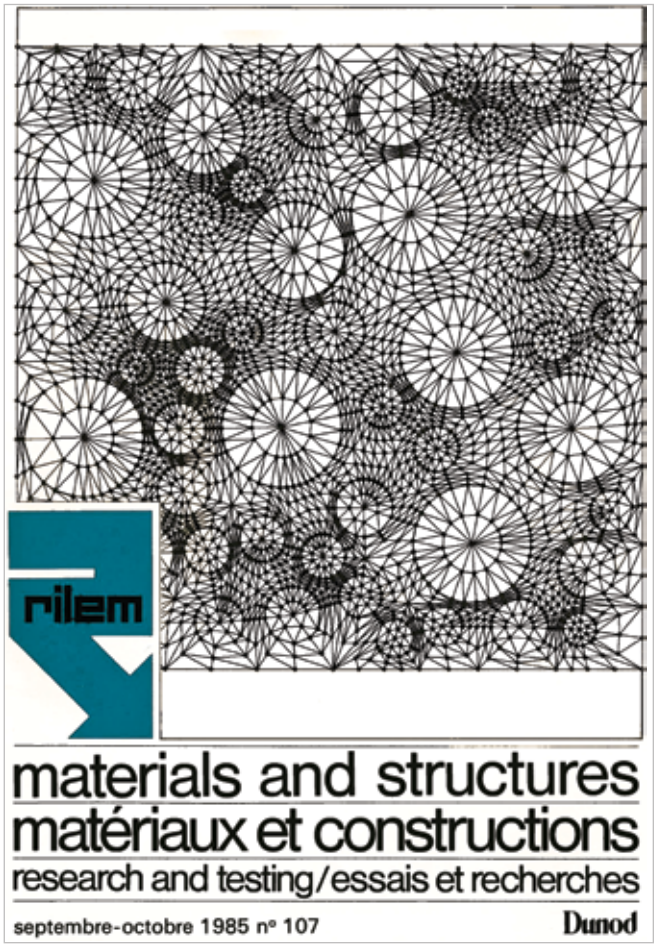


(Credit: *Materials and Structures*)

Cover of journal in which "Le beton numerique" was published. The cover shows the 2D finite-element mesh incorporating the circular sand and gravel particles.

The computational materials science of concrete:

Past, present and future

By Edward J. Garboczi

Computational materials science of concrete is now a viable discipline that bridges the gap between fundamental principles and practical applications.

The development of my career parallels the historical growth of the computational materials science of concrete. Thus, my hope is that my personal point of view might help some of the younger ACerS members see how a career can develop (mostly unplanned). In addition, I believe the facts of history can be much more stimulating when acquired in a biographical context, another reason for the added personal touch.

From its humble beginnings back in the 1960s, computational materials science has come a long way. The field currently is getting even more exciting with ideas like integrated computational materials engineering[1] being seriously considered in the beginning of national programs like the Materials Genome Initiative.[2] Of course, the development of computational materials science, in general, has gone hand-in-hand with the startling increases in computer processing speed and memory over the same time period. The computational materials science of concrete is no different. With this type of work now firmly established for cement and concrete, this seems like a good time to examine the origins of this field from a viewpoint—possibly a bit distorted by looking through my personal lens—and to delineate where it needs to go in the future, with, I hope, a more objective vision.

My background

In 1985, I earned a PhD in condensed matter physics from Michigan State University, with a thesis on computer simulation models of the elastic properties of amorphous semiconductors. That was 27 years ago, but my thesis work exemplifies the rapid development of computers since then. For example, to finish up my thesis research, I tied up the Michigan State University mainframe computer—all two megabytes of its RAM—for several weeks! My thesis advisor was Michael Thorpe, and, in subsequent years, I have told him that I am still doing random condensed matter physics, but just at a different length scale!

From 1985 to 1988, Armstrong World Industries in Lancaster, Pa., employed me to work on calcium phosphate cements for inorganic ceiling boards, and I began learning a magic word: "microstructure." At Armstrong, we were attempting to develop a macro-defect-free (MDF) twin-roll process for our patented inorganic cement. In this pursuit, I studied the MDF literature of the 1970s and 1980s and was eventually led to a special 1983 issue of the *Philosophical Transactions of the Royal Society of London* journal devoted to new developments in cements for the 1990s.[3] Reading this issue fired my interest in cement and concrete in general, and I moved to NIST in 1988, replacing my friend Hamlin

Jennings who had left in 1987 for Northwestern University.

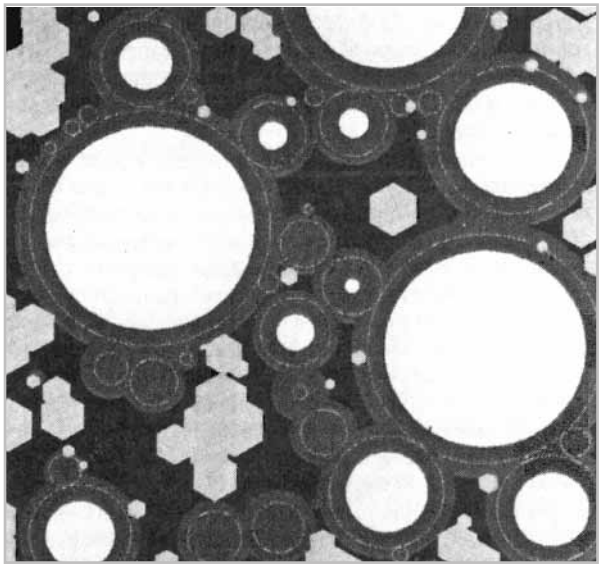
At NIST, my division chief was visionary scientist and research leader Geoffrey Frohnsdorff (1928–2006), who had a key article in that 1983 issue.[4] Frohnsdorff cofounded the ACerS Cements Division, one of his many accomplishments that have helped our field tremendously. He was a modest man, who, at his retirement, said that his career had consisted of "two or three good ideas … and the rest of the time spent signing bits of paper." One of his good ideas was, in 1968, developing, programming, and publishing the first computer model of cement hydration.[5]

Model building

Although it was not as clear to me at the beginning as it would be later, my long-term goal at NIST was to make the computational materials science of concrete a viable discipline, one that could work with experiment as an equal partner to solve the hard problems posed by this complex, random, multiscale material.

Several physicist friends told me, "Physicists do not work on messy problems like concrete." Civil engineering workers told me that fancy models were not going to be useful to "the average concrete worker." But, I was open to the idea of going against "common-sense" because Albert Einstein, one of my heroes, said, "Common-sense is (only) the collection of prejudices acquired by age eighteen." I wanted to break through the common-sense prejudices about concrete that were then current among most materials scientists.

I explicitly went to NIST, at first, to further develop a 3D cement-paste microstructural model that Jennings and Johnson were the first to create in 1986.[6] This model approximately handled processing (e.g., cement hydration) and microstructure, and, thus, had two of the three components of the classic processing–microstructure–properties triad. However, I quickly realized that there was a major limitation on the model's future usefulness: It would be very difficult to compute material properties based on how it stored the model's microstructure, viz., an approach based on centers and radii of spherical particles in a virtual container. I needed to get this property computation part operational. Without that component, the model could not be truly called a materials science model, and its results could not be directly compared with experiment.



(Credit: Jennings and Johnson; *Journal of the American Ceramic Society*)

Illustration of 2D cross section of the Jennings–Johnson 1986 cement paste model.

To put the properties issue in context, one of the difficult problems in the field that spurred this model development was the need to understand concrete durability. Because controlled degradation experiments typically took decades (and even accelerated experiments could take months), an accurate microstructure development–property model would enable virtual experiments to be run in much shorter times. A second hard problem was material optimization. Concrete has many possible components, and it is a massive job to attempt experimentally to optimize among them. An accurate model would do this much faster.

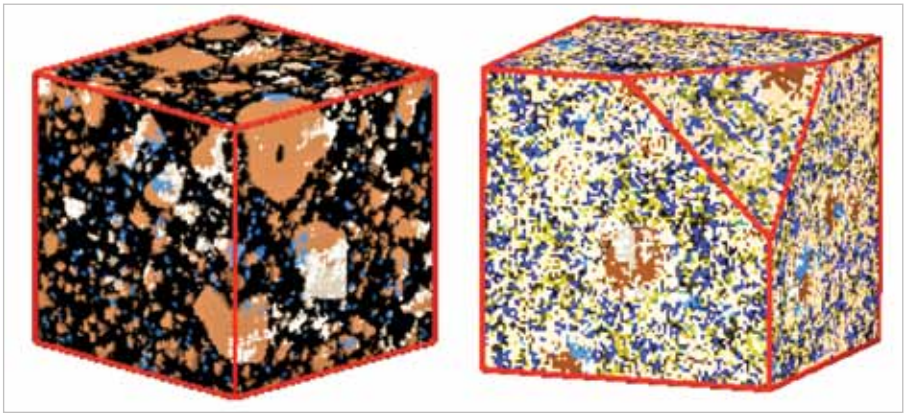
Four sources helped inspire my ideas about how to calculate properties based on microstructure, which ultimately led to a new type of model. The first source was the field of my PhD work, amorphous semiconductors. Researchers in the 1960s had developed computational models of atoms, randomly linked together to simulate the structure of these materials, on which to calculate properties and give computer-based theoretical predictions to guide and interpret experiments.

The second intellectual source was "Le béton numerique," by Roelfstra, Sadouki, and Wittmann.[7] They developed a simple 2D model of concrete structure, which was then "wired up" with a finite-element mesh to predict properties.

The third intellectual source was an algorithm, developed by my friend Larry Schwartz at Schlumberger,[8] that used the random trajectories (random walks) of mathematical points in a pore space to compute the diffusivity of a porous material for a Jennings–Johnson spheres-in-a box type of model.

My fourth source came from what I learned with my colleague Dale Bentz about representing microstructure information, whether from a model or from some type of microscope or camera, in a digital image format. Combining the idea of random walks, digital images, and property calculations led Dale and me to a new type of cement paste model whose random digital structure was built up by random walks of various chemical phases.

The digital image format breakthrough was the last piece needed for the ability to compute any property via finite-difference and finite-element algorithms, based on the 3D digital lattice structure, although with some restrictions on microstructure length scales. I concentrated on developing new algorithms explicitly written for property computation in digital image models, while Dale worked on further developing the cement paste microstructure model, soon to be called CEMHYD3D.[9] Realistic (complex) cement mineralogy was added with the help of Paul Stutzman. The support of and collaboration with the National Science Foundation (NSF) Center for Advanced Cement-Based



VCCTL/CEMHYD3D cement paste model—before hydration (showing multiphasic cement particles) and after hydration (showing the many new phases and remnants of the old phases).

Materials (ACBM), headquartered at Northwestern University, also was very important to this effort in the 1990s.

The combination of these four ideas led to what truly could be called a computational materials science model. Around 1990, I started using the actual term "computational materials science of concrete."

Practically speaking, concrete is a multiscale material with random structure from the nanometer to the centimeter and beyond. Given this, I next wanted to move into concrete-length scales so that the term "computational materials science of concrete" could be used more truthfully. To do this, we first used spheres as simple models of aggregates, such as sand and gravel, packed into a cement paste matrix. We later developed 3D models of real-shaped sand and gravel, based on X-ray computed tomography, which brought more accuracy to our concrete models. (These random shape measurement and mathematical procedures also are used to analyze other particulate materials outside the concrete world, including simulated lunar soil, shredded tires for waste treatment systems, breast cancer tumors, crushed waste glass, chemical explosives, and metal powders for additive manufacturing.)

Aside on simple vs complex models

We now have most of these models integrated into the VCCTL—Virtual Cement and Concrete Testing Laboratory—that covers the micrometer to millimeter length scale of concrete. Companies are starting to have success using this computational materials science code to guide and replace some of their testing. Because the VCCTL is a "desktop computing" software package, some of our more complicated models that require much more computer power cannot be part of it. But, the VCCTL becomes more effective with more desktop computing power and, of course, that is constantly improving.

There is one continuing argument about the computational materials science of concrete that occurred at the beginning of the field and unfortunately continues today. Basically, the experimentalists and industrial workers want models that run quickly on a desktop computer. On the other hand, theorists want to use powerful, more complex and realistic models, requiring parallel supercomputers that can handle the fundamental chemistry and physics of cement-based materials.

My view is that this is an artificial competition and unfortunately often involves disparaging the one by insisting that the other is the only valid approach. Indeed, there is a difference between models that daily can help experimentalists and industry, and models that help us understand complex, fundamental issues. The correct way to link the two is to build simple, valid, nonfitting models that can run on desktop computers that are abstractions from validated, realistic, complex models that are run on parallel supercomputers.

Unfortunately, at least in the concrete world, simple models are often used that have not been linked to more complex and realistic models. As Einstein also said, "Everything should be made as simple as possible, but not simpler." One use for the computational materials science of concrete is to solve problems accurately and to validate and improve the simpler models in everyday use in the cement and concrete world.

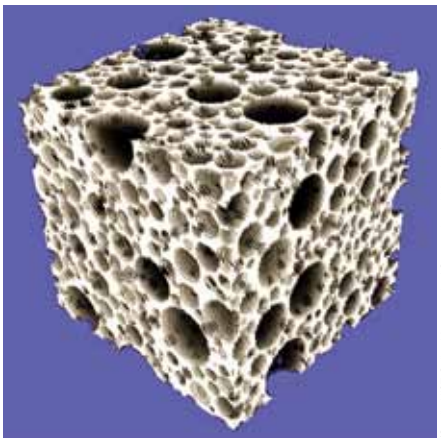
Encouraging signs

Currently, various signs suggest that the computational materials science of concrete field is fairly healthy. There is now much more acceptance in academia and industry of this endeavor than there was 20 years ago. What has caused this sea change? Clearly, we can attribute it to success in solving real problems and accurate comparisons to real experiments.

In addition, the educational component has been crucial. The annual three-day ACBM-NIST Computer Modeling Workshop has taught the rudiments of the computational materials science of concrete to more than 600 people since its beginning in 1990, and the *NIST Educational Monograph* ("An electronic monograph: Modeling and measuring the structure and properties of cement-based materials," see http://concrete.nist.gov/monograph), begun in 1997, has educated thousands more across the world in these topics. The influence of the *Monograph* is reflected in the metric that between 6,000 and 10,000 independent IP addresses currently access it each month.

As a reviewer for and reader of journals, I find much more computational work now than in the past, in the United States, Europe, and Asia. I even find civil engineers doing molecular dynamics, a practice that 20 years ago I never expected to happen in my lifetime.

Computer power continues to increase rapidly with parallel processing and graphical processing units (GPUs). As always, progress in this field goes hand in hand with progress



(Credit: NIST Online Monograph of Cement-based Materials.)

Figure X. As this image shows, computing the transport through the cement paste surrounding the aggregates is a difficult problem. In the image, the model aggregate has been rendered transparent, so that only the cement paste matrix is seen.

in computer hardware and software. For cement-based materials like concrete, the NSF explicitly has been calling for combined experimental and computational approaches. Molecular- and nanometer-scale models are starting to be generated at many institutions. For example, a large center for this type of modeling is at the Massachusetts Institute of Technology, where work is funded by members of the cement and concrete industry. This type of support is very encouraging.

So, for the first time, we have the opportunity to start assembling a multiscale suite of models, version 1.0, because concrete, as I have said before, is really a multiscale material. There will be many gaps, but it is good to identify the gaps so we know where to focus future attention. In the world of materials, in general, ideas such as Integrated Computational Materials Engineering (ICME), advanced by the National Academy of Science, are taking hold. ICME—the idea that experiments should be performed together with realistic computational models—is the intellectual driving force behind the Materials Genome Initiative (MGI), which can be summarized as "new materials by computational design."

Where do we go from here?

The goal of MGI, an initiative closely related to manufacturing, is to accelerate materials design via computation enabled by data repositories, models, and information transfer links. There is a large opportunity for concrete in this approach—if we can take the models to the next step to make them more suitable for design. The current elaboration of the computational materials science of concrete is based on the experimental knowledge gleaned over the past 50 years. Although this has been very beneficial, we are using up our "seed corn." Our models cannot progress further without a new influx of experiments. There are many materials design problems, such as finding the best composition for concrete for a specific geographic region and application, that our computational materials science models cannot handle, and will not be able to handle without the input of new fundamental measurements.

The new measurements needed mainly involve knowledge of the basic hydration reactions, thermodynamics, dissolution of cement minerals, and nucleation and growth mechanisms for each hydration product.[10] The heart of the computational materials science of concrete is building cement paste microstructure (and nanostructure) by simultaneously modeling chemistry and the growth of structure, i.e., the formation of microstructure via chemical reaction. Adding sand and gravel, although a complicated composite problem, remains only a composite problem that we generally have learned how to tackle. But, what we do not know about hydration could fill a book. Admittedly, other types of measurements are needed, including controlled fracture and viscoelastic measurements as well as the basic chemistry and physics of degradation. These also will support model growth, but the hydration-related measurements that I discuss below are more fundamental and, therefore, should have priority.

The basic hydration reactions involving portland cement and water are known only in a broad form, and much less is known about reactions involving widely used byproducts, such as fly ash and blast furnace slag. Details of what phases form in solution and the rate-controlling mechanisms and their associated rate constants are not well-known, at least not well enough for sophisticated hydration models, such as HydratiCA,[11] to be able to function as a design tool for new cement-based materials that incorporate industrial byproducts.

(Credit: Z. Qian, E. Schlangen, G. Ye, and E.J. Garboczi, unpublished.)

Figure X. This image shows a simple concrete microstructure, including only the larger aggregates, created using a recent model developed by workers and Delft and the author.

Portland cement is made up of many mineral phases, most of which dissolve in water and most of which contain impurities that can affect the mechanisms and rates of hydration reactions ~~of~~ their own and those of other minerals. The solubility limits of known impurities in cement minerals and hydration products are not known, and accurate models of the activity of solute components at the high ionic strengths commonly found in cement-based binders are not available. For the dissolution of each cement mineral, the rate of pure dissolution in isolation as a function of composition, temperature, and undersaturation needs to be measured. With these data in hand as a reference, the influences of other important solute ions, such as sulfates and aluminates, can be isolated and measured to determine unequivocally the interactions among cement phases.

The nucleation rates for hydration products, as functions of solution composition and temperature, are not known. The actual nucleation mode in the presence of various surfaces needs to be determined. For heterogeneous

nucleation, the strength of adhesion of a product to mineral surfaces and the ability of products to affect further dissolution of covered mineral surfaces are not known.

Finally, the rate of pure hydration product growth in supersaturated solutions, as a function of temperature and supersaturation degree, needs to be measured. The rate of uptake of various solution impurities in hydration products also needs to be determined. Likewise, the effect on the phase growth rate because of impurity uptake is not presently known. These fundamental building blocks will provide the basic thermodynamic and kinetic foundations that HydratiCA and other realistic models need to advance to the next level of materials design.

Prospectus and need for a data repository

The properties listed above can be measured using sophisticated techniques developed in the past 20 years, with some adaptation. But, it will take a lot of work to get these measurements done.

As these data are gathered, once and for all, this information should be put into a cement materials thermokinetic data repository (that NIST would be happy to host on its servers). Validated experimental data as well as data from molecular scale models from all over the world could feed into it. To make effective use of this data repository, the cement and concrete community will be pushed toward better material characterization. This effort needs long-term, sustained, coordinated support. The funding sources will have to be patient. "People love chopping wood. In this activity one immediately sees results," said Einstein. But these measurements are not just chopping wood, because once they are done, they are done!

This data repository will be incredibly useful—but in what ways? Undoubtedly, the new data will enable the further development and successful use of precise, comprehensive models. These models then will be able to accurately

- Predict time-dependent properties;
- Predict the effect of new chemistries (e.g., oil well cements, calcium sulfoaluminate cements);
- Predict the influence of existing chemical admixtures; and
- Inspire new chemical admixtures by running models to show the possible effects of changes in mechanisms.

If new supplementary cementitious materials (such as fly ash, slag, limestone, rice hull ash, expanded clays, waste glass, and recycled concrete) are controlled by the same types of phases and chemistry, then these models also will be able to accurately incorporate the use of these materials. If not, more measurements will need to be made, using techniques that were developed for cement. The same can be said for degradation reactions: If the same type of necessary fundamental measurements are made, they will enable the development of accurate degradation models, thus enabling service life prediction. The latter is a particularly important issue when considering infrastructure renewal decisions.

If we can develop this data repository, the computational materials science of concrete truly can become multiscale and reach the higher level needed to become a paradigm of the ICME and the MGI approach. Industry, then, would be able to engineer concrete for any application and make effective use of a wide range of component materials. For a large concrete construction job, given a range of local materials as well as environmental conditions and structural requirements, these models will show how to select and best use the available materials to achieve job specifications.

Final personal thoughts

I have enjoyed attempting to bring comprehension to my own little part of concrete research, which itself is a very small part of all scientific endeavor. I have received a great deal of help along the way. Let me start my thanks with Einstein by quoting him one last time: "The most incomprehensible thing about the world is that it is comprehensible." Despite his matchless achievements as a physicist (and his remarkable quotability), I will venture to differ with him about incomprehensibility. I believe, "in the beginning, God created the heavens and the earth"—we were created in his image, so that is why the world, also his creation, is, in general, comprehensible to us. I am grateful for this basis of meaning and enjoyment for my pursuit of science.

I would like to especially thank my PhD thesis advisor, Mike Thorpe,

Publicly available modeling programs through NIST

- Virtual Cement and Concrete Testing Laboratory
- Computation of Water-to-Cement Distance Function
  - for computing the water-to-cement distance (proximity) function
- Stokes Three-Dimensional Permeability Solver
  - for the computation of the permeability of any digitized porous microstructure
- Internal Curing with Lightweight Aggregates
  - models and publications concerned with mixture proportioning and other aspects of internal curing
- Concrete Optimization Software Tool (COST)
  - for optimizing concrete mixture proportions based on response surface methodology experimental design principles and statistical analysis
- A Prototype Service Life Prediction Program
  - for service life prediction of steel-reinforced concrete exposed to chloride ions
- CEMHYD3D Computer Program
  - 3D cement hydration and microstructure development modeling
- HCSSMODEL Computer Program
  - 3D concrete microstructure modeling
- Finite element/finite difference programs
  - for computing the linear elastic and linear electric properties of digital images in 2D and 3D
- 4SIGHT Computer Program
  - for concrete performance prediction under various exposure conditions
- CONCLIFE Computer Program
  - for estimating the service life of concrete pavements and bridge decks exposed to sulfate attack and freeze-thaw deterioration
- SULFATE2 Computer Program
  - for computing the resistance of concrete to sulfate attack
- Cement Hydration Modeling
  - for modeling cement hydration and percolation processes

For more information, see: http://ciks.cbt.nist.gov/~bentz/phpct/cmml.html

Garboczi.

and NIST colleagues Jeff Bullard, Dale Bentz, Paul Stutzman, Clarissa Ferraris, Nicos Martys, Ken Snyder, Jack Douglas, Geoff Frohnsdorff, and Jim Clifton. My principal colleagues at ACBM were Suru Shah, Francis Young, Tom Mason, and Hamlin Jennings. ACBM and NIST have played crucial roles in the development of my career and in the development of the computational materials science of concrete.

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Editor's note

This article has been adapted from the 2012 Della Roy Lecture, given June 11, 2012, at the annual meeting of the ACerS Cements Division, University of Texas-Austin.

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