the EU Integrated Project ‘I-SSB’, Qingliang Yu MSc develops self-compacting hemi-hydrate based mortars containing lightweight aggregates and doped TiO\textsubscript{2} (for indoor air purification), using the new mix design concept.

Paste/mortar/concrete flow is analyzed by standard empirical tests, such as slump flow (Hägermann, Graf, Abrams) and V-funnel, and can also be measured by using a rheometer, which indicates yield stress and viscosity. Models for the viscosity of geometrically graded slurries are in development. As simplest case, the viscosity of a concentrated suspension of monosized particles was determined first (Figure 9).

Hunger (2010) analyzed the void content (water and some air) of various self-compacting mortars and concretes, and Hüsken (2010) that of earth-moist mixes. Their solids are graded geometrically (e.g. see Figure 8). By comparing the computed solid fraction with an analytical expression for geometric packings
(Brouwers (2006)), it was found that the packing fraction falls between those of random close packing and random loose packing, with the earth-moist mixes being closest to random close packing, as would be expected. The water in the mix is needed to fill the voids and to lubricate the grains; Hunger (2010) found a water film thickness of 24 nm surrounding the powders in a flowing paste. In the M2i project ‘Nanosilica in concrete’, George Quercia MSc applies the mix design concept to submicron particles. First results indicate that nanoparticles are surrounded by the same water film thickness as the powders (so 24 nm) when they are present in a flowing paste. Nanosilica particles produced by Alberto Lázaro MSc in the EU Integrated Project ‘Promine’ will also be used (Figure 10). He is continuing the research by Lieftink (1997) and Jonckbloedt (1997), who first investigated this patented production method (Schuiling (1987)). By the inclusion of nanosized particles the size range of a concrete can be extended to seven decades (Figure 7), enhancing cement efficiency and improving product properties (Vijayareethinam (2009)).
For the design of concrete, traditional methods and prescriptions concern for instance the cement content, the total content of particles smaller than 250 µm and the water-cement ratio. The developed design method allows for a more performance-based mix design. Many mixes of members of the sponsor group\(^5\) 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 by-products. For use as binder in mortar, concrete or stabilized waste, insight into the hardening reactions of cement clinker, cementitious by-products and contaminants (if present) is required, which is the rationale for mineral oxide engineering.

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Former members are: Delta Marine Consultants, Jaartsveld Groen en Milieu, Rokramix, Agentschap NL Soil+, Betoncentrale Twenthe, Betomortelcentrale Flevoland, Kijlstra Beton, Struyk Verwo Groep, Hülskens, Dusseldorp Groep and Eenland Recycling.
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 (1948), the hydration reactions of the four major clinker phases (C$_3$S, C$_2$S, C$_3$A, C$_4$AF) and their hydration products, such as C-S-H and CH etc., were quantified (Brouwers (2004, 2005)). Next, reaction models for alkali-activated slag and slag-blended cement were established based on stoichiometric calculations (Chen (2007)). 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.

Blast furnace slags typically possess C/S ratios of about unity, and part of the CH produced by the hydration of C$_3$S and C$_2$S in the clinker is available to increase the C/S ratio of C-S-H formed from the slag. Chen (2007) 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 11).

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
(Bentz (1997)) was adopted and extended (Van Eijk (2001), Chen (2007)). Performing cellular-automata like rules on the matrix of voxels simulates the hydration (Figure 12). During one hydration cycle the phases of all voxels are updated based on their current phase, the phase of their adjacent voxels and a set of rules describing dissolution, reaction and diffusion. During one hydration cycle, part of the cement mineral phases and gypsum that is exposed to water may dissolve and react in the same step with this water, forming the diffusing hydration products. Performing random walks in the solution until they precipitate or react further simulates this diffusion of reaction products. All reactions take place on a volume base. After a defined number of total diffusion steps a new cycle starts and a new part of the cement dissolves, creating new diffusing species.

Van Eijk (2001) and Chen (2007) 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 µm) demonstrated the robustness of the improved model (Chen (2007)). Currently, CEMHYD3D has been extended with several additional possibilities which enable the hydration of particles as small as 0.2 µm, designated as ‘multi-scale’ feature (De Korte (2011)). The model is modified for the reaction of hemi-hydrate to gypsum (as final product). Because the hydration time of hemi-hydrates is very short compared with that of cement, the cycle-time relation has to be shortened. CEMHYD3D has therefore successfully been extended with the option of ‘multi-time’ modeling (De Korte (2011)).
Chen (2007) combined mineral oxides of by-products to develop a shrinkage compensating admixture for OPC. Furthermore, in the author’s group several secondary binders have been designed and tested for members of the sponsor group. Motivated by clause 5.2.5.3 of the EN 206-1 standard referred to earlier, these companies dose cementitious by-products to the mix themselves, and follow the relevant attestation route. This development has increased utilization of cementitious by-products from different sources, both domestic and abroad, so not only the traditional blast furnace slag and powder coal fly ash. Both suppliers and appliers (concrete companies) of these by-products are assisted by investigating their suitability as binder. For their application as binder in concrete, three technical criteria are important as well as price (sometimes negative if it relates to ‘waste’), constant composition and availability in sufficiently large quantities:

i. The workability, hardening and related strength development.
ii. The durability of the product.
iii. The presence of contaminants.

A first indication for the suitability of a material as cement substitute is obtained by preparing standard mortars and studying the fresh workability, and the subsequent strength development during 28 days. For this purpose also CEMHYD3D is used. For these cementitious by-products, it can also be opportune to blend them to obtain a composition of particle sizes and mineral oxides with optimum reactive characteristics.

Durability, for instance, can be assessed by measuring water absorption, water intrusion and freeze-thaw resistance, and by accelerated chloride migration tests. An emerging test is the rapid chloride migration (RCM) test, developed by Tang (1996). The apparent diffusion coefficient determined by this method follows from assuming Cl⁻ concentration profiles in the specimen as shown in Figure 13 (left).

figure 13

Chloride profiles in a concrete during RCM test: at the left the assumed profile, and at the right a measured profile and the model prediction by Spiesz et al. (2010).
A typical example of a true measured profile, however, can be seen in Figure 13 (right). Przemek Spiesz MSc is working on a new diffusion model based on non-linear (Freundlich) Cl⁻ absorption by the concrete and non-equilibrium with the pore water, yielding excellent agreement (Figure 13). This new model yields a more realistic diffusion coefficient together with the mass transfer coefficient. This research, sponsored by the Dr. Ir. Cornelis Lely Foundation, will be expanded in a joint STW (IS₂C) project with Prof. Dr. Ir. J.C. Walraven (Delft University of Technology); three PhD candidates will study the chloride and moisture transport in cracked and uncracked concrete.

As well as strength and durability, the purity and maximum level of contamination, both in composition and in leaching, are also relevant. In the Netherlands, the Soil Quality Decree sets limits for the composition and leaching of granular materials, designated as ‘non-shaped’, with respect to chloride, mercury, molybdenum and strontium, for example. If these requirements are not met, the granular material is considered as ‘waste’, and landfill of these materials is a remaining option. The composition and leaching are also leading in selecting the type of applicable landfill, namely for mineral or hazardous waste, and the relevant safety measures as laid down by the Landfill Decree.

By stabilization, e.g. combining different materials and adding primary binders (quicklime/hemi-hydrate/cement), a granular (‘non-shaped’) or a shaped product can be obtained for non-hazardous landfill, and sometimes it is even possible that the treated material meets the Soil Quality Decree requirements of a building material.

Contaminated soil and dredged soil material are interesting ‘wastes’ that can be combined with primary binders and cementitious by-products (whether or not contaminated). By treatment of these ‘wastes’, cheaper landfill or even the application as building material becomes feasible. This saves the production of primary building materials, as well as the space and costs associated with landfill. A few stabilization projects have been carried out in which contaminated soil and dredged spoils have been made into acceptable building materials (Brouwers et al. (2007) and De Korte (2011)). Non-shaped stabilized wastes can be used as road base material, traffic noise barriers, etc. Shaped stabilized wastes may be used as plain (unreinforced) concrete products such as slabs and blocks.

Miruna Marinescu MSc uses the hydration models to relate (Freundlich) binding and transport of anions (chloride) and cations (heavy metals) and microstructure. This is related to binder recipe and hardening conditions (Marinescu and Brouwers (2010)). Fixation of heavy metals is important for the stabilization of waste, binding of chloride for stabilization and the durability of concrete structures, as seen before.
1. Chair and unit
It has been seen that changing regulations and standards enable performance-based recipes rather than prescriptive-based recipes, driving product innovation. The scientifically and technically driven design and production methodology will be continued to obtain more sustainable, durable and functional materials and products based on the binders cement, hemi-hydrate and quicklime. 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, to achieve "more with less".

Furthermore, clay products have been investigated by the staff already present in the chair (Ir. Bert van Schaijk and Dr. Ir. Ton van der Zanden), such as the production of fired bricks and moisture transport in bricks. Bricks, roof tiles and sewage pipes are examples of fired clay products. The raw materials, clay and loam, are granular materials and contain similar minerals to cement and concrete. It is conceivable that particle size and mineralogical engineering can also be applied to these materials. The testing of the raw materials and the fired clay products is also similar, and the fired clay materials and the hemi-hydrate and quicklime binders share the same history. Unfired clay and loam are also used for the construction of buildings. Walls covered inside with a layer of loam work well to control air humidity. Combined with straw, loam is used as a construction material to build walls. This building technique is more than 10,000 years old, and one-third of the world population still live in earth buildings. Unfired clay is also used in infrastructure constructions such as dike cores, and in the production of cement (Figure 4). In addition to all these mineralogical materials, it would also be interesting to study biological materials, plastics and metals, in particular steel, which is the common partner of concrete in concrete structures.

For adjacent chairs in my Building Physics and Systems unit, physical (especially thermal), chemical and biological properties of building materials are of interest. In my group we have developed materials with increased thermal mass by incorporating microencapsulated PCMs (Hunger et al. (2009)), which are currently field-tested (Entrop (2011)). We are also developing materials that are self-cleaning and air purifying (also indoor), and we are modeling air quality using a Langmuir-Hinshelwood model for the kinetics and CFD for the fluid dynamics (Hunger (2010),
Hüsken (2010), Yu and Brouwers (2009), Yu et al. (2010)). In other words, there are ample opportunities for cooperation.

2. Faculty
Cooperation with the three other units is also obvious. The mechanical and physical material properties are relevant for the Structural Design and Construction Technology unit. Together with this unit, Dipl.-Ing. Götz Hüsken has developed an SCC for the B-invented innovative concrete foundation (‘B-smart’). We will also cooperate in the development of steel fiber concrete and impact resistant concrete. Discussions are ongoing with the Architectural Design and Engineering unit about the development of 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. The air purifying and self-cleaning concrete just referred to is already of interest to architects. This type of concrete has been applied in the Dives in Misericordia church in Rome (architect Richard Meier), and the George Harrison Memorial Garden in London. And the lightweight SCC mentioned above is suitable for monolithic concrete buildings. Projects in the field of energy saving in the existing building stock are possible with the Urban Management and Design Systems unit. The environmental performance of buildings we assess with tools such as Greencalc+, BREEAM, LEED etc. The building envelope then plays an important role. These topics are addressed by Entrop (2011), in a PhD project sponsored by Agentschap NL (EOS-LT). The cellular automata approach of cement hydration used in my group (Figure 12) also connects to the travel simulation models developed in this unit.

3. University
At the University of Twente (UT) there was always a good cooperation with the chairs of Mechanics of Forming Technology (Prof. Dr. Ir. J.H. Huétink), Production Technology (Prof. Dr. Ir. R. Akkerman) and Elastomer Technology and Engineering (Prof. Dr. Ir. J.W.M. Noordermeer), concerning metals, plastic composites and rubbers, respectively. Other types of materials such as plastics, metals, semiconductors, photovoltaic etc. are being developed in other TU/e groups. We have contacted some of these groups already, and we will continue exploring the possibilities of sharing models, experimental facilities and setting up joint projects. In 2004 TU/e selected the New Materials cluster as one of three focal areas. Perhaps the former TU/e coordinating working group for materials needs revitalization.
4. Clients
The satisfaction of our clients with our major products, education, alumni, know-how and artifacts, is both the source of and the justification for our teaching and research funding. The activities of our Department of Architecture, Building and Planning are obviously aimed at the building sector, which comprises about 10-15% of the GDP of most countries. About 70% of the total value of the Netherlands consists of (residential and utility) buildings (CBS (2009)).

The building industry is by nature locally oriented, and makes use of local materials and mostly local manpower (which includes our alumni). The simple fact that material prices are low and volumes large mean that building materials, products and components travel only short distances. This principle affects the whole building chain, in other words all disciplines in our Department of Architecture, Building and Planning: building is a local activity.

The background and expertise of the department staff more or less reflect the activities of the complete building chain. Furthermore, considering the excellent computational, experimental and demonstration facilities, our department can perfectly address the questions and needs of our clients. This ability to secure contract funding is becoming increasingly important because of the declining levels of direct funding by the government. Thanks to the close relations with the building sector, our department is well positioned to cope with this trend.

The cooperation with the sector also provides a valuable channel for our students, who constitute one-quarter of the total student population at this university, to building practice and career start. In my chair we will continue working with and expanding the sponsor group (almost completely domestic) and our partners abroad. This cooperation has resulted in many joint research projects, currently enabling 11 contract research positions (of which two at UT).

In Dutch public debates, also at TU/e (Lintsen (2010)), the role of the university in our modern society is often discussed, or the fundamental question is raised: “whereto is the university on earth?” Research at universities of technology is often difficult to distinguish from research in technical institutes and by research departments in larger companies. Scientific education is a unique feature of a university, and I therefore believe that our most important task is the education of young people, and preparing them for their future, so that their study investment proves to be a sustainable one. Our answer to the above question could therefore read: “Our university is on earth to make students happy during their student days and afterwards”. I am therefore pleased that almost 50 students have taken my elective Master course in Sustainable Building, which was taught for the first time in the past quartile.
The inaugural lecture is a suitable occasion to look back on my life, and to thank people and organizations. First, I would like to thank the management of the university, my department and unit for putting their trust in me and giving me all the opportunities to develop my activities. I greatly acknowledge my introduction by Dik-Gert Mans and Cees Kleinman to Bert Snijder of the Department Board, the subsequent firm direction of the application procedure by the dean Jan Westra, as well as the help of managing director Paul Scholte with the transfer of my group to Eindhoven.

I did not move to Eindhoven alone. Martin Hunger, Götz Hüsken, Qingliang Yu, Milagros Ballari, Przemek Spiesz and Miruna Marinescu, I appreciate your courage to follow me, and I am pleased to see that you and your families have settled down here successfully. This also applies to Alberto Lázaro and George Quercia, who started recently at TU/e, and who will soon be joined by two new PhD candidates. Bram Entrop and Ariën de Korte, although you preferred to stay at the University of Twente, my interest in your PhD projects and in supervising you is unaltered.

My previous job was at the University of Twente. Nineteen years ago, Herman Wind and Henk van Tongeren recruited me as associate professor for their newly established Department of Civil Engineering and Management. They gave me the opportunity to develop my technical research in a predominantly management-oriented department. Later, during the never ending discussions on the preferable management line, I appreciated Huib de Vriend’s attention and support for my technical research.

After the transition to the newly established Faculty of Engineering Technology in June 2001, I was able to work pleasantly in joint projects with Geert Dewulf and Joop Halman, and perhaps we can formulate new joint projects in the field of Sustainable Building. The encounter with Hans-Ulrich Hummel in Boston, a few months after the 9/11 events and in the week George Harrison passed away, and the subsequent contact with Maria Founti, opened the doors to joint European

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6 Head of R&D and board member Knauf Gips, and professor of Inorganic Chemistry at the University of Erlangen-Nürnberg, Germany.

7 Professor of Thermal Engineering at the National Technical University of Athens, Greece.
projects, which meant a tremendous boost for the development of my research group. And finally, I would like to thank my former dean, Rikus Eising, for the smooth transfer of my group to Eindhoven. At UT several projects are still running under my supervision, and I will do my best to complete them as foreseen at their start.

After graduating from this university, my first job was at Akzo Nobel Central Research Arnhem. I started working there at the same time as Hans Meerman, a graduate from the same study and year, with whom I worked very closely and pleasantly together, and we never lost sight of each other. After one year of working it appeared that my research on plastic heat exchangers was suitable for a PhD thesis, but after two years this company project was ending. I appreciate the help of my managers, Geert Vegt and Hugo Korstanje, for encouraging and helping me to complete the PhD thesis.

This also brings me to my private life. It was my older brother Bert who paved the way to an academic study and career. He is 14 years older, and I have always been able to count on his advice and help. And now we have become colleagues at this university; but the older brother will always be the older one. I would also like to thank my wife for her support and patience. And finally, I would like to close by thanking my parents, who not only created me, but also raised me. Now I am at the same age as my father when I was born, and I am pleased that he can attend this lecture. In this lecture I have referred to George Harrison. I would like to conclude with the words of the Dutch composer and singer “Vader Abraham”:

“Bedankt lieve ouders, bedankt dat mijn wiegje in uw huis eens mocht staan”.

Thank you for your attention.

I have spoken.

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8 “Thank you dear parents, thank you that my little cradle was once welcome to stand in your house.”
References


CBS (2009), Nederland is 3.500.000.000.000 euro waard, Webmagazine, 14 December 2009 (in Dutch).


Plum, N.M. (1950), 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.
Wikipedia (2010), Buckminster Fuller, Wikipedia online encyclopaedia.
Jos Brouwers (1963) graduated in mechanical engineering (1986) at Eindhoven University of Technology. He joined Akzo Nobel Central Research Arnhem to work as research engineer and project leader in the field of plastic production processes and products such as synthetic fibers. He gained his PhD (1990) in the Department of Applied Physics at Eindhoven University of Technology with a thesis on ‘Film models for transport phenomena with fog formation, with application to plastic heat exchangers and condensers’. In 1992 he moved as associate professor to the Department of Civil Engineering and Management at the University of Twente. He was responsible for education and research in the fields of construction materials and sustainable building. In July 2007 he became guest professor at Wuhan University of Technology in China. In September 2009 he was appointed full-time professor of Building Materials in Eindhoven. He has published about 180 publications, of which more than one-third in refereed journals; and he is sole author of almost half of them. His research is among others funded by the European Commission, STW, Dr. Ir. Cornelis Lely Foundation, M2i, Agentschap NL and the province of Overijssel, and by a sponsor group consisting of public and private organizations. He established this sponsor group in 2003, and its members support the chair in kind (materials, equipment, know-how and data) and with a contribution in cash9.

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9 Each member contributes € 5000 (excluding VAT) per year.
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