UDC 691.542

A HOMOGENIZATION METHOD FOR STIFFNESS CHARACTERISTICS OF CEMENT PASTE UNDER VISCOELASTIC BEHAVIOR

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Abstract

The microstructure of cement paste is extremely complex and heterogeneous, consists of randomly distributed phases with an arbitrary geometry, formed during the hydration process. The key phase of cement paste – calcium silicate hydrate exhibits distinct viscoelastic behavior causing creep in cement-based composites. These reasons make the problem of evaluating effective stiffness characteristics rather difficult, since stress-strain relationships under viscoelastic behavior are usually described using the principle of aging-time superposition, represented in the form of the Stiltes integral, which has not an analytical solution.

Existing approaches to solving this problem involve two principles: the Laplace–Carson transform and the effective medium theory. This makes possible to find a solution for the evaluate effective stiffness characteristics under viscoelastic behavior, but only for a limited geometric shape of inclusions in the form of an ellipsoid and its related shapes. However, such shapes are not fully matching the real geometric shape of most phases of cement paste, especially for capillary porosity.

The paper presents one more approach to solving the problem of effective stiffness characteristics of cement paste based on a FEA homogenization facilitates to evaluate effective stiffness properties for an arbitrary phase geometry, through introducing into the variational formulation the numerical inversion of the Stieltjes integral describing its viscoelastic behaviour. In addition, this approach best implements the solidification mechanism for the history of the aging stress-strain relation during the hydration process.

Keywords: cement paste, viscoelasticity, homogenization, solidification theory, FEA.

ПРИНЦИП ГОМОГЕНИЗАЦИИ ЖЕСТКОСТНЫХ ХАРАКТЕРИСТИК ЦЕМЕНТНОГО КАМНЯ ПРИ ВЯЗКОУПРУГОМ ПОВЕДЕНИИ

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Реферат

Микроструктура цементного камня чрезвычайно сложна и неоднородна, состоит из хаотично распределенных фаз с произвольной геометрией, образуемых в процессе гидратации. При этом следует учитывать, что основная фаза цементного камня – гидросиликат кальция – проявляет ярко выраженное вязкоупругое поведение, обуславливая возникновение ползучести в цементных композитах. Эти причины делают задачу оценки его эффективных жесткостных характеристик достаточно сложной, поскольку напряженно-деформированное состояние в условиях вязкоупругого поведения принято рассматривать с позиций теории нелинейной наследственности, представляемой в виде интеграла Стилтьеса, не имеющего аналитического решения.

Существующие подходы к решению обозначенной проблемы сочетают два принципа: преобразование Лапласа – Карсона и положения теории эффективной среды, что позволяет находить решение задачи эффективных свойств композитов при вязкоупругом поведении. Однако поскольку получаемые решения в рамках такого подхода основаны на положениях теории эффективной среды, это приводит к достаточно существенному ограничению, накладываемому на геометрическую форму фаз композита, которые могут быть представлены только в виде эллипсоида и его производных форм, что не совсем соответствует реальной геометрической форме большинства фаз цементного камня, в особенности капиллярной пористости.

В статье представлен еще один подход к решению задачи эффективных жесткостных характеристик цементного камня при вязкоупругом поведении, основанный на положениях гомогенизации методом конечных элементов, позволяющей оценивать эффективные жесткостные характеристики композитов с произвольной геометрической формой фаз, в вариационную формулировку которого вводится численное обращение интеграла Стилтьеса, описывающего вязкоупругое поведение цементного камня. Кроме того, этот подход наилучшим образом реализует положения теории солидификации при формировании истории напряженно-деформированного состояния в период гидратации.

Ключевые слова: цементный камень, гомогенизация, вязкоупругость, теория солидификации, МКЭ.

Introduction

Cement paste[1] is a crucial phase of cement-based composites which in many respects determines their mechanical behavior at an early age. It is a composite consisting of a solid phase (hydration products and unhydrated cement), a liquid phase (water), and a gas phase (air) with a complex and heterogeneous structure formed during the hydration process.

One of the key features of cement paste is that the solid phase has the distinctly viscoelastic behavior which originates in the calcium silicate hydrates, and causes creep in cement-based composites [1].

There are many prediction models for evaluating the effective stiffness characteristics of cement paste based on the principles of multiscale modeling, and dealing with the following techniques of homogenization:

1. Analytical homogenization, including two class of effective theories: effective medium theory and differential effective medium theory [2].
2. Numerical homogenization based, including two methods based on: finite element analysis (FEA) [3] and the Fourier transform [4].

The obvious drawback of these models is that they consider cement paste through linear elastic behaviour. At the same, there are only a few models taking into consideration viscoelastic behavior of cement paste. The models [5, 6] use the Laplace–Carson transform which converts non-ageing linear viscoelastic behavior into linear elastic one, allowing to directly apply the analytical homogenization schemes. Then, the numerical inversion of the Laplace–Carson transform is used for the aging vis-

[1] Here the term «cement paste» refers to the hardened cement paste.

https://doi.org/10.36773/1818-1112-2024-135-3-38-42

coelastic solution. The model [7] uses a principle based on Volterra integral operators, allowing to directly apply the Mori-Tanaka scheme for the aging viscoelastic solution.

Despite the fact that these models allow for the possibility of linear viscoelastic behavior, they still have restrictions imposed by the analytical homogenization schemes related to the geometric shape of the inclusions, assuming to be spherical in most cases. This considerable assumption may decrease accuracy of predicting the effective stiffness characteristics, since it is a well-known fact that the shape of most hydration products is not spherical. This is especially so for capillary porosity with a complex morphology, including various geometric shapes with irregular spatial distribution.

This paper presents approach based on the FEA homogenization, which is not sensitive to a geometric shape of inclusions, through introducing into the variational formulation the numerical inversion of the following Stieltjes integral, expressing the principle of superposition [8]:

$$\boldsymbol{\varepsilon}(t) = \int_0^t \mathbb{J}(t,t')\ d\boldsymbol{\sigma}(t'), \quad (1)$$

where $\boldsymbol{\varepsilon}(t)$ – is the second-order microscopic strain tensor at time t representing the age of a cement-based composite;

$\mathbb{J}(t,t')$ – is the fourth-order compliance tensor representing the strain at time t caused by a stress that has been acting since time t';

$\boldsymbol{\sigma}(t')$ – is the second-order microscopic stress tensor at time t'.

The time-dependent compliance tensor contains a time-dependent elastic part and a time-dependent viscous part [8]:

$$\mathbb{J}(t,t') = \mathbb{C}(t')^{-1} + \mathbb{J}_v(t,t'), \quad (2)$$

where $\mathbb{C}$ – is the fourth-order elasticity tensor;

$\mathbb{J}_v$ – is the fourth-order viscoelastic compliance tensor.

The principle of superposition relates the stress and strain histories states that the response to a sum of two stress (or strain) histories is the sum of the responses to each of them taken separately [8]. For the microstructural development of cement paste caused by a hydration process, the stress and strain histories can be expressed throw the solidification theory that assumes that fictitious clusters of cement paste are gradually added to the existing ones. Cement paste is considered as a set of all formed clusters (see Figure 1). Since clusters are formed at different ages, history variables for different clusters are treated as mutually independent variables [9, 10].

The presented approach based on the FEA homogenization implements the principles of the solidification theory to express strain and stress history.

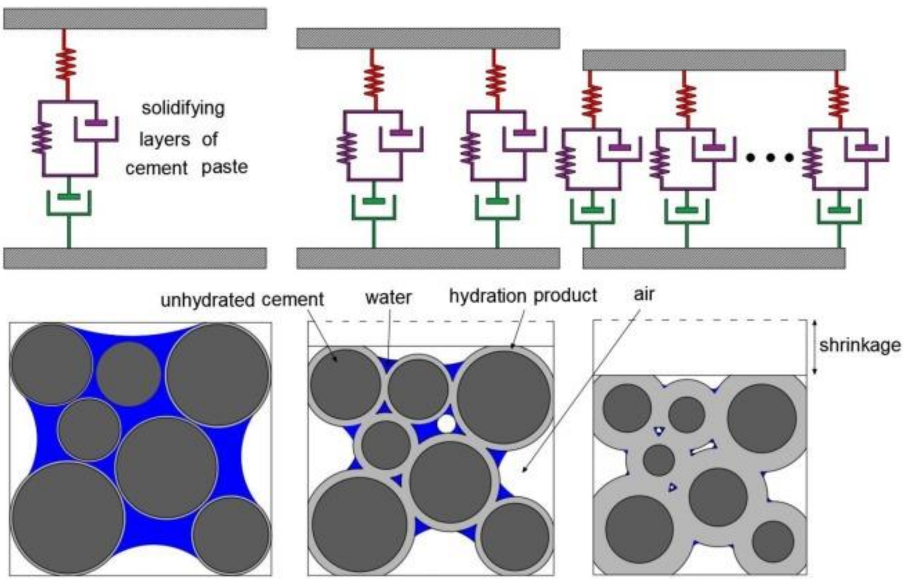

Figure 1 – Schematic representation of solidifying clusters of cement paste, according to [10]

Basic assumptions and principles

1. The microstructure of cement paste is considered at two scales:

– Level 1: Unhydrated cement consists of the C_3S[1], C_2S, C_3A, and C_4AF minerals, and hydration products consists of the CSH, CH, $C_6A\bar{S}_3H_{32}$, $C_4A\bar{S}H_{12}$, C_3AH_6, and FH_3 compounds.

– Level 2: Cement paste consists of homogenous unhydrated cement, a homogenous solid of hydration products, and porosity, including a liquid phase (water) and a gas phase (air).

2. The hydration-induced evolution of the volume fractions of clinker, hydrates, and pores is quantified based on the stoichiometry of hydration reactions of clinker phases. The hydration reactions of Portland cement were taken from the model of Tennis and Jennings [11].

3. Hydration of clinker phases is modeled through the kinetics model of Parrot and Killoh [12].

4. The morphology of cement particles is modeled as spherical, and the hydration products as prolate or oblate spheroidal that is isotropically oriented in the representative element volume (REV).

5. The effective elastic tensor of homogenous unhydrated cement ($\mathbb{C}_{uc}$) and a homogenous solid of hydration products ($\mathbb{C}_{hp}$) are estimated using the Self-Consistent scheme [2].

6. The REV of cement paste is a voxel-based grid considered no more 50×50×50 μm in size with a resolution of 1 μm^3/voxel.

7. The voxel-based microstructural model [13] is used for the spatial distribution the phases of cement paste over the REV during the hydration process.

8. Continuous hydration time is discretized into n time intervals $\Delta t_i = t_i - t_{i-1}$, $i = \overline{1,n}$.

9. A hexahedral mesh is generated based on the voxel-based REV, and consists of the following finite elements:

– for strain field: a trilinear 8 nodes hexahedron with 3 degree-of-freedom (DOF) per node;

– for stress field: a trilinear 8 nodes hexahedron with 1 DOF per node.

10. The hexahedral mesh is divided into four subdomains related to the phases of cement paste. Each finite element has the same elasticity tensor within a subdomain.

11. Periodic boundary conditions are implemented to approximate the REV as an infinite system with a structural periodicity.

[1] The cement chemist notation is used.

12. Since, only the CSH phase has viscoelastic behavior, the modified model from [14] is used for the compliance tensor:

$$\mathbb{J}(t,t') = \mathbb{C}_{hp}(t')^{-1} + \mathbb{J}_{CSH}(t,t') = $$
$$= \mathbb{C}_{hp}(t')^{-1} + \mathbb{B}\frac{1}{C_v(1-\nu^2)} ln\left(1+\frac{t-t'}{\tau}\right), \quad (3)$$

where C_v – is the contact creep module calculated by [14];

$\mathbb{B}$ – is the fourth-order tensor for transformation into three-dimension stress-strain relationship, which depends on the elastic properties of the CSH phase [8];

ν – is the Poisson's ratio of the CSH phase;

τ – is the characteristic viscous time, 1,66 s [14].

13. The numerical inversion of the integral (1) is used according to the trapezoidal rule [8]:

$$\boldsymbol{\varepsilon}_i = \sum_{j=1}^{i}\left[\frac{1}{2}\left(\mathbb{J}(t_i,t_j) + \mathbb{J}(t_i,t_{j-1})\right) : \Delta\boldsymbol{\sigma}_j\right] = \sum_{j=1}^{i}\mathbb{J}(t_i,t_{j-1/2}) : \Delta\boldsymbol{\sigma}_j \text{ for } i \geq 1, \quad (4)$$

where $\boldsymbol{\varepsilon}_i$ – is the second-order stress tensor of i-th time interval;

t_i, t_j – is time related to the end of i-th and j-th time interval, respectively;

$t_{j-1/2}$ – is time related to the middle of j-th time interval;

$\Delta\boldsymbol{\sigma}_j$ – is the incremental of the second-order stress tensor of j-th time interval;

«:» – is the double dot product.

Then, the difference of the second-order stress tensor at i-th time interval ($\Delta\boldsymbol{\varepsilon}_i$) can be expressed by [8, 15]:

$$\Delta\boldsymbol{\varepsilon}_i = \mathbb{J}(t_i,t_{i-1/2}) : \Delta\boldsymbol{\sigma}_i + \sum_{j=1}^{i-1}\left[\left(\mathbb{J}(t_i,t_{j-1/2}) - \mathbb{J}(t_{i-1},t_{j-1/2})\right) : \Delta\boldsymbol{\sigma}_j\right] \text{ for } i > j. \quad (5)$$

Consequently, the stress tensor at i-th time interval is calculated by:

$$\boldsymbol{\sigma}_i = \boldsymbol{\sigma}_{i-1} + \Delta\boldsymbol{\sigma}_i . \quad (6)$$

14. The relation between the macroscopic stress and strain tensors is used is used to calculate the effective constitutive fourth-order tensor $\mathbb{C}_{eff}$ [11]:

$$\langle\boldsymbol{\sigma}\rangle_V = \mathbb{C}_{eff} : \langle\boldsymbol{\varepsilon}\rangle_V, \quad (7)$$

where $\langle * \rangle_V$ – is the average of a field $\boldsymbol{f}$ over the REV, $\langle\boldsymbol{f}\rangle_V = \frac{1}{V}\int_V \boldsymbol{f}(\boldsymbol{x})dV$.

Variational formulation

Following continuum micromechanics, the composite microstructure can be considered as a REV composed of homogeneous phases, and subjected to a macroscopic strain field ($\bar{\boldsymbol{\varepsilon}}$) prescribed at its boundaries. The local strain field of an arbitrary point in the REV $\boldsymbol{\varepsilon}(\boldsymbol{x})$ can be split into $\bar{\boldsymbol{\varepsilon}}$ and a periodic fluctuation strain $\tilde{\boldsymbol{\varepsilon}}(\tilde{\boldsymbol{u}}(\boldsymbol{x}))$, which accounts for the presence of heterogeneities [16].

Then, using the principle of virtual work and introducing an additional vectoral Lagrange multiplier as an additional unknown to make the average of $\tilde{\boldsymbol{\varepsilon}}$ over the REV is vanish, the following variational formulation gives [17]:

Find $(\tilde{\boldsymbol{u}},\boldsymbol{\lambda}) \in V$ such that:

$$\int_V \boldsymbol{\sigma}(\tilde{\boldsymbol{u}}(\boldsymbol{x})) : \tilde{\boldsymbol{\varepsilon}}(\tilde{\boldsymbol{v}})dV + \int_V \boldsymbol{\lambda}\cdot\tilde{\boldsymbol{v}}dV + \int_V \boldsymbol{\theta}\cdot\tilde{\boldsymbol{u}}\,dV = 0 \;\forall(\tilde{\boldsymbol{v}},\boldsymbol{\theta}) \in V, \quad (8)$$

where $\tilde{\boldsymbol{u}}$ – is the trial displacement function;

$\tilde{\boldsymbol{v}}$ – is the test displacement function;

$\boldsymbol{\lambda}$ – is the vectoral Lagrange multiplier;

$\boldsymbol{x}$ – is the position of an arbitrary point in the REV;

V– is the REV;

«·» – is the dot product.

And:

$$\boldsymbol{\sigma}(\tilde{\boldsymbol{u}}(\boldsymbol{x})) = \begin{cases} (\bar{\boldsymbol{\varepsilon}}_i + \tilde{\boldsymbol{\varepsilon}}(\tilde{\boldsymbol{u}}_i)) : \mathbb{C}_{uc} \;\forall\boldsymbol{x} \in V_{uc} \\ \boldsymbol{\sigma}_{i-1} + \Delta\boldsymbol{\sigma}_i \;\forall\boldsymbol{x} \in V_{hp} \\ (\bar{\boldsymbol{\varepsilon}}_i + \tilde{\boldsymbol{\varepsilon}}(\tilde{\boldsymbol{u}}_i)) : \mathbb{C}_w \;\forall\boldsymbol{x} \in V_w \\ (\bar{\boldsymbol{\varepsilon}}_i + \tilde{\boldsymbol{\varepsilon}}(\tilde{\boldsymbol{u}}_i)) : \mathbb{C}_{air} \;\forall\boldsymbol{x} \in V_{air} \end{cases}, \quad (9)$$

$$\tilde{\boldsymbol{\varepsilon}}(\boldsymbol{u}) = \frac{1}{2}(\nabla\boldsymbol{u} + (\nabla\boldsymbol{u})^T), \quad (10)$$

where $\mathbb{C}_w$, $\mathbb{C}_{air}$ – is the fourth-order elasticity tensor of water and air, respectively;

V_{uc}, V_{hp}, V_w, V_{air} – are the subdomains of the REV referring to the phases of unhydrated cement, hydration products, water, and air, respectively.

The is the incremental of the stress tensor $\Delta\boldsymbol{\sigma}_i$ is expressed from (5), where $\Delta\boldsymbol{\varepsilon}_i = \Delta\bar{\boldsymbol{\varepsilon}}_i + \Delta\tilde{\boldsymbol{\varepsilon}}_i$.

Solidification of cement paste

According to the solidification theory, increment of stiffness of cement paste during a hydration process is given in relation to increment of a cluster thickness, which is represented by increment of the volume of hydration products (see Figure 1), and it is assumed that the properties of clusters do not vary with time [9]. The total number of fictitious clusters is equal to the number of time intervals.

The above principle can be easily applied to discrete REV, where a separate cluster is considered as a set of hydration product voxels that have been produced at i-th time interval. The use of the voxel-based microstructural model, for instance [9], allows creating a history of the formation of such clusters, as well as to keep the spatial position of the cluster voxels in the REV.

This also means that the finite elements within a mesh subdomain belonging to each cluster can be identified and explicitly associated with the stress-strain relation on the corresponding time interval unlike the classical approach where the fictitious clusters are assumed to be a dimensionless variable that is equal to the increment in degree of hydration.

Modelling results

The Portland cement paste with parameters reported in Table 1 was used for the simulation.

The parameters of the constitutive phases of cement paste report in Table 2. The elastic properties of the phases in Table 2 were taken according to [19].

Table 1 – Parameters of cement paste

Mix proportions, kg/m³		Water to cement ratio	Density of cement, kg/m³	Fineness of cement, m²/kg	Mineral composition of cement (mass %)
Portland Cement	Water				
370	185	0,5	3150	345	C_3S: 54,5; C_2S: 17,3; C_3A: 8,9; C_4AF: 7,6; Gypsum: 5

Table 2 – Parameters of the constitutive phases of cement paste

Parameter	Phase										
	C_3S	C_2S	C_3A	C_4AF	CSH	CH	$C_6A\bar{S}_3H_{32}$	$C_4A\bar{S}H_{12}$	C_3AH_6	FH_3	Gypsum
Young's modulus, GPa	137,4	135,5	145,2	150,8	23,8	43,5	24,1	43,2	93,8	22,4	44,5
Poisson's ratio	0,299	0,297	0,278	0,318	0,24	0,294	0,321	0,292	0,32	0,25	0,33
Aspect ratio	1,0	1,0	1,0	1,0	0,01	0,1	100	10	1,0	1,0	1,0

The stiffness characteristics of water: the bulk modulus is 2,2 GPa, the Poisson's ratio is 0,499. The stiffness characteristics of air were taken to be close to zero.

The REV resolution of 10 voxels/edge was used in simulation to reduce the computational cost.

Six elementary load cases consisting of macroscopic uniaxial strain and shear solicitations were applied at each time step by assigning constant unit values $\bar{\varepsilon}_{ij}$: $\bar{\boldsymbol{\varepsilon}} = \boldsymbol{e}_i \otimes \boldsymbol{e}_i$ – for uniaxial strain, and

$$\bar{\boldsymbol{\varepsilon}} = \frac{1}{2}\left(\boldsymbol{e}_i \otimes \boldsymbol{e}_j + \boldsymbol{e}_j \otimes \boldsymbol{e}_i\right) \text{ – for shear strain} \quad (11)$$

where $\boldsymbol{e}_i$ – is the unitary bases;

«$\otimes$» – is the tensor product.

Two additional creep compliance functions $J(t, t')$ were used for a comparative analysis of the effective properties:

1) adapted ACI model [20]:

$$J(t,t') = \frac{1+\varphi(t,t')}{E_{hp}(t')};\ (t,t') = 2{,}35 \cdot \frac{(t-t')^{0,6}}{10+(t-t')^{0,6}}, \quad (12)$$

where $E_{hp}(t')$ – is the effective elastic modulus of hydration products.

2) adapted CEB MC90 model [14]:

$$J(t,t') = \frac{1}{E_{hp}(t')} + \frac{\varphi(t,t')}{E_{hp,28}};\ \varphi(t,t') = \left[\frac{(t-t')}{500+(t-t')}\right]^{0,3}, \quad (13)$$

where $E_{hp,28}$ – is the effective elastic modulus of hydration products at 28 days, 33 GPa.

To evaluate the stiffness tensor considering the percolation of the solid phase of cement paste, a power law in the following normalized form was used:

$$\mathbb{C}_{eff} = \mathbb{C}_{eff}^{FEA}\left(\frac{\alpha-\alpha_{per}}{1-\alpha_{per}}\right)^{\gamma}, \quad (14)$$

where $\mathbb{C}_{eff}^{FEA}$ – is the effective fourth-order stiffness tensor of cement paste according to the FEA-based homogenization;

α – is the hydration degree of cement;

α_{per} – is the hydration degree of cement corresponding to the percolation threshold of the solid phase;

γ – is the exponent, 1.

The modeling results are presented in in Figures 2 and 3.

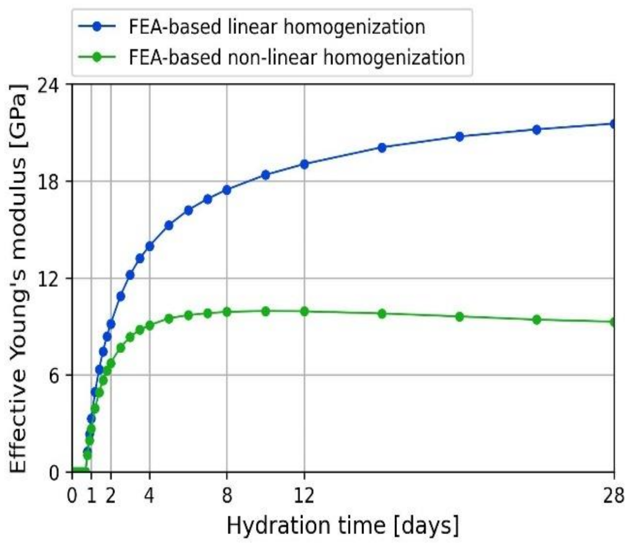

Figure 2 – Effective behavior of cement paste

Conclusions

1. Modelling the effective stiffness characteristics of cement paste is a rather difficult task, due to the combination of the facts that its structure consists of randomly distributed phases with an arbitrary geometry, and a solid phase has viscoelastic behaviour.

2. The article presents the approach to solving the problem of effective stiffness characteristics of cement paste given the above issues based on a FEA homogenization makes it possible to evaluate its effective properties for arbitrary phase geometry, throw introducing into the variational formulation the numerical inversion of the Stieltjes integral describing the constitutive model of viscoelastic of the solid phase of cement paste.

3. An important advantage of above approach is that the voxel REV used to generate the mesh can also implement solidification theory principles to express the strain and stress history associated not with fictitious but with each identified cluster formed during the hydration process.

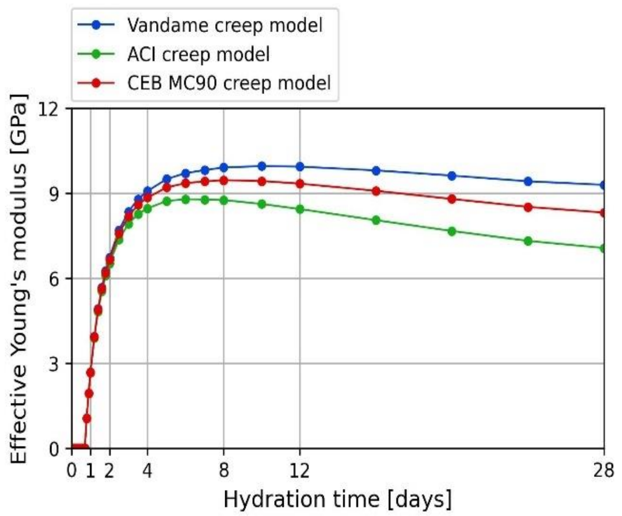

Figure 3 – Effective behavior of cement paste based on non-linear FEA-based homogenization using different creep models

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Material received 11/11/2024, approved 28/11/2024, accepted for publication 28/11/2024