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MULTIOBJECTIVE OPTIMIZATION OF CEMENT PASTE PERFORMANCE

V. Šmilauer[*], M. Lepš[**], Z. Vitingerová[***]

Summary: *Resulting properties of hardening cement paste are influenced by several stochastic parameters whose effect has not been yet fully determined. We attempt to explore the sensitivity of parameters on the hydration heat and Young's modulus covering pastes made from ordinary Portland cement. Virtual modeling is based on CEMHYD3D model, quantifying the effect of cement fineness, its chemical composition, gypsum content, w/c, and curing conditions. Gypsum content, autocorrelation of clinkers, C4AF and saturated/sealed curing conditions were found to be insensitive in terms of released heat or Young's modulus. W/c is the only governing factor for the Young's modulus at the above-mentioned degrees of hydration or times. Possible application of virtual modeling is demonstrated on multiobjective optimization of cement paste properties. The goal is to minimize hydration heat while maximizing Young's modulus. In this particular case, the Pareto set clearly shows the importance of w/c parameter.*

1. Introduction

Concrete, perceived as a multiscale material, inherits several properties from the cement paste. The level of cement paste plays important role in the design of tailored material since its composition and time evolution can be controlled, monitored, and influenced. Higher level of mortar or concrete is relevant rather for aggregates, being usually not the weakest element in mechanical performance or durability.

The optimization of concrete properties has been a long time the domain of experiments. Statistical mixture design methods may provide certain guidance to select extreme combinations and to interpolate among experimental results (Simon, Lagergreen & Snyder, (1997)). Virtual modeling is another approach to complement experimental point of view; it is a promising tool not only for a design itself but for the verification of certain assumptions in a numerical way.

The objectives of this paper include stochastic sensitivity analysis between input parameters of cement paste and its response in terms of hydration heat and Young's modulus.

[*] Ing. Vít Šmilauer, Ph.D.: Department of Mechanics, Faculty of Civil Engineering, CTU in Prague, Thákurova 7; 166 29 Praha 6; tel: +420 224 354 369, fax: +420 224 310775; e-mail: smilauer@cml.fsv.cvut.cz

[**] Ing. Matěj Lepš, Ph.D.: Department of Mechanics, Faculty of Civil Engineering, CTU in Prague, Thákurova 7; 166 29 Praha 6; tel: +420 224 354 369, fax: +420 224 310775; e-mail: leps@cml.fsv.cvut.cz

[***] Ing. Zuzana Vitingerová: Department of Mechanics, Faculty of Civil Engineering, CTU in Prague, Thákurova 7; 166 29 Praha 6; tel: +420 224 355 417, fax: +420 224 310775; e-mail: zuzanka@cml.fsv.cvut.cz

Such approach is hardly to be achieved experimentally due to the hundreds of evaluations which are time-consuming, or exhibit statistical nature of cement paste and testing device. The reproducibility in virtual tests is guaranteed although the link microstructure-property does not have to exist or can be even wrong. Therefore virtual modeling is exploited particularly for well established and validated simulations, i.e. for the heat evolution and Young's modulus of cement paste.

CEMHYD3D serves as a microstructural model for the simulation of cement paste at the resolution of 1 μm (Bentz, (2005)). Standard input values were used and the model capabilities were extended with the prediction of C-S-H_{LD} and C-S-H_{HD} to include additional mechanical stiffening at confined capillary space (Šmilauer & Bittnar, (2006)). Parameter β = 0.0003 was used throughout for the mapping of CEMHYD3D cycles to a real hydration time. Microstructure size of 50 x 50 x 50 μm was assumed to be representative up to approximately 28 hydration days. The drawback of using such a small size is a limited maximum grain diameter to 31 μm. The consequence is a slight overestimation of hydration degree at later stages especially in coarse cements. All simulations took place at isothermal 20 ^{o}C.

The selection of input parameters is chosen in such a way that CEMHYD3D is able to take them realistically into account in the simulations. Their ranges correspond approximately to contemporary limiting values of Portland cements used in civil engineering, Tab. 1.

Table 1: Input parameters with limits used in virtual tests

Parameter	*Minimum*	*Maximum*
Fineness [m^2/kg]	200	600
Gypsum content [vol. of cement]	0	0.1
Autocorrelation file designation	0	9
C_3S [cement mass fraction]	0.4	0.8
C_2S [cement mass fraction]	0	0.35
C_3A [cement mass fraction]	0	0.15
C_4AF [cement mass fraction]	0	0.15
W/c	0.2	0.6
Curing condition regime	0-saturated	1-sealed

The particle size distribution (PSD) is expressed by the value of Blaine fineness [m^2/kg]. Rosin-Rammler cumulative function is fitted to PSD data (Garboczi, Bullard & Bentz, (2004)) with x [μm] being the particle diameter

$$G(x) = 1 - \exp(-bx^n) \quad (1)$$

$$n = -0.00083333\ fineness + 1.1175 \quad (2)$$

$$b = 0.000754\ fineness - 0.143 \quad (3)$$

Before the simulation of hydration, spherical cement grains have to be thrown into the computational volume. Since Portland cement is composed basically from four clinker minerals, each grain has to be consequently divided into silicates, aluminates and later to individual clinker minerals, relying on a triplet of autocorrelation functions (Garboczi, Bullard & Bentz, (2004)). Ten triplets obtained from CCRL cements were taken from the NIST cement database to explore their effect (Garboczi, Bullard & Bentz, (2004)).

Saturated or sealed curing conditions result in a different morphology of cement paste. While the saturation ensures enough water to percolated capillary space, sealed conditions slow down hydration process due to the lack of water, implemented as emptying of larger capillary pores with an influence on reactions.

Typically, two hundred input datasets were randomly generated using LHS method which guaranteed low mutual correlation among input parameters and covered the range of feasible input combinations. Since four clinker minerals have to form a unity, weak correlation among them was reported.

2. Role of parameters influencing the heat of hydration

Bentz, Waller & de Larrard, (1998) used CEMHYD3D in the simulation of temperature rise under adiabatic conditions. It is well established that heat of hydration origins from individual chemical reactions in which the kinetics of clinker minerals determine heat release rates. The effect of input parameters on the overall heat is examined either in terms of the degree of hydration (DoH) or elapsed hydration time, Fig.1.

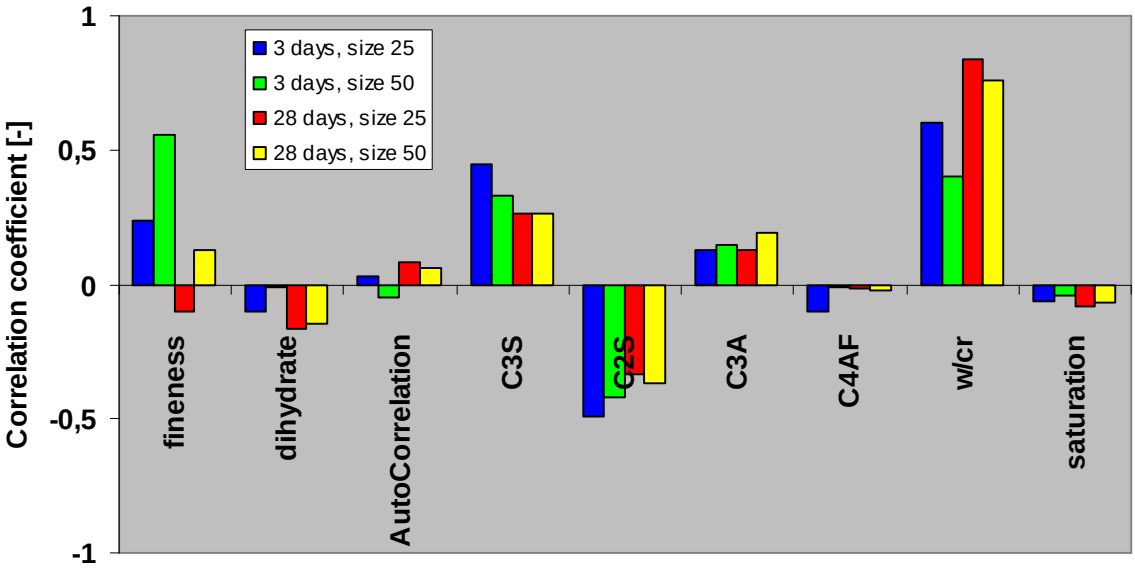

Figure 1 : Correlation between input parameters and hydration heat at two degrees of hydration and two hydration times

Regardless on the above-mentioned evaluation criteria, there are negligible effects in the amount of gypsum, type of autocorrelation file, C_4AF content, and saturated/sealed curing conditions on released heat. Decreasing C_3A content and increasing C_2S amount have the only significant impact when microstructures are hydrated to the same hydration degrees but not times.

When the released heat is quantified at 3 and 28 days of hydration, the effect of input parameters is more complex. Released heat increases due to higher fineness, especially at early ages, since higher surface area of cement grains accelerates the overall kinetics. The effect of C_3S and C_3A content is about the same, which seems to be contradictory to experiments, where Portland cements with higher C_3A are known to release more heat at early

ages. It must be born in mind that increasing the C_3A amount itself results in the decrease of the other clinkers and that C_3S content is always more dominant over C_3A in Portland cements. Therefore there are two contradicting mechanisms which defeat each other, but the mechanism is uncovered in the comparison against achieved hydration degree. An increase in w/c has an impact on higher released heat since hydration is not slowed down by limited capillary space. This effect is more significant at 28 days.

3. Role of parameters influencing Young's modulus

Determination of Young's modulus is based on numerical linear elastic homogenization, utilizing fast Fourier transform (FFT) (Moulinec & Suquet, (1994)). Periodic microstructure 50 x 50 x 50 μm from CEMHYD3D model is filtered through solid percolation routine (Šmilauer & Bittnar, (2006)) and sampled to a grid of 50 x 50 x 50 Fourier points. The filtering has an affect up to approximately degree of hydration of 0.3 for the range of input parameters in Tab. 1. The average time consumption for FFT-based homogenization is around 10 minutes on 3.2 GHz CPU. Intrinsic elastic properties are taken from (Šmilauer & Bittnar, (2006)), including water filled and empty capillary porosity, C-S-H_{LD} and C-S-H_{HD}.

The stochastic statistical sensitivity between input parameters and Young's modulus is depicted in Fig. 2. W/c ratio is the only governing parameter, followed by cement fineness. Obviously, decreasing w/c reduces the capillary porosity while increasing the modulus. More homogeneous composites, having the same volume fractions, are known to exhibit higher stiffness which can be interpreted as increasing cement fineness where smaller grains cause more uniformly distributed phases.

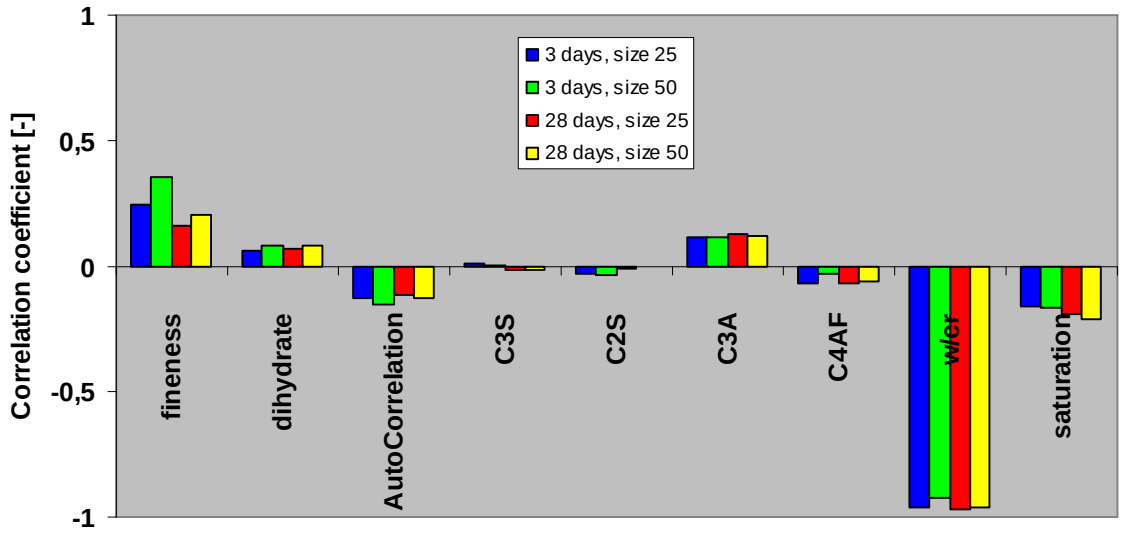

Figure 2 : Correlation between input parameters and Young's modulus at two degrees of hydration and two hydration times

W/c is the most influencing input parameter for the Young modulus so the results may be plotted against w/c only. The situation is depicted in Fig. 3. Over 250 sets of different microstructures were generated, let to hydrate, filtered with solid percolation routine and homogenized by FFT-based method. Only 50 microstructures were selected for the degree of hydration of 0.8.

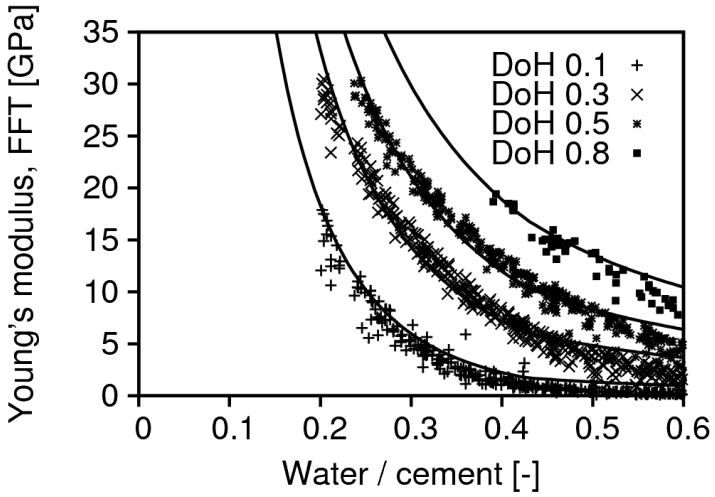

Figure 3: Relationship between Young's modulus and w/c ratio and the fit to a power function

The evolution of Young's modulus in Fig. 3 can be approximated with a power function. The function should match the asymptotes and take into account percolation threshold (setting) at low degrees of hydration. The solid percolation threshold, $DOH_{p,}$ was found to be the function of w/c (Šmilauer, (2007)). The evolution of Young's modulus against degree of hydration, DoH, should be concave for all cases. The following formulae, valid in the range $0.2 \leq w/c \leq 0.6$, were obtained by nonlinear fitting to equally-weighted points from Fig. 3

$$\text{Young's modulus [GPa]} = a(DoH - DoH_p)^b, \text{ greater than 0} \quad (4)$$

$$DoH_p = 0.0485\ w/c \quad (5)$$

$$a = (0.15\ w/c^{1.39})^{-1} \quad (6)$$

$$b = 2.15\ w/c + 0.08,\ b \leq 1 \quad (7)$$

The coefficient *a* expresses the asymptotic Young's modulus, approximately for complete hydration (in reality complete hydration does not occur below w/c = 0.36 (Hansen, (1986))). For example, at w/c = 0.2 *a* = 62.4 GPa and at w/c = 0.6 *a* = 13.56 GPa. The coefficient *b* quantifies the concavity which is higher in lower w/c, i.e. the Young's modulus grows faster in lower DoH regardless on *a*. Coefficient *b* must be less or equal than 1. The maximum Young's modulus that can be attained in Portland cement paste is approximately 50 GPa for w/c=0.2. In this particular case, the maximum degree of hydration can reach 0.55 according to Hansen, (1986). Unhydrated clinker minerals boost the stiffness of hydrated cement paste considerably, more than the amount of formed C-S-H. The coefficient of correlation between Eq. (4) and the data in Fig. 3 is 0.986.

4. Multi-objective optimization

A majority of realistic optimization problems do not rely on one objective function only but require simultaneous optimization of several objective functions. For example, one objective function can be merged with the amount of released heat at a given time and an optimal set of input parameters can be found. On the other hand, such combination can be detrimental to

other properties, such as stiffness, shrinkage, or crack formation. Several contradicting goals are typical in multi-objective optimization and a compromise has to be found to satisfy partially all of them.

The scalar concept of optimality has to be replaced with Pareto optimality in the multi-objective optimization. Pareto optimal solution presents a set, for which no better set can be found while minimizing (or maximizing) *all* of the objective functions. The goal is to find such a set of input parameters which maximize the Young modulus while minimizing hydration heat at the time of 28 days of hydration.

Multi-objective optimization was based on an evolutionary algorithm called Pareto Archived Evolution Strategy (PAES) (Knowles & Corne, (2000)) with Global parallel model (Cantú-Paz, (2001)). Each individual in optimization represents a unique set of input parameters. The PAES algorithm chooses one individual from the population and applies mutation. The offspring is evaluated for dominancy and is rejected or given back into the population archive. The situation after 150 cycles is depicted on twelve Pareto points in Fig. 4 (left). The set presents some feasible combinations with maximum Young's moduli and minimum hydration heat at 28 days for input data from Tab. 1.

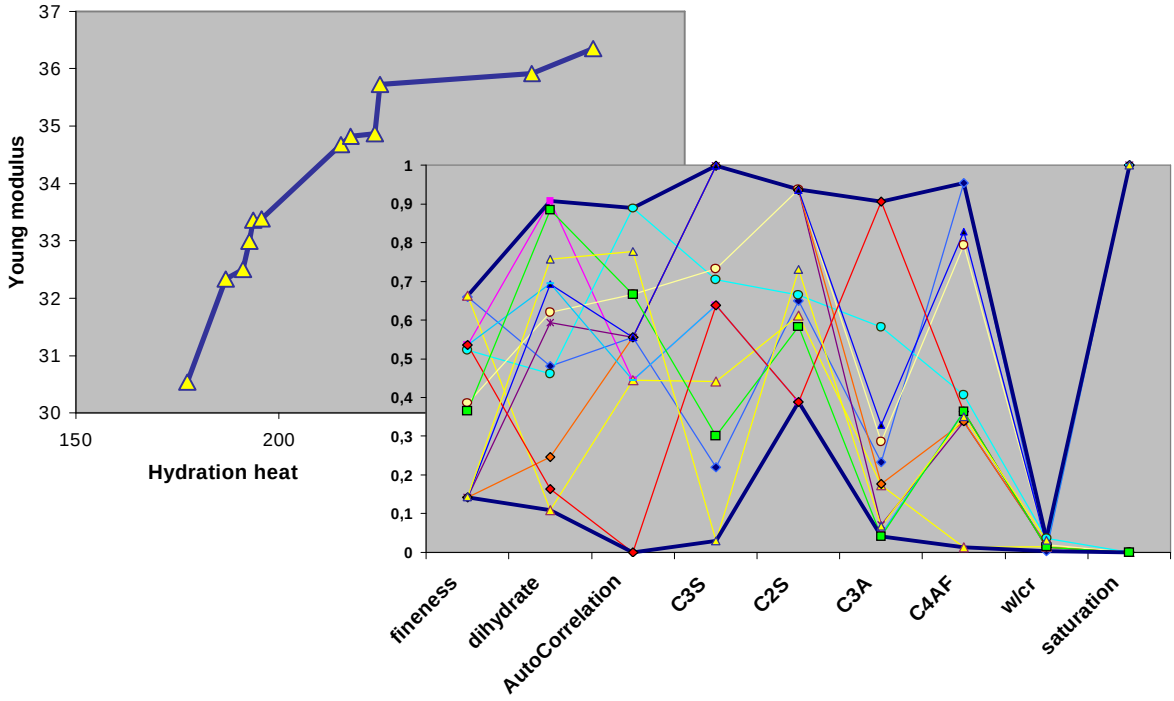

Figure 4: Optimal Pareto front (left) and corresponding input parameters (right)

The bounds of hydration heat were found in the range 177 – 277 J/(g of cement) with Young's moduli between 30.54 and 36.35 GPa. Corresponding feasible sets of input parameters are plotted in Fig. 4 (right) in relative measures. The optimization results point to rather low cement fineness, lower C3S amount, and higher C2S content. All w/c's tend to fall at the minimum of 0.2. A scatter in the majority of input parameters allows imposing other restrictions, not considered in this particular multi-objective optimization. The results can be used further in the design of experiment to include certain input combinations easily overlooked in such complex systems as cement paste.

5. Conclusions

The stochastic sensitivity of cement input parameters was explored on released heat and Young's modulus. While the effect of the individual input parameters on hydration heat is not straightforward, the w/c is the only governing parameter for the evolution of Young's modulus at 3 or 28 days or degrees of hydration of 0.3 or 0.5. Young's moduli were fitted to analytical formula, depending only on w/c and degree of hydration.

Multi-objective optimization showed feasible combinations for maximizing Young's modulus while minimizing released heat. The input parameters demonstrated the need for low w/c close to 0.2. The criteria for multi-objective functions can be set up arbitrarily, enabling the design of tailored materials prior to experimental verification.

Acknowledgement

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