Material Properties Prediction for Long Term Operation of Nuclear Power Plants Civil Engineering Structures: Challenges at EDF

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Abstract

To manage the long term operation of nuclear power plants civil engineering structures, EDF has adopted an approach based on inspection, structural analysis and material ageing knowledge. This approach allows to monitor the degradation of civil works, and optimize maintenance, to ensure a safe operation.

The assessment of concrete ageing plays a significant role in this approach, and is based on both experimental and modelling works, to anticipate the long-term behavior of concrete. EDF research and development (R&D) is thus developing a software tool to help EDF engineering services to predict the ageing of cementitious materials. This tool is called Virtual Cement and Concrete Ageing Analysis Toolbox v2 ($Vi(CA)_2T$ v2). To be predictive on a large number of concrete mixes, it must rely on physics-based models. These models can be gathered in two groups:

- hydration models, based on physics, kinetics and chemistry of cement hydration, aimed at predicting the evolution of the volume fractions of cement phases,
- micromechanical models, based on mean field homogenization, aimed at predicting transport and mechanical effective properties from the volume fractions of cement phases.

This contribution proposes an overview of the challenges faced by the development of such models, which must integrate as much physical information as possible, while still maintaining a reasonable computing time.

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I. INTRODUCTION

A. Industrial context and needs

For managing the long term operation of nuclear power plants, EDF has adopted an integrated approach [1], combining structural analysis, inspection and material ageing knowledge. These three pillars constitute inseparable elements of the approach:

- without structural analysis, projecting the structure behavior into the future is not possible,
- without inspection, diagnostics on the current state of the structure cannot be obtained,
- without material ageing knowledge, structure computations cannot be fed with proper material behaviors.

This contribution deals with the material ageing aspect, which aims at:

- as far as inspection is concerned, for non-destructive techniques which involve indirect measurements, providing a reliable relation between the quantity of interest (water content for example) and the measured quantity (electric conductivity or permittivity for example);
- as far as structural analysis is concerned, providing the appropriate material properties and behaviors, to be used as inputs of FEM computations for example.

In short, the physical, transport and mechanical properties and behaviors of *the* concretes making up the structure at stake are required.

In this respect, the complete experimental characterization of every concrete of every civil engineering structure EDF has to manage is out of scope:

- as made up from local raw materials, each concrete is unique, and EDF has estimated as around 1000 the number of different mix designs,
- drilling core samples from the structure is not always feasible, and only gives access to the current state of the material, not to the whole history.

Thus, modelling approaches represent an attractive alternative, as they allow to estimate, from the knowledge of both the initial mix design and the ageing conditions in the structure, the properties and behavior of concrete. To be predictive and adaptable to a rather wide range of concrete mixes, these models clearly have to be physics-based rather than phenomenological.

Concrete being a highly variable, multi-scale material, where multi-physics processes occur, multi-scale models represent an appealing option. Indeed, these approaches, based on micromechanics, are able to bridge the scale where the physical processes occur to the scale of interest for the engineer. In other words, it is possible to go down to the scale where the complex behaviors and couplings observed macroscopically can be physically investigated and possibly un-coupled (see [2] for example regarding the creep and damage coupling).

B. A software tool dedicated to concrete

Recent progresses regarding the physics-based modelling of cementitious materials (see [3], [4], [5], [6], [7] to cite a few references) make the approach promising. However, knowledge is scattered worldwide, and models from the literature may not be directly usable by engineers, as they often rely on rather involved techniques. Thus, structural engineers had, for a long time, to mainly rely on empirical models whose predictions and adaptability from one mix to another can be questionable.

To overcome this situation, Le Pape initiated at EDF R&D the development of a software tool dedicated to concrete [8], and usable by engineers. As a reference to the father of modern cement industry, Louis Joseph Vicat, the tool is called Vi$(CA)_2$T, for Virtual Cement and Concrete Ageing Analysis Toolbox. It is based on the concept of a "virtual lab" or "virtual material": from the mix design details, it is able to estimate physical properties (such as the evolution of hydration heat, capillary porosity or water content) and mechanical behaviors (such as the evolution of the Young's modulus, or the basic creep rate at early and late ages). To do so, it embeds several kinds of models:

- Cement paste hydration models: combining chemistry and kinetics, from the initial mix design information, the amounts of phases (anhydrous phases, hydrates, water, porosity) are estimated as a function of time.
- Micromechanical models: taking advantage of upscaling techniques, from the amount and elementary behavior of phases, the effective mechanical behavior is estimated (such as stiffness or some characteristics of basic creep).

The tool has recently been revamped to version 2 by Sanahuja and Reviron [9], to further improve the engineer-friendliness, and to incorporate either refined or new models, to further extend the range of predicted properties and behaviors. The new code structure has been designed with continuous improvement in mind: new models can be implemented as they either appear in the open literature or are developed in-house. The current main improvements with respect to version 1, regarding modelling, are:

- hydration in presence of silica fume, in addition to the four anhydrous phases of clinker (C_3S, C_2S, C_3A, C_4AF) and gypsum, and considering simple heat transfer conditions (isotherm, adiabatic, or including some losses);
- morphological models (at the basis of upscaling techniques) can now be defined from a straightforward text file, instead of requiring to implement the corresponding code;

- the complete basic creep function can now be estimated, instead of just the asymptotic creep rates (at $t \to 0$ and $t \to \infty$), but still considering the microstructure as frozen once the stress loading step has been applied.

Moreover, quality assurance aspects have been improved, adding unit and verification tests.

C. Main challenge

As such a software tool must be usable by EDF engineering services, a crucial feature is the responsiveness: heavy computational models, taking several hours or days, are not conceivable in this context.

The main challenge posed by the development of such a tool is thus: *how to integrate models as much physics-based as possible, while still maintaining computation time below a couple of minutes on a typical desktop computer?* This main challenge calls several other ones, for each part of the software (hydration, microstructure modelling, upscaling). The latter are detailed in section 2. Then, an overview of the EDF strategies to overcome these challenges is proposed in section 3, with a specific focus on basic creep, which is especially critical for prestressed containment buildings.

II. RESPONSIVE AND PHYSICS-BASED MODELLING OF CEMENTITIOUS MATERIALS: A FEW CHALLENGES

This section proposes an overview of the challenges associated to designing models to predict the effective behavior of cementitious materials.

A. Hydration modelling

Hydration modelling is the starting point of every study performed with Vi(CA)$_2$T. The aim of the hydration model is to predict the time evolution of:

- the various elementary phases of cement paste (as aggregates are assumed to be chemically inert): anhydrous, hydrates, water and, by extension, capillary porosity,
- the heat produced by hydration reactions and/or the temperature evolution;

from the following input data:

- mix design information,
- thermal conditions (either isothermal, adiabatic or considering a given expression of heat losses).

Time response constraints prevent the use of hydration models providing a full 3D description of the evolving microstructure of cement paste, such as CEMHYD3D [10], HYMOSTRUC [11] or µic [12]. Thus, a simplified approach, which does not need to describe the microstructure, is adopted. Kinetics are modelled based on the laws of Avrami [13] and of Fuji and Kondo [14], while stoichiometry is enforced through commonly used simplified hydration reactions of clinker components (C_3S, C_2S, C_3A, C_4AF) and silica fume [15]. The heat conservation equation is also written.

This modelling yields an initial value problem involving a system of first-order ordinary differential equations:

$\{1\}\dot{X} = f(X,t)$ and $X(0) = X_0$

where X is a time-depending vector made up of the amounts of C_3S, C_2S, C_3A, C_4AF and silica fume present in the modelled system, and of temperature. This initial value problem is numerically integrated using a solver available in the Python SciPy package (method using Backward Differentiation Formulas, from the Variable-coefficient Ordinary Differential Equation solver), suited for stiff problems. Parameters for the kinetics model are extracted from [3] for clinker phases and from [16] for silica fume. The latter have been validated for silica fume contents up to 10%. The model is able to correctly reproduce the influence of silica fume on hydration kinetics [Fig. 1.].

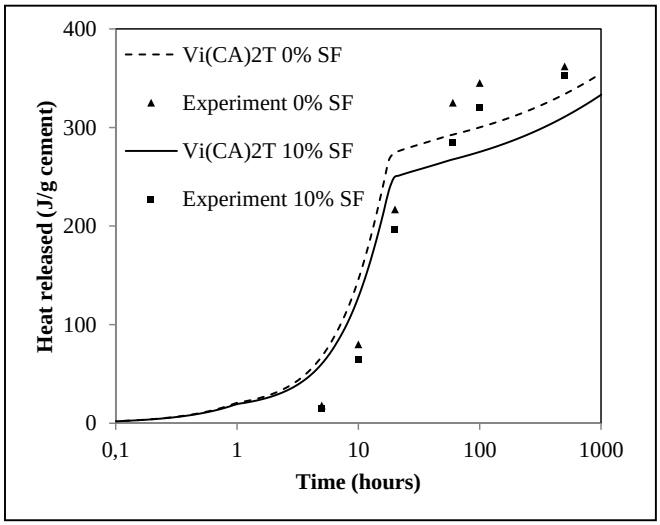
The main challenge here is to model as accurately as possible the physico-chemical interactions between the various anhydrous and hydrated phases. Integrating the influence of other supplementary cementing materials, such as fly ash and slag, yields another level of difficulty.

B. Microstructure modelling

The micromechanical models used to estimate the effective behavior take as inputs:

- the volume fraction of each phase,
- the elementary behavior of each phase,
- a morphological model describing the geometrical arrangement of phases.

Fig. 1. Cumulated heat released estimated from Vi(CA)$_2$T and experimentally measured (data from [17]), for mortars without and with 10% of silica fume (SF).



The effective behavior highly depends on the morphological model. Thus, significant efforts should be driven towards accurate description of microstructure.

Upscaling can be done either numerically, with full field homogenization (explicitly described morphology, as meshes or images [18], [19]), or analytically, with mean field homogenization (implicitly described morphology). Time response constraints prevent the use of full field homogenization in $Vi(CA)_2T$, thus the morphological model has to be simplified. Note that full field computations are still useful, as means of evaluation of the ability of mean field homogenization to accurately take into account specific morphological features.

The basic framework of morphological modelling driven towards mean field homogenization is briefly recalled here, considering as an example the case of elasticity. First, some scale separations must be introduced: phases appearing as continuous and homogeneous at a given scale can be detailed, in terms of microstructure, at a lower scale. Thus, upscaling is performed step by step, up to the scale of interest. Second, at each scale, to ensure semi-analytic computations, morphology has to be described choosing features among a restricted set:

- ellipsoidal particles, to take advantage of the Eshelby inclusion result [20],
- morphologically representative patterns [21], such as *n*-layered spherical particles [22].

These particles can be arranged in two different ways:

- particles embedded into a continuous phase, called matrix (Mori-Tanaka scheme [23]),
- particles not separated by a matrix but in direct contact, that is, arranged as a polycrystal (self-consistent scheme [24]).

Thus, at each scale, a so-called homogenization scheme can be built from the morphological model, and translated into a semi-analytical procedure to estimate the effective stiffness.

Regarding concrete, a rather natural scale separation can be introduced between aggregates (including sand) and the cement paste: concrete can be considered as aggregates inclusions in a cement paste matrix. Refined models would introduce more scale separations in the aggregates sieve curve, for example between coarse aggregates and sand. To simplify, aggregates can be modelled as spheres, at least for linear behaviors, as the influence of the shape on the effective behavior has been found to be small [19]. If required, an interfacial transition zone can be introduced around inclusions, using morphologically representative patterns.

Morphology of cement paste is much less clear, and there are still debates in the community. While the (possibly remaining) anhydrous grains can be represented as spherical inclusions, hydrated phases are much harder to describe, especially the C-S-H gel. Observing details of the latter (at scales starting from a few nanometers) is indeed difficult. Models of C-S-H gel based on globules [25], fibers [4], or lamellae [26] have been proposed. At the cement paste scale, several morphological models can be found in the literature: [3], [27], [4], to name a few. Based on some microstructure observations and on a trial and error process comparing the evolution of effective stiffness with respect to experimental data, a simplified morphology has been designed for cement paste [28], and straightforwardly extended to concrete [29], see [Fig. 2.].

It has the advantage to describe setting: at the beginning of hydration, the effective stiffness predicted is zero, and the onset of the latter occurs at a given hydration degree, which depends on the water to cement ratio. The model is found to overestimate by around 15% the experimental Young's modulus of several concretes [Fig. 3.]. Note that the experimental scatter (measurements at one year have been performed by two different labs) is of the same order of magnitude.

Another challenge would be to model or describe the porous network, or at least the pore size distribution, which are useful for transport or shrinkage estimations.

Fig. 2. Proposed multi-scale morphological model of concrete (2D sketch; morphology is actually 3D) [28], [29].

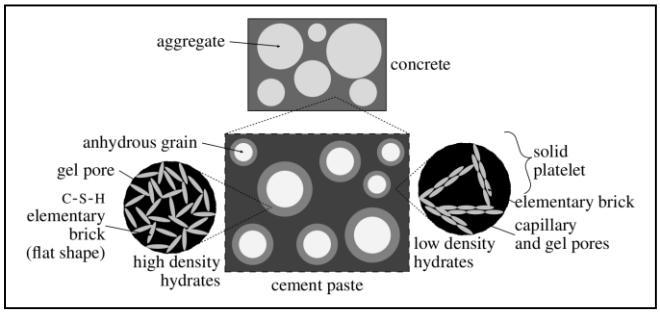
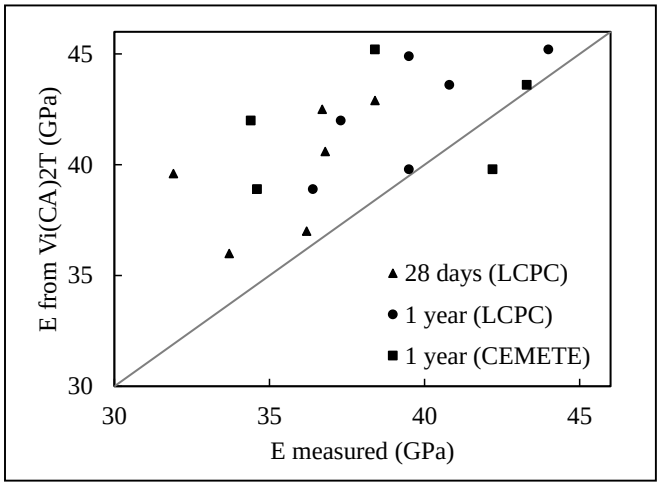


Fig. 3. Young's modulus of several concretes measured (data from [30]) and estimated from $Vi(CA)_2T$, at both 28 days and 1 year.



C. Knowledge on micro-mechanisms

Estimating mechanical properties or behaviors beyond elasticity often require to start from informations on the main micro-mechanisms responsible of the effective behavior at stake.

For example, as far as basic creep is concerned, consensus has not been reached yet on the main nano- or micro-mechanisms responsible of this macroscopic behavior. Many hypotheses can be found in the literature [31], among which transfers between capillary and adsorbed water [32], water transfers to newly created microcracks [33], C-S-H sheets viscous sliding [34], …

As this debate regarding the relevant creep mechanisms seems far from being closed, an assumption has to be made. The sheet sliding mechanism, in C-S-H bricks, is considered here. Thus, transforming the elastic multi-scale model into a basic creep one only involves the modification of the elementary behavior of the C-S-H bricks for the strain mechanisms involving sliding of sheets. A Maxwell model is considered as a first step, so that the corresponding characteristic time is the only extra parameter with respect to the elastic model.

D. Upscaling

While upscaling elasticity is straightforward (at least once the morphological model has been chosen, and the elementary stiffness and volume fractions are known), upscaling other properties such as strength or basic creep represents an ambitious challenge as far as cementitious materials are concerned. Regarding strength, even if recent approaches seem promising (for example [35], [36]), the proper description of damage, micro-cracking and cracks coalescence seems to be a genuine challenge for current mean field homogenization approaches.

Regarding basic creep, when the microstructure does not evolve during the stress loading, the correspondence principle [37] can be taken advantage of. Indeed, the Laplace-Carson transform changes non ageing linear viscoelastic behaviors into elastic ones. Elastic homogenization approaches can thus be directly reused. From the morphological model validated in elasticity [Fig. 2.], the effective uniaxial compliance functions of various concretes have been estimated [Fig. 4.]. The micromechanical model provides the right order of magnitude but systematically overestimates the experimental creep. Many sources of improvement can be pointed out. Among them, neglecting the time evolution of microstructure (due to hydration, to possible densification or polymerization of hydrates, …) is required to take advantage of the correspondence principle, but is an important source of approximation. Indeed, as illustrated on [Fig. 5.], not considering microstructure evolution during loading yields an overestimation of the real creep compliance, in this simplified case of hydrates massively precipitating into pores [38].

Fig. 4. Uniaxial basic creep compliance of 6 concretes, loaded at 28 days, measured (dots, data from [30]) and estimated from Vi(CA)$_2$T considering frozen microstructure once the stress step has been applied.

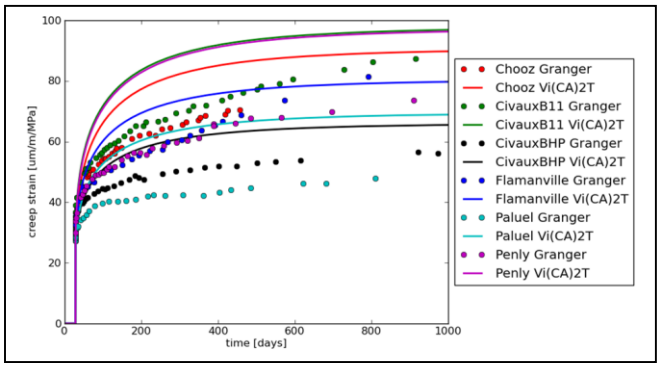
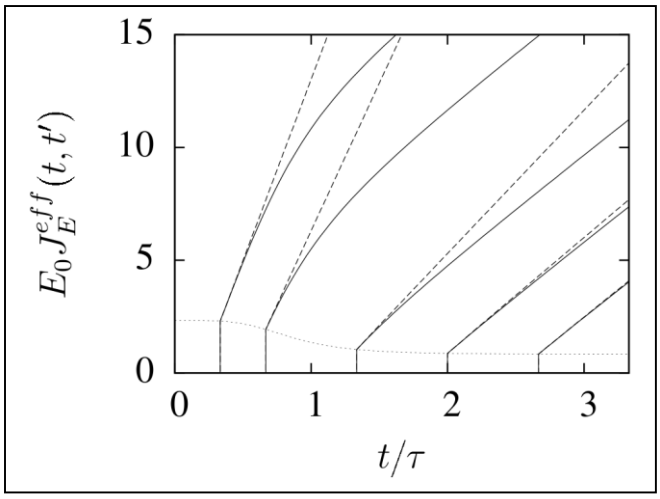


Fig. 5. Ageing effective uniaxial compliance functions, when hydrates massively precipitate in pores, without (dashed lines) and with (plain lines) considering microstructure evolution during stress loading; dotted line represent the initial elastic strain, estimated from elasticity upscaling [38].



The transport properties, even if not detailed here, represent another difficulty, as the high values (sometimes infinite) of contrast between phase properties makes the effective properties highly dependent on morphology. The latter can only be represented in an approximate way by mean-field homogenization, even if recent approaches on unsaturated media appear promising [7], [39].

III. OVERVIEW OF EDF APPROACHES CALLED BY THESE CHALLENGES

This section proposes an overview of some technical approaches developed at EDF to overcome some of these challenges, focusing on microstructure and estimation of ageing basic creep.

A. Microstructure

As far as morphological models of cementitious materials are concerned, many options can be found in the literature, and consensus does not seem to be reached yet. The ability to quickly and easily integrate and test new morphological models is thus crucial. A specific module has been developed to interpret morphological models described as simple text files. The latter describe morphology as a directed graph, where:

- the vertices are either elementary phases or composites made up (at a lower scale) of elementary phases and/or composites; vertices can embed shape information;
- the directed edges can be of two kinds, to represent either the relation "made up of, at a lower scale" or the relation "included in, at the same scale".

This approach allows flexibility, and new morphological models can be easily integrated by non-developers. To illustrate the graph structure, the concrete morphological model of [Fig. 2.] is represented on [Fig. 6.].

B. Ageing creep

Regarding creep, the challenges posed by both the requirement of efficient computations and the evolving nature of the microstructure conducted to develop a specific upscaling approach. These morphological evolutions come from progressive phase transformations, that is either dissolution, precipitation, or solid to solid transformations. Several of these processes can occur at the same time. The idea, inspired by the solidification theory from Bazant [40], is to replace the evolving microstructure by a constant one, introducing fictitious phases whose behavior is ageing linear viscoelastic [41]. The constant morphology has an equivalent behavior to the original evolving one. The constant nature of the equivalent microstructure then allows to take advantage of mean field homogenization, which has been recently adapted [42] [43] to generic ageing linear viscoelastic behaviors, defined in integral form:

$$\{2\}\boldsymbol{\sigma}(t) = \int_{t'=-\infty}^{t} [3k(t,t')\mathbb{J} + 2g(t,t')\mathbb{K}] : \mathrm{d}\boldsymbol{\varepsilon}(t')$$

with *any* isotropic bulk *k* and shear *g* relaxation functions.

Preliminary application to model precipitating materials (a pre-existing matrix and various precipitation processes into pore space) showed that the effective ageing behavior is highly dependent on both the precipitation mechanism and kinetics [44]. A first application to cement pastes, albeit simplified, is proposed in [45]. The evolving morphology is reproduced on [Fig. 7.] and consider both dissolution of anhydrous and precipitation of hydrates. Effective creep functions are plotted on [Fig. 8.]. Even if the elementary behaviors of phases are non ageing, the effective behavior is ageing. Note that the "frozen microstructure" assumption yields very different results, and that even the initial creep rate is not properly estimated, contrary to what happened in the simple precipitation case of [Fig. 5.]. This may be due to the dissolution process, as already pointed out by [46] with full field simulations. Applications to more realistic evolving morphologies are ongoing.

Fig. 6. Graph structure corresponding to multiscale morphological model depicted on [Fig. 2.].

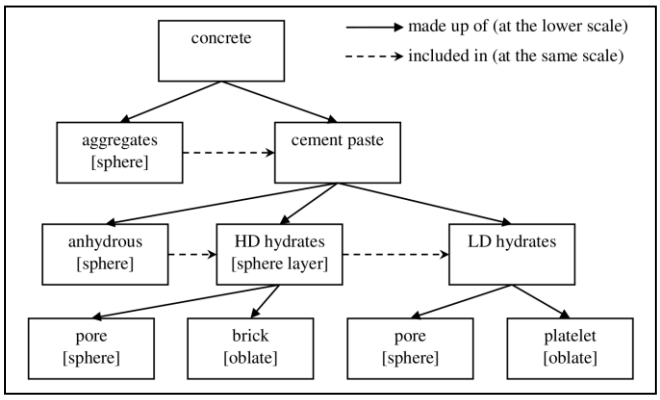


Fig. 7. Simplified evolving microstructure of cement paste (w/c=0.6): snapshots at various hydration degrees [45].

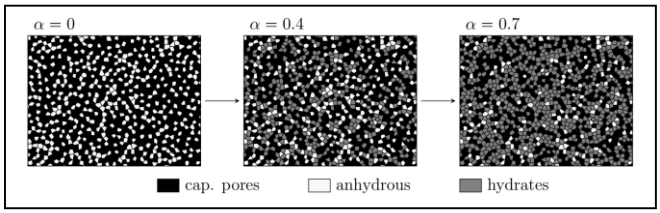
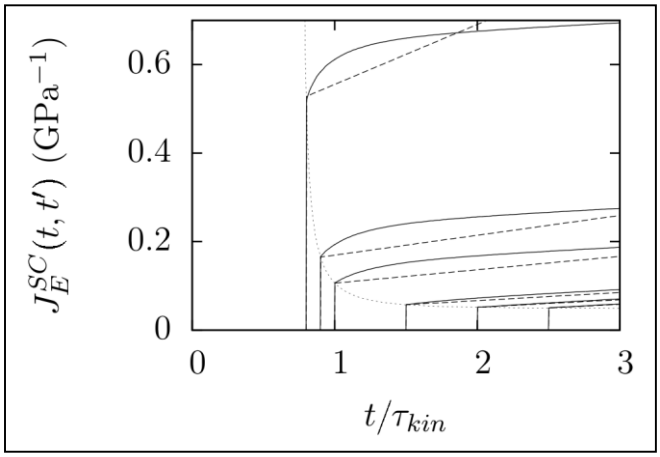


Fig. 8. Ageing effective uniaxial compliance functions estimated for simplified cement paste, with frozen microstructure (dashed lines) or considering microstructure evolution (plain lines) during stress loading; dotted line represent the initial elastic strain [45].



IV. CONCLUSION AND PROSPECTS

The physics-based estimation of effective behavior of cementitious materials raises a number of challenges. The latter are partly due to the lack of knowledge or consensus regarding morphology or micro-mechanisms, and partly due to technical difficulties regarding upscaling of specific behaviors or properties. Integrating these models into a tool useful for EDF engineering services poses even more challenges, mainly related to the need of responsiveness: an ageing creep estimation may not take more than a couple of minutes.

The first kind of challenges calls the development of advanced experimental techniques to better characterize microstructure and micro-mechanisms, if possible. If not possible, trial-and-error based approaches can be an option: assumptions are made on either the microstructure or the micro-mechanisms, and predictions are compared to macroscopic experimental results. This allowed to successfully validate the morphological model of cement paste proposed in [28], upon which the concrete model integrated into $Vi(CA)_2T$ v2 is based, with several improvements to take into account the diversity of anhydrous and hydrated phases.

Regarding ageing basic creep due to the evolving nature of the microstructure, the approach [45] starts to be mature enough. The main challenge is now to apply it on realistic (albeit simplified) microstructure evolution scenarii (as a consequence of hydration, of hydrates densification or polymerization, or even of various degradation mechanisms, etc...).

Some of the challenges exposed in this contribution are ambitious, and posed to the entire cementitious materials scientific community. Regarding EDF efforts, it is now time to measure progress. For example, concerning basic creep:

- The tool started a few years ago with the prediction of only the asymptotic creep rates at very early ($t \rightarrow 0$) and very late ($t \rightarrow \infty$) ages [47] under the assumption of a frozen microstructure once loaded.
- Then estimation of the complete creep curve has been implemented, still assuming a frozen microstructure [29].
- Currently, microstructure evolutions during loading can be taken into account [44], still profiting from mean field homogenization and its (somewhat but reasonable, in the case of ageing viscoelasticity) fast response.

Moreover, without the $Vi(CA)_2T$ v2 platform, it would have been much more time consuming to apply the various (hydration, elasticity, creep) models to different concrete mix designs, to compare to experimental results. In the near future, these comparisons will be automated, including the fit of some lower-scale model parameters which are not accessible from usual tests.

Finally, regarding homogenization approaches and microstructure description, continuous progresses in computer hardware might allow, in the near future, to reconsider the limitation to mean field approaches. Indeed, technologies such as GPGPU computing seem appealing to accelerate full field computations, maybe up to the point that detailed microstructures become reasonably usable by engineering tools. Improving computational efficiency of the material code would also allow to call the latter for each Gauss point of the structure FEM computation, to benefit from "scale" coupling. Before tackling this ambitious challenge, a first approach can be to fit or optimize, on simulations at the material scale, the parameters of mechanical behavior laws to be used at the structure scale [48].

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