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# A LINEAR ELASTIC FRACTURE MECHANICS-BASED STUDY OF THE INFLUENCE OF AGGREGATE SIZE AND GEL MECHANICAL PROPERTIES DURING ALKALI-SILICA REACTION

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## Abstract

We present a fracture mechanics model for Alkali-Silica Reaction for concretes made up of dense aggregates. Chemistry and diffusion (ions and gel) are not modelled. The focus is put on the mechanical consequences of the replacement of the outer layer of the aggregate by a less dense gel. A schematic cracking pattern is assumed: a ring-shaped crack appears in the cement paste surrounding the spherical aggregate depending on the pressure build-up. The onset of cracking is determined using an incremental energy criterion. The stored elastic energy and deformation of a given configuration are determined (using FEA) assuming that each aggregate behaves as if it was embedded in an infinite cement paste matrix. We note a very different behaviour of aggregates of different sizes. The macroscopic deformation is estimated by adding the aggregate's individual contributions. A rate of attack is identified that leads to recover the usual sigmoid ASR expansion curve.

**Keywords:** Alkali-aggregate reaction, Free expansion, Linear Elastic Fracture Mechanics, Energy criterion

## 1 INTRODUCTION

The alkali-silica reaction (ASR) has been discovered in the USA by Stanton [1]. During ASR, the silica released by the dissolution of the aggregates combines with alkali and calcium to produce swelling gels. These gels can appear in various places (aggregate itself, interface transition zone or through the porosity of the cement paste) depending on the distribution of the reactive silica throughout the aggregate, the availability of calcium and the transport properties of the concrete. It is often stated in the ASR literature that the growth of gel-filled microcracks is responsible for a large part of the macroscopic expansion. In this article, following previous models like that of Bazant [2], we try to make a link between the aggregate size and the properties of the gels, and the macroscopic swelling. To do so, we simplify the inputs of the problem as much as possible, assuming for example that the gel appears at the aggregate/cement paste interface and that this interface is of negligible strength, in order to build the basis of a more sophisticated model that will consider the effect of an external mechanical loading and more physics regarding the attack of the aggregate itself.

First, we explain briefly why we propose a new fracture model for ASR and its differences with existing ones. Then follow the description of the considered elementary volume and the computation of the energies used in the energy fracture criterion presented right after. The simple rule used to sum the contributions of different elementary volumes to macroscopic expansion comes next. Later, we explain which parameters influence the results of the model, focusing on aggregate size and the properties of the ASR gel. Finally, we discuss the potential of our model for reproducing experimental expansion curves.

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## 2 RESTRICTIONS OF THE MODEL PROPOSED IN THIS ARTICLE

In this first attempt to understand the behaviour of an aggregate submitted to an attack, we only study a simplified mechanical problem related to ASR. Our goal is to understand the role of cracking of the cement paste in the swelling of concretes submitted to ASR. Therefore, we restrict our model to the simplest elements leading to the appearance of a crack in the cement paste under gel pressure. We want to understand how the size of aggregates influence cracking and hence, ASR expansion. The model follows many ideas of Bažant's mechanical model for ASR [2]. In this model a schematic 14 days attack is considered, corresponding to the accelerated expansion test ASTM C 1260. The decrease of the concrete's resistance and its expansions due to fracture of the cement paste between a periodical glass aggregate pattern are estimated thanks to Linear Elastic Fracture Mechanics. Let us explain the main differences here. First, in our model, no assumption is made about the periodicity of the aggregates in the cement paste. Instead, each aggregate is assumed to be embedded in infinite cement paste. Second, the basis for crack propagation is a Finite Element Analysis (FEA), instead of an interpolation between analytically known stress intensity factors. On this point, our approach is close to that of Xiao [3] where the authors study, through FEA, the evolution of stress intensity factors for ring cracks surrounding inclusions when varying various parameters, except that we work directly on energies. We also compute the volume available to the gel by FEA which makes it simpler than what is done for example in [2] where it is deduced from the cracked medium's compliance which is obtained by integration (with respect to the crack size) of the stress intensity factor. Then, our determination of average deformations far from the reaction sites is quite simple. Finally we make no assumptions about the compressibility of the ASR products relatively to the cement paste and we briefly discuss its importance (like in [4]).

Our goal in this article is to build a reliable method to create and propagate cracks that we'll be able to use under external loadings to study the anisotropy of expansion, later on. We plan to use this methodology to finally be able to extract macroscopic information about the anisotropy of swelling and decrease of material properties that can be used in structure-size FEA.

## 3 DESCRIPTION OF THE BEHAVIOR OF AN ELEMENTARY VOLUME

Our elementary volume is a single spherical aggregate of radius  $R_p$  surrounded by an infinite cement paste which is free at infinity (Figure. 1). Hence, the interaction between aggregates is neglected. The attack of the aggregate is represented by replacing the aggregate by a gel from the outside to the inside. The relative attack depth is called  $\alpha \in [0;1]$ , that is at a given time, an aggregate of radius  $(1-\alpha)R_p$  remains. The aggregate is supposed to easily separate from the cement paste due to the mechanical weakness of the interface transition zone (ITZ) surrounding it. The aggregate and cement paste are considered linear isotropic elastic of properties  $(E_a, \nu_a)$  and  $(E_c, \nu_c)$ . The gel bulk modulus is  $K_{gel}$ . Under zero pressure, the volume of gel replacing a given volume of aggregate is  $\delta$  times bigger. Hence, the undeformed gel volume at a given attack degree writes  $V_0(\alpha) = \frac{4}{3}\pi\delta R_p^3 [1 - (1-\alpha)^3]$ . Let us stress that  $\delta$  could be an effective value of the ratio of the gel volume to the aggregate volume accounting for physical phenomena such as the invasion of a part of the porous space by the gel (particularly the ITZ, of bigger porosity). It would then depend on the gel pressure and the aggregate size. To keep our model simple, we consider it as a constant.

We study the cracking of the cement paste under the increasing pressure created by gel accumulation around the aggregate. Quantities relative to the gel, the aggregates, and the cement paste, respectively have the indexes *gel*, *a*, and *c*.

#### 4 EXPRESSION OF GEL PRESSURE AND ELASTIC ENERGIES

The cracks we are considering are ring cracks developing in a symmetry plane of the spherical cavity (See Figure 1). Therefore, we do not take the influence of neighbouring aggregates on crack propagation into account. A crack around a cavity of radius  $R_p$  is characterized by its relative size  $x$  defined as the difference of the crack's outer and inner radii, divided by  $R_p$ . The gel pressure first has to be computed. The compressed gel volume writes:

$$V(\alpha, x) = V_0(\alpha) \left(1 - P(\alpha, x)/K_{gel}\right) \quad (1)$$

where  $P(\alpha, x)$  is the homogeneous gel pressure where as for  $V(\alpha, x)$ , we only keep explicit the dependence in the degree of attack  $\alpha$  and the relative crack size  $x$  which are the two variables describing the evolution of our elementary volume. To find the gel pressure, this gel volume has to be compared to the volume left available by the deformation of the solid components. When a crack exists, no closed form solution is available. Hence, we resort to FEA to compute the variation of volume available to the gel, at given crack size, due to the deformation of the cement paste. Dimensional analysis leads us to write this quantity  $\Delta V_c(\alpha, x) = \frac{4}{3}\pi \left(R_p^3 P(\alpha, x)/E_c\right) \Delta v(x)$  where  $\Delta v(x)$  is dimensionless. Hence, the volume accessible to the gel writes:

$$V(\alpha, x) \approx \frac{4\pi}{3} R_p^3 \left[ \left(1 + \frac{P(\alpha, x)}{E_c} \Delta v(x)\right) - (1-\alpha)^3 \left(1 - 3P(\alpha, x) \frac{1-2\nu_a}{E_a}\right) \right] \quad (2)$$

where in the term concerning the compression of the aggregate, the assumption  $P(\alpha, x) \ll E_a$  has been used to keep the expression linear in the pressure. Thanks to this approximation, equating expressions (1) and (2) yields a linear expression of the pressure which can hence be easily written as:

$$P(\alpha, x) \approx \frac{(\delta-1) \left[1 - (1-\alpha)^3\right]}{\frac{\delta \left[1 - (1-\alpha)^3\right]}{K_{gel}} + \frac{\Delta v(x)}{E_c} + 3(1-\alpha)^3 \frac{1-2\nu_a}{E_a}} \quad (3)$$

The elastic energies of all components are needed. For the aggregate and the gel, their expressions are in closed form. For the gel it writes:

$$E_{gel}^{el}(\alpha, x) = \frac{2\pi}{3} \delta R_p^3 \left[1 - (1-\alpha)^3\right] \frac{P^2(\alpha, x)}{K_{gel}} \quad (4)$$

while concerning the aggregate:

$$E_a^{el}(\alpha, x) = 2\pi R_p^3 (1-\alpha)^3 \frac{1-2\nu_a}{E_a} P^2(\alpha, x) \quad (5)$$

Regarding the cement paste, dimensional analysis leads us to write it under the following form:

$$E_c^{el}(\alpha, x) = 2\pi R_p^3 \frac{P^2(\alpha, x)}{E_c} e(x) \quad (6)$$

where  $e(x)$  is dimensionless. Now let us stress the fact that the equality of the work done by the pressure in the variation of volume of the cavity and the stored elastic energy leads to:

$$\Delta v(x) = 3e(x) \quad (7)$$

Thanks to this, the determination of  $e(x)$  by FEA (see Figure 2 Left) gives us access to the stored elastic energies and the pressure (Equations (3-7)), for any Young's moduli of the cement paste and the aggregate and bulk modulus of the gel (but at fixed Poisson's ratios since  $e$  in fact also depends on  $\nu_c$ ).

The closed form solution for the uncracked situation gives us the uncracked value of the dimensionless energy:  $e(0) = (1 + \nu_c)/2$ .

## 5 THERMODYNAMIC STUDY OF CRACK INITIATION AND PROPAGATION

We decided to create the cracks from scratch, avoiding the question of the initial crack size. When there is enough energy stored in the system to create a crack of given surface, it can happen. We consider this necessary condition as sufficient and therefore, we use the equality of released energy and dissipated energy as a crack initiation criterion.

The dissipated energy during crack growth is defined as:

$$E^{diss}(x) = G_c \pi R_p^2 \left[ (1+x)^2 - 1 \right] \quad (8)$$

which is the product of the surface of the crack by a material parameter  $G_c$  (surface fracture energy). This energy is to be compared to the energy released during the creation of a crack of size  $x$  which we define as:

$$E^{rel}(\alpha, x) = E_{total}^{el}(\alpha, 0) - E_{total}^{el}(\alpha, x) \quad (9)$$

where, taking advantage of Equations (4), (5), and (6):

$$E_{total}^{el}(\alpha, x) = 2\pi R_p^3 \left[ \frac{\delta [1 - (1-\alpha)]^3}{3K_{gel}} + (1-\alpha)^3 \frac{1-2\nu_a}{E_a} + e(x) \right] P^2(\alpha, x) \quad (10)$$

Then, as can be seen on Figure 2. Right, while the loading parameter  $\alpha$  increases, the inequality  $E^{rel}(\alpha, x) \geq E^{diss}(x)$  is more and more likely to be verified for a given relative crack size  $x$ . When this inequality is first verified, the two energies as functions of the relative crack size are also tangent. Therefore, for a given aggregate size and set of material constants, the initially uncracked system instantaneously jumps to a first crack of size  $x_c$ , created at the attack degree  $\alpha_c$  which verifies:

$$\begin{cases} E^{rel}(\alpha_c(R_p), x_c(R_p)) = E^{diss}(x_c(R_p)) \\ \frac{\partial E^{rel}}{\partial x}(\alpha_c(R_p), x_c(R_p)) = \frac{dE^{diss}}{dx}(x_c(R_p)) \end{cases} \quad (11)$$

Then, the propagation of the crack follows the energy rate equation (Equation 12) which gives the relative crack size corresponding to each attack degree  $x(\alpha)$ .

$$\frac{\partial E^{rel}}{\partial x}(\alpha, x) = \frac{dE^{diss}}{dx}(x) \quad (12)$$

Now, starting from the knowledge of the volume fraction of each aggregate size at the macroscopic level and that of the pressure and the crack size for each aggregate size at the microscopic level, we need to define a macroscopic deformation.

## 6 MACROSCOPIC DEFORMATION

This part explains our method to compute macroscopic deformations starting from the crack volume obtained for each aggregate size from crack propagation under gel pressure as explained above. First we consider one aggregate in infinite cement paste and a domain  $\Omega$  including the aggregate. Its current state is described by the couple  $(x, P(\alpha, x))$ . Let us consider  $\underline{\underline{U}} = \int_{\partial\Omega} (\underline{u} \otimes \underline{n})^s dS$  where  $\underline{u}$  is the displacement field,  $\underline{n}$  the outward normal to  $\Omega$ , and  $s$  is used to take the symmetric part of the tensor. Takana-Mori's theorem [5] states that if  $\Omega$  is a sphere,  $\underline{\underline{U}}$  does not depend on its size and position as long as it includes the cavity.

Suppose there is only one aggregate size  $R_p$  representing a volume fraction  $f$  throughout the concrete. Then the sphere  $\Omega$  has to be chosen such that  $f = \frac{4}{3}\pi R_p^3 / |\Omega|$  to make sure that the volume fraction of the aggregate in the domain used to compute the average deformation due to the attack is equal to the volume fraction in the concrete considered. Finally we define the macroscopic deformation  $\underline{\underline{E}} = |\Omega|^{-1} \underline{\underline{U}} = \left[ f / \left( \frac{4}{3}\pi R_p^3 \right) \right] \underline{\underline{U}}$ .

We now need to take into account the aggregate size distribution. To each aggregate size  $R_{p,i}$  corresponds a volume fraction  $f_i$  in the concrete. Each aggregate class contributes to the macroscopic

deformation through a partial average deformation defined as  $\underline{\underline{E}}_i = \left[ f_i / \left( \frac{4}{3} \pi R_{p,i}^3 \right) \right] \underline{\underline{U}}_i$  where  $\underline{\underline{U}}_i$  is computed relatively to the aggregate of size  $R_{p,i}$ , a crack of size  $x_i$ , and a pressure  $P_i = P(\alpha_i, x_i)$ . The macroscopic deformation is then defined as  $\underline{\underline{E}} = \sum_{\substack{\text{aggregate} \\ \text{size } i}} \underline{\underline{E}}_i$ .

A corollary of Takana-Mori's theorem [5] for which we will give our own proof in a later paper relates the volumetric part of  $\underline{\underline{U}}_i$  to the dimensionless stored elastic energy in the cement paste surrounding the considered aggregate (defined in Equation 6 and related to the volume variation of the cavity through Equation 7, and computed by FEA) through the expression:

$$tr \underline{\underline{U}}_i = \frac{R_{p,i}^3 P(\alpha_i, x_i)}{E_c} \frac{4\pi}{3} \frac{1+\nu_c}{1-\nu_c} [1-2\nu_c + e(x_i)] \quad (14)$$

Equations 13 and 14 finally give us access to the macroscopic volumetric deformation:

$$tr \underline{\underline{E}} = \sum_{\substack{\text{aggregate} \\ \text{size } i}} f_i \frac{P(\alpha_i, x_i)}{E_c} \frac{1+\nu_c}{1-\nu_c} [1-2\nu_c + e(x_i)] \quad (15)$$

## 7 NUMERICAL RESULTS: EFFECT OF AGGREGATE SIZE

In this section, we want to apply our model to see if it allows explaining the expansions observed in ASR. To do so, we need some reliable values of the different parameters involved. Recalling that  $x_{\text{inf}}$  depends on  $E_c/K_{\text{gel}}$ ,  $\nu_c$ ,  $\delta$ ,  $E_c/E_a$ ,  $\nu_a$  and  $R_{\text{inf}}$  depends on these quantities plus  $G_c/E_c$ , we are going to study the influence of the aggregate radius  $R_p$ , the ratio of gel to aggregate volume  $\delta$  and the gel bulk modulus  $K_{\text{gel}}$  on first cracking and crack length at full attack. The other parameters which vary less or are easier to determine for a given concrete will be set to the values of  $E_c = 20\text{GPa}$ ,  $E_g = 60\text{GPa}$ ,  $\nu_c = \nu_a = 0.25$ ,  $G_c = 40\text{J.m}^{-2}$  (see [6]).

### 7.1 Study of the first and final cracking

*Smallest aggregate size that can lead to the fracture of the cement paste:  $R_{\text{inf}}$*

Let us first study how  $R_{\text{inf}}$  varies when changing  $\delta$  and  $K_{\text{gel}}$ . It is important to study this Behaviour, because the swelling of the gel and its compressibility are not well known. In 2005, Phair claimed to publish the first measurements of ASR gel bulk modulus [8]. The measured values are of the order of 10 GPa. His experimental set-up seems to measure the undrained bulk modulus of the gel. The gel is porous, so its drained modulus might be very different, and it seems that since gel creation is very slow, water movements have time to occur during the pressure build-up and hence, the drained bulk modulus of the gel is what we need in our model. Moreover, the different sorts of gel that are produced in different chemical environments might also have very different and time-dependant values of these parameters as observed on

the storage and loss moduli of synthetic alkaline-calcium silica gels by Gaboriaud [9]. Since the swelling of the gel is often explained by water absorption, they might also differ significantly according to the saturation degree of the concrete. Hence, in a predictive model, they would probably have to be determined by an optimization procedure. We can see (Figure 3 Left) that increasing the coefficient of expansion  $\delta$  or the compressibility  $K_{gel}$  decreases the radius of the smallest aggregate that leads to fracture initiation. When  $\delta \longrightarrow 1$ , cracking becomes impossible, as well as when the gel is too soft ( $K_{gel} \longrightarrow 0$ ).

#### *First and final crack sizes varying $R_p$ , $K_{gel}$ , and $\delta$*

These three parameters are very important to determine the crack initiation and propagation around an aggregate correctly as can be seen on Figures 3 Right, 4 Left, and 4 Right, where we represent the first ( $x_c$ ) and final ( $x_{max}$ ) crack size when changing these parameters. The unchanged parameters in each case have the values  $K_{gel} = 1GPa$ ,  $\delta = 1.03$  and  $R_p = 1.5mm$ .

A similar behaviour observed varying  $\delta$ ,  $K_{gel}$ , or  $R_p$  (Figures 3 Right, 4 Left, and 4 Right): for small values, cracking is impossible even at full attack ( $\alpha = 1$ ) then, cracking becomes possible. The initial crack size ( $x_c$ ) further decreases, and the final crack size ( $x_{max}$ ) increases when increasing  $\delta$ ,  $K_{gel}$ , or  $R_p$ .

#### **7.2 Study of the complete evolution of a crack, influence of an initial flaw**

Once the crack is initiated, we can follow its evolution when degree of attack increases further. We still are in the approximation of a spherical attack, even if the presence of the crack increasingly leads this assumption to become wrong. In this article we mostly study the situation where there is no initial flaw (all crack sizes are initially zero), but we will show in this section a few evolutions of cracks with non-zero initial size. For one aggregate size, here  $R_p = 1.5mm$ , we try different initial relative crack sizes, ranging from zero to the aggregate size. We represent the crack size  $x$ , the pressure  $P$  and the volumetric deformation  $tr \underline{\underline{E}}$  as functions of  $\delta$ , the relative attack depth. We consider 10 different cases: initial flaws ranging from  $x = 0$  (blue dotted line) to  $x = 1$ . We see the existence of a critical flaw size above which no jump in crack size occurs anymore (Figure 5 Left), here approximately  $x = 0.4$ . Other simulations showed that this critical flaw size depends on the aggregate size. The smaller the flaw, the bigger the pressure has to be to propagate a crack (Figure 5 Right). If cracking occurs, expansion is ultimately the same, but a significantly different behaviour is observed at the beginning (Figure 6). At early times, expansion is roughly doubled if a large flaw is considered, compared to the case with no initial crack. This effect seems very important here, when we only have one aggregate size involved.

## **8 NUMERICAL RESULTS: FIT TO AN EXPERIMENTAL EXPANSION CURVE**

We must keep in mind that usual ASR expansion curves have a sigmoid shape characterized by a latency time, a characteristic time and an asymptotic expansion value. We can wonder if our model can be used to fit such an expansion curve, even if it would not mean that the model is predictive in any way. We use an expansion test found in Riche's PhD thesis [9]. In this test on  $7*7*28cm$  prismatic samples, high

alkalinity cement is used. The reactive aggregate is flint ranging from  $5mm$  to  $20mm$ , for a total volume fraction of around 22.5%, while a volume fraction of 45% is occupied by a limestone non-reactive aggregate. We have two choices to explain the end of expansion: scarcity of aggregates, or scarcity of chemical products to attack the aggregates. Even if the formulation has a very high equivalent alkali content (around  $5\text{ kg}/\text{m}^3$ ), it is unlikely that it is sufficient to attack  $500\text{ kg}/\text{m}^3$  of reactive aggregates. Hence, we assume that here the expansion stops even if some aggregates are left. Following Bažant [2], we assume that at a given time, the attack depth is the same for all aggregate sizes. We then identify a function  $r(t)$  which links the attack depth to the physical time through  $\alpha(R_p, t) = r(t)/R_p$ . Thanks to this deformation of the time scale, we obtain a realistic but artificial expansion profile (Figure 7). The values used here are  $K_{gel} = 1\text{ GPa}$  and  $\delta = 1.05$ . These values were chosen to achieve a sufficient asymptotic deformation.

More physics is needed to explain the shape of this curve. To explain why expansion starts slowly, at least two options. The first is that diffusion of the chemical species attacking the aggregate take time. The second one is that the gel created in the beginning of the reaction does not contribute to cracking but fills the porosity of concrete. These two points will be studied as next steps of our research, starting with a study of the rheological properties of synthetic gels.

## 9 CONCLUSION

A model for Alkali-reaction was proposed. It is a strictly mechanical model, partly inspired by that of Bažant [2]. Aggregates are supposed to behave as if they were embedded in an infinite cement paste. The chemical attack is modelled by progressively replacing each aggregate by a less dense gel. The subsequent pressure increase leads to cracking of the surrounding cement paste. The initiation and propagation of the crack are governed by energy balance only. An interesting behaviour is observed. Aggregates of different sizes have a different impact on the overall expansion. First of all, even fully attacked, smaller aggregates are shown not to lead to any cracking of the cement paste. The pressure reached for these aggregates is important, but according to our energy criterion the cement paste can take such pressures when there is no (or a very small) initial crack, because the elastic energy stored in the cement paste around a small aggregate is not sufficient to provide the energy needed to create a crack. Finally we apply the model to try to reproduce the expansion obtained in an experiment lead on a concrete specimen by Riche [9] and manage to identify the attack rate, which is one of the inputs of our models, to reproduce the usual sigmoid expansion curve.

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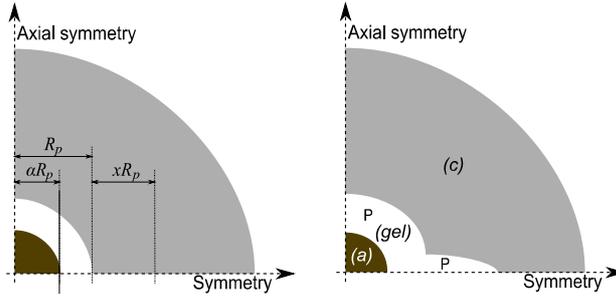


Figure 1. Undeformed and deformed configurations. (a): remaining aggregate. (c): cement paste.

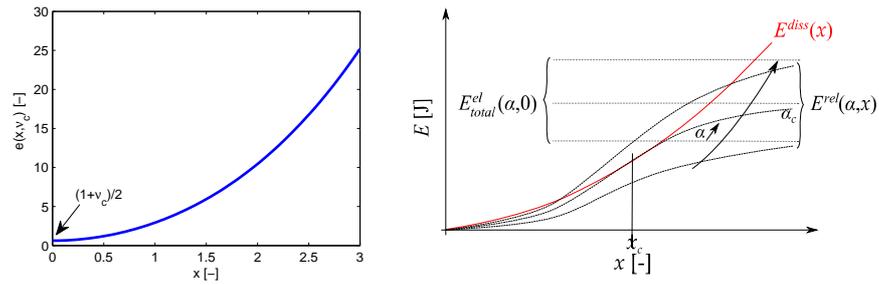


Figure 2. Left: Dimensionless stored elastic energy, determined by FEA for different crack sizes  $x$ . Right: Schematic representation of the dissipated and released energies as functions of the relative crack size  $x$  while increasing the attack degree  $\alpha$

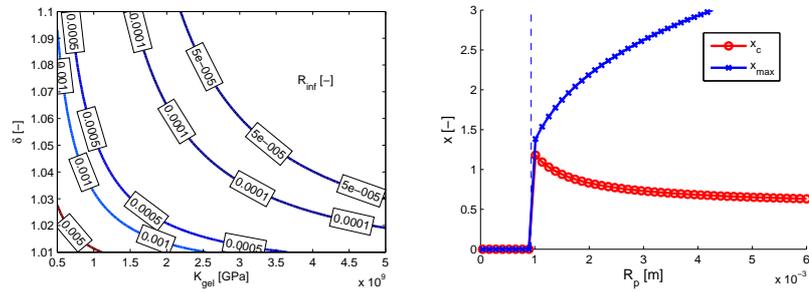


Figure 3. Left: Minimum radius to crack the cement paste:  $R_{inf}(\delta, K_{gel})$ . Right: Relative initial ( $x_c$ ) and final ( $x_{max}$ ) relative crack size as a function of aggregate size.  $K_{gel} = 1GPa$  and  $\delta = 1.03$

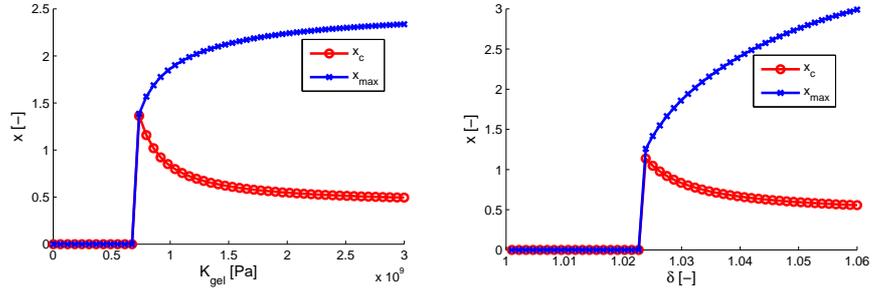


Figure 4. Left: Relative initial ( $x_c$ ) and final ( $x_{\max}$ ) relative crack size as a function of the gel bulk modulus.  $\delta = 1.03$  and  $R_p = 1.5 \text{ mm}$ . Right: Relative initial ( $x_c$ ) and final ( $x_{\max}$ ) relative crack size as a function of the ratio of gel volume to aggregate volume.  $K_{gel} = 1 \text{ GPa}$  and  $R_p = 1.5 \text{ mm}$ .

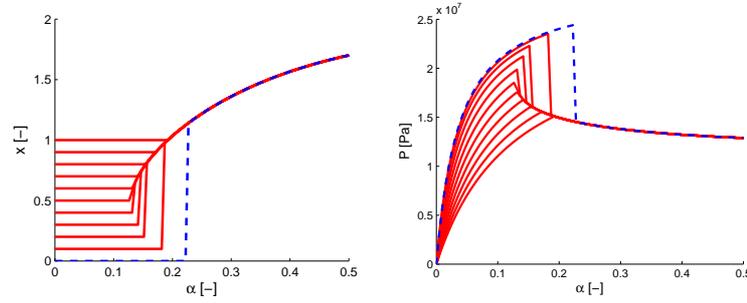


Figure 5. Left: Relative crack size  $x(\alpha)$  (initial flaws ranging from  $x = 0$  (dotted line) to  $x = 1$ ).  $K_{gel} = 1 \text{ GPa}$ ,  $\delta = 1.03$ , and  $R_p = 1.5 \text{ mm}$ . Right: Pressure  $P(\alpha)$  (same conditions as Fig. 5. Left)

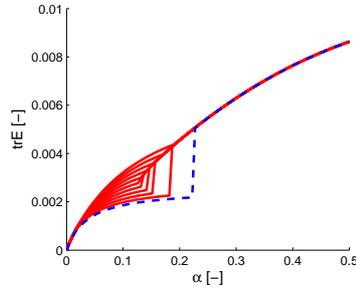


Figure 6. Volumetric deformation  $tr \underline{\underline{E}}(\alpha)$  (same conditions as Fig. 5. Left)

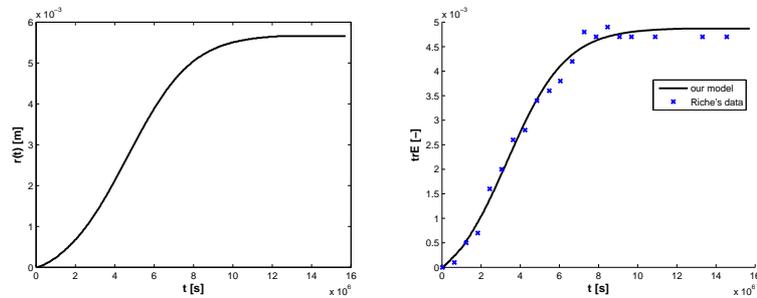


Figure 7. Optimized volumetric expansion profile.