The Past, Present, and Future of Computational Materials Science of Concrete

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Abstract

The computational materials science of concrete has developed rapidly in the last 15 years. This development has been strongly tied to the even more rapid advances in computer processor speed and memory during this same time, without which progress in the computational materials science of complicated materials such as concrete would not have been possible. As the increase in computer power continues unabated, further development in the computational materials science of concrete may be constrained by a lack of trained researchers. In this presentation, we will briefly review the past and present state of this field, and make some projections as to what will happen in the next decade or so, especially if more people become trained and involved.

1. Introduction

Concrete is a complex random composite material. In addition, concrete is also a multi-length scale material. From the nanometer to the millimeter scale, concrete is a different random composite at each length scale [1]. Concrete is not only different at each length scale, it is also an interactive composite, where the amount and properties of one phase (aggregate volume and surface area) affects the properties of other phases (bulk and interfacial transition zone cement paste) [2]. Percolation processes [3-5] and composite interactions [6-8] play key roles in the performance of concrete, and help to explain the overall dependence of transport properties like ionic diffusivity [9] and fluid permeability [10] on the microstructure.

Because of this randomness and complexity, analytical methods for quantitatively relating microstructure and properties of concrete are generally ruled out from the start. That is not to say that analytical equations are not useful, but they can only be used by "smearing" out some aspect of the complex random microstructure. Therefore, they cannot be used to directly relate microstructural details to properties. By default, sophisticated computer models are necessary to describe the microstructure and transport properties.

But having sophisticated computer models is not enough. As will be seen below, the computational materials science of concrete (CMSC) has been intimately wrapped up with the development of computer processing speed and the growth of computer memory over the last 15 years. For further advances to be made, both the algorithm development and the hardware necessary for proper implementation of the algorithm must be available simultaneously. Computer hardware development goes on independently of concrete

research. But the development of models for predicting the microstructure and properties of concrete is dependent on having researchers available to carry out the work.

What we mean by the term "computational materials science of concrete" is this: computer-based models of microstructure at the relevant length scale, operated on by algorithms that give accurate measures of physical properties. The microstructure models ideally do incorporate much of the basic physics and chemistry of the microstructure formation process. However, they do not have to, as long as the eventual microstructure reflects reality. This working definition is similar to the definition of "fundamental models" that we have previously contributed [11]. We should therefore mention that this short paper is not in any sense a comprehensive review of the field, but rather is an informal, personal look at this field in terms of where it has come from, where it is at currently, and where it might be going, with a focus on work we have done ourselves and with others.

Since this is a symposium to honor Francis Young on his retirement, we do want to mention that his encouragement and advice over the last 11 years have been invaluable. At the start of ACBM in the beginning of 1989, CMSC was just in its infancy. The support of ACBM, which was in no small part due to Francis' personal support of the work, played a large role in our part of the development of CMSC.

2. Development of the Computational Materials Science of Concrete

It is good to remember where CMSC came from, before going on to describing what it is now and what it might be in the future. This description of its development is incomplete, but gives the main intellectual sources, at least for us.

From our point of view, there were really four intellectual sources. First came the work on the structure of amorphous semiconductors like silicon and germanium in the 1960's and 1970's (which was the background of one of us, EJG). Here the problem was first really faced of not having a periodic lattice upon which to do calculations of various material properties. Physicists had before developed crystal physics to a high degree, and had even allowed for crystal defects like dislocations. However, the problem of amorphous semiconductors, or of glass, was entirely different. There was no underlying crystal lattice. How was one to do any calculations at all? Analytical approximations were tried, with only a limited degree of success [12,13]. Then models were built, where several hundred atoms (which pushed the computing power back then to the limit) were linked together randomly. Algorithms were applied to these models to compute properties, which then were compared to experiment in an attempt to explain the experimental results. In essence, these models and their associated computations were the beginning of the computational materials science for amorphous materials, at the atomic level.

Second came two developments in the materials science of concrete community, which appeared to be unrelated to the previous amorphous semiconductor work, but which were similar to it. These were both highly original, highly innovative developments. In 1984-5, Wittmann, Roelfstra, and Sadouki published two important papers on numerically simulating the structure and properties of concrete in 2-D [14,15]. In these papers, which anticipate all our work at the concrete level, simple models were

developed for simulating the shape and arrangement of aggregates in concrete. A finite element array was then applied to these models in order to compute properties like thermal conductivity and elastic moduli.

In the very next year, 1986, Jennings and Johnson published work on a three-dimensional (3-D) model of cement paste microstructure development for C_3S pastes [16]. This was the equivalent of the amorphous semiconductor models, but at the cement particle scale, not at the atomic length scale. This effort carried the development of CMSC down to the micrometer scale of cement. Particles of various size, mimicing a cement particle size distribution, were dispersed randomly in 3-D. Various rules were applied to these continuum spherical particles to simulate the dissolution of cement and the growth of hydration products. We have been told that a digitized structure for the model was originally contemplated, but computer power at the time was deemed insufficient [17]. This is an example of the interplay between computer hardware developments and algorithm developments, as was mentioned in the Introduction. The percolation properties of this model were analyzed by Navi et al. [18] in the 1990's. The further development of this model has been impeded because of the difficulty in carrying out the cement hydration process and subsequent calculation of properties using non-digitized continuum particles.

The fourth development, which completed the preliminary steps that led to our part of the development of CMSC over the last 11 years, was a paper showing how a random walk algorithm could be applied to continuum models to compute electrical and diffusive transport in their pore space [19]. We sought to apply this algorithm to Jennings' cement paste model, since it was a continuum model. While learning this random walk algorithm, we experimented with digitizing the microstructure of simple models and then using random walks on the digital lattice. The combination of the ideas of random walks, digital images, and a cement paste hydration microstructure development model led directly to the first NIST cement paste hydration model. Fortunately, at the time of its first development, 1989, we had just enough computer power to barely implement a 3-D model of sufficient size (100^3 pixels). Three-dimensional models are necessary to accurately compute properties of these highly random materials. Two-dimensional models and real 2-D images are generally insufficient because the percolation properties of these systems are quite different in two and three dimensions.

Once the model was on a digital lattice, a suggestion by Thorpe, one of the leaders in the amorphous semiconductor modelling work, led us to the realization that any finite element or finite difference algorithm will work on a digital lattice. Therefore, almost any physical property could be simulated, thus greatly increasing the ability of the cement paste model to be compared to experimental results. With the development of percolation theory [20] and composite theory [21-23] for digital lattices, all the pieces were in place for the further rapid development of CMSC.

3. Present-Day Activity

At present, we would say that CMSC is at a consolidation phase. Models of cement paste and concrete have been made, and many individual properties can be computed and compared to experiment. Much of the current state of affairs can be seen in the

Electronic Monograph developed by us and others, which contains, in HTML book form, a record of most of the developments in CMSC over the last 11 years [24]. As part of this monograph, a "digital tool-kit" has been described, detailing what computational tools are available to operate on computer-based models of random materials [25]. The monograph also describes experimental materials science results obtained at NIST, many of which are strongly coupled to computational results.

There is increasing synergy between the experimental and computational aspects of the materials science of concrete in our group at NIST. This has been true for centuries in physics, but is only now being made use of in understanding the microstructure-property relationships in concrete. For example, the NIST 3-D cement hydration model (now called CEMHYD3D) is only able to accurately model microstructure development and properties because of the extreme care taken in generating representative, in terms of particle size and phase distribution, starting 3-D cement particle images. The basis for these starting images is an image segmentation algorithm applied to real experimental 2-D cement powder images and a correlated image generation algorithm originally developed in the 1970's [26].

Currently, the cement paste microstructure model is being improved and revised almost daily [27,28]. This model, the concrete microstructure model, and many of the application programs have been documented and are freely available to any user [29]. More and more people, including industrial companies, are starting to use the software, at least partly due to the ACBM/NIST Computer Modelling Workshop, which is now in its 11^{th} year. Over 200 people from academia, industry, and government, including almost 100 students, have been introduced to computational materials science via this workshop, and we are now approaching a "critical mass" necessary for much more activity in this area to take place.

New uses have been proposed for the computational materials science tools that have been developed, with most involving combinations of tools to study phenomena that are more complicated than those studied to date. For example, studying the early–age cracking of high performance concrete involves first hydrating the cement paste and computing the self-desiccation in terms of which pores dry out first. Then elastic and viscoelastic computations must be made for stresses, allowing for capillary tension in the pore water and possible changes in the surface energy of the pore water as ions are dissolved in it. These stresses can in turn cause cracking, which can then alter the pore structure and thus alter the self-desiccation. A different combination of tools has been used to simulate and study the drying shrinkage of porous Vycor glass [30]. Other groups are also carrying on CMSC activities, mention of which are beyond the scope of this short paper.

4. Future Trends

To discuss the future of CMSC is an exercise in speculation. The most we can do is to extrapolate some trend lines, and state where the field seems to be going and what the field needs to be doing. Whether the field ever gets there, or gets somewhere else altogether different, depends on future trends in computing power, available researchers, funding opportunities, and interest by industry. At present, there is still only a limited

number of researchers in this field. However, this situation seems to be changing, as this field becomes more accepted, both by academic and by industrial users, and as computer power becomes more easily available. NIST is also dedicated to making computational tools accessible on the Internet as they become available and as we have manpower to document them.

One item that should remain fairly constant is the rough divide that exists in the field of CMSC between those researchers who think in terms of "modelling" and those who think in terms of "theory." These terms are overly broad, but they are meant to point out the difference between those who wish to *replace* experiments with modelling and those who wish to *explain* experiments with computer-based theory.

We think that the primary function of CMSC is to explain and suggest experiments, much like the use of theory in physics. As was stated above, the relationships between microstructure and properties for concrete are too complex for analytical methods to be very useful. Computational materials science provides the theory that is needed to understand current experiments and help plan new experiments. However, once the validity of a computer model is unequivocally demonstrated, one can then progress to replacing some experiments with models.

Those who want to replace experiments with modelling wish to save time and money by replacing expensive experiments with cheap (once the cost of development is past!) computer simulations. If the materials science of some aspect of concrete is well-understood, and the models developed give results that have been well-validated by careful experiments, then at least some of the experimental work involved can indeed be replaced by computer modelling. At NIST, we are in the process of developing an industrial consortium called the Virtual Cement and Concrete Testing Laboratory [31], which is intended to replace many standard tests on cement and concrete with well-validated computer models. At least some of the models developed so far in CMSC are at this stage. This will hopefully speed up the R&D process to develop new cement-based materials. We invite cement and concrete companies to join with us in this development. The future of this activity will also depend on advances in computer power, both at the research end, so that models can do more, and also at the user end, so that the average user can take better advantage of the ability of models to replace experiments.

As for the future of CMSC in explaining experiments, this will depend on both computer processing speed and memory improvements, although perhaps more on processing speed improvement. The memories available on the large shared-memory clusters (32 or more processors, 1 gigabyte per processor) are large enough to do many computations that were impossible in the past. In general, however, doing large computations, involving 10 or more gigabytes, is still too slow. Parallel processing will help, but often many processors are not available at one time. Also, the current models that are used the most – cement hydration and finite element electric and elastic codes – have not been optimized for parallel processing. They can be, but this has not been done to date. Intrinsic processor speed must also increase greatly to allow many problems to be solved.

The following is a partial list of the problems that will challenge CMSC in the next decade or so.

- The viscoelasticity of concrete is a difficult problem, but very important for understanding real concrete mechanical properties, both at early and later ages. Regular linear viscoelasticity would be hard enough, but concrete is a non-linear, aging viscoelastic composite. The aging is due both to changes in the amount of constituents over time due to hydration, and changes within the phases themselves (aging of C-S-H). Coupling moisture flow to viscoelasticity is also a difficult and important problem. The correct modelling of viscoelasticity of course affects all other properties and processes that involve mechanical stresses.

- Simulating the rheology of concrete suspensions will also become an active area, making use of modern developments in computational fluid dynamics. Among the parameters that need to be studied are the effect of the size distribution and shape of the aggregates, and the effect of chemical admixtures.

- Developing a predictive model for the nanostructure of C-S-H will be a focus of much effort in the next decade. Structural information at the atomic level from NMR and molecular dynamic modelling [32,33] will need to be coupled in to achieve a truly predictive model of C-S-H. This will need also to be able to understand and predict the change of structure and properties of C-S-H with time.

- The hydration of blended cements and cements with mineral admixtures, along with the effect on properties, is an important problem that will definitely be worked on computationally over the next decade. We believe that the future of the cement and concrete industry is closely tied to the future of blended cements.

- The challenge of correctly modelling transport properties like chloride diffusivity, fluid permeability, and sorptivity, especially in unsaturated concrete, will not go away for some years to come. Here is a case where better experiments, which are beginning to be made, drive the development of better (and more complicated) models.

- Nearly all performance properties of concrete change as chemical and physical degradation processes take place. Computational materials science, in conjunction with careful experiments, should be able to analyze and predict these changes. This will be crucial for a proper understanding of the service life of concrete, which is determined by the coupling of properties to environment factors.

- The simulation of the degradation processes occurring in concrete, with application to the prediction of service life, is a case where models *should* replace experiments, in order to compress 50-100 years of service life into one computer run. Even longer times are necessary for concrete used for the long-term containment of nuclear and hazardous waste. A special challenge is the growth of cracks and deterioration of mechanical properties during the degradation.

- The prediction of strength, either tensile or compressive, which involves the application of fracture mechanics to crack growth processes, is a challenge that will occupy CMSC for some time to come.

• Finally, temperature affects hydration mechanisms, microstructure, and degradation processes. A successful CMSC must be able to take the role of temperature quantitatively into account.

All of these areas will demand new algorithmic developments, as well as increased hardware speed and capacity. People who combine skills in basic materials science and computer programming will be essential in new algorithmic development.

We want to re-emphasize the important need for close cooperation between experimental and computational materials science. This is how important advances will be made in concrete science and technology in the future. One example of what can be accomplished via this kind of cooperation is in the work on the electrical properties of plain and fiber-reinforced cement paste and mortar, and the effect of electrode shape and roughness, which has been carried out collaboratively between Northwestern University and NIST [34-46]. Close cooperation means experimentalists designing experiments to make the job of modelling easier, and modellers forcing themselves to model real systems, not just simple systems. In this way, the models can help explain the experiments, and the experiments can show the shortcomings of the model.

In spite of the many problems that remain to be solved, which will keep this field scientifically fruitful for years to come, we also expect to see important *applications* of CMSC to concrete practice within the next decade. We invite those interested in this important field to collaborate with us to advance CMSC.

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