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Simplified model for the transport of Alkali-Silica Reaction gel in concrete porosity

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Abstract
In this work, we focus on estimating the proportion of gel created by Alkali-Silica Reaction (ASR) in concrete that can enter the Interface Transition Zone (ITZ) surrounding an aggregate at a given pressure. We consider the gel as a non-Newtonian fluid following a Darcy law with a threshold pressure gradient while invading a porous medium. We show that at equilibrium, the mass of gel that penetrates in the ITZ can be considered as proportional to the pressure surrounding the aggregates while the ITZ is not full yet, and then constant once the ITZ is full. This is fairly consistent with the common use of a "connected porosity" in the models dealing with mechanical features of ASR to estimate the volume of gel lost in the porosity. We also study briefly the kinetics of this process and the influence of the gel compressibility.

1. Introduction
As an attempt to understand why, although the gel can penetrate in the porosity of concrete after Alkali-Silica Reaction (ASR), there still exists an important pressure around the aggregate that can lead to cement paste fracture, we wrote a very simple model accounting for the diffusion of an incompressible fluid following Darcy law with a threshold, around an attacked aggregate. This model is motivated by interesting results from (Gaboriaud et al. 2005) showing that synthetic gels very close to ASR gels have a threshold fluid behavior in bulk experiments, which might suggest that it is also the case when invading a porous medium. In many models of concrete degradation during ASR, a certain amount of gel is allowed to flow into the cement paste (Bažant and Steffens 2000; Multon et al. 2010; Charpin and Ehrlacher 2012; Charpin 2013). This phenomenon delays the pressure build-up in the porous cement paste surrounding the aggregates, and needs to be taken into account to correctly estimate damage or cracking, for certain kinds of reactive aggregates which induce significant cracking in the cement paste. Our paper aims at studying the invasion of the cement paste by the gel, in order to justify this approach.

2. Physical set up of the problem
Our goal in this article is to explain why it is relevant, while modeling cracking around aggregates submitted to ASR, to assume that there is a zone that a limited quantity of gel can easily invade. The existence of this buffer zone, which is mainly due to the presence of a high porosity zone around the aggregates called Interface Transition Zone (ITZ), has an
important impact on the pressure in the gel-filled cavity, hence on the possibilities of fracture. It is usually taken into account in mechanical models for ASR as a "connected porosity" length (Bažant and Steffens 2000; Multon et al. 2010; Charpin and Ehrlicher 2012). To explain this phenomenon, let us first set the equations needed to describe it and the first simplifications we make.

2.1. Solid porosity

First we account for the mass conservation of the gel in the porosity. The total porosity of the concrete is named $\phi_s$. The solid skeleton is assumed infinitely rigid. Therefore, the porosity $\phi_s$ is constant in time. It might however take different values in a thin region surrounding the aggregate (the ITZ) which is of high porosity and can act as a gel reservoir, and the rest of the cement paste, farther from the aggregate.

2.2. Gel compressibility

The volume fraction occupied by the gel is $\phi(x,t)$. It is such that $0 \leq \phi(x,t) \leq \phi_s$. The gel is a very special material. It is itself a porous material, and the matrix of this porous material also exhibits viscous relaxation (Gaboriaud et al. 2005), all these properties evolving with time, as the gel ages and changes chemical environment. Like other porous materials, its stiffness depends on the speed of loading, due to fluid movements inside the gel. Therefore, it is very difficult at the moment to choose an appropriate constitutive behavior for the gel. The main goal of this article is to study the effect of the flow threshold, therefore, we choose a very basic linear elastic law to model the gel elasticity. The gel density writes:

$$\rho(x,t) = \rho \left(1 + \frac{P(x,t)}{K}\right)$$

Where $\rho_0$ is the density at zero pressure, $P(x,t)$ is the pressure written as a function of space $x$ and time $t$, and $K$ is the bulk modulus of the gel.

2.3. Mass conservation

The poromechanics equations are written following the works of Coussy (2004). The volume flux of the gel is called $q$. The mass conservation equation therefore writes:

$$\frac{\partial (\rho(x,t)\phi(x,t))}{\partial t} + \text{div} (\rho(x,t)q(x,t)) = 0$$

2.4. Transport law

For the transport law, our idea is that if it was simply a Darcy law, all pressure gradients would equilibrate and the pressure would remain insufficient to crack the cement paste. We think the gel might have a threshold shear stress below which does not flow, as is reported for various polymeric gels. However, this is a microscopic property and its upscaling to a macro threshold requires a lot of information about the shape and size of the porosity of concrete, and is a very difficult task which requires large computations (Sochi 2010). For now we simply assume that this behaviour can be summed up assuming that below a given macroscopic pressure gradient, the gel does not flow (Protopas 1998). Above this gradient, we assume the flux to increase linearly with the pressure gradient. Therefore, we chose the following elementary transport law for the gel:
Where $k$ represents the permeability of the cement paste and $\tau_o$ the pressure gradient threshold, both different in the ITZ and the bulk cement paste. Both these quantities are mixed properties of the gel and the cement paste. Notice that we here assume that $k$ is independent of the gel saturation of the porous space, which is certainly not the case since there is a vast literature discussing the proper choices for the dependence of permeability on saturation (van Genuchten 1980). However, our main focus here will be on the equilibrium properties of the ingress of the gel in the porous medium, not on the kinetics.

3. Dimensionless system in the homogeneous spherical case

For now we assume that the cement paste is homogeneous, that is the porosity is constant and equal to $\phi_h$. The gel appears in a spherical cavity in the cement paste, which we assume to be at constant pressure $P_{imp}$ due to a chemical process creating gel. Here we are interested in the ingress of the gel in the cement paste, which is a phenomenon of relatively short distance compared to the aggregate size. Therefore, we neglect interactions between the various aggregates composing the concrete and assume the cavity where the gel is created is embedded in an infinite cement paste porous medium.

3.1. Use of spherical symmetry

Let us start assuming that a spherical cavity is filled with gel under a constant pressure $P_{imp}$. The surrounding cement paste is assumed homogeneous for now. The first simplification is due to the spherical symmetry. The mass conservation (Eq. 2) rewrites:

$$\frac{\partial (\rho (r,t) \phi (r,t))}{\partial t} + \frac{\partial (\rho (r,t) q (r,t))}{\partial r} + 2 \frac{\rho (r,t) q (r,t)}{r} = 0$$  \hspace{1cm} (4)

We also assume that due to the fact that all the gel comes from the cavity, the pressure decreases moving away from the cavity. Therefore the transport law (Eq. 3) simplifies to:

$$\begin{cases} 
\text{If } \frac{\nabla P}{\nabla P} < \tau_o, & q = 0 \\
\text{If } \frac{\nabla P}{\nabla P} > \tau_o, & q = k \left( \nabla P + \tau_o \right) 
\end{cases}$$  \hspace{1cm} (3)

$$\begin{cases} 
\nabla q = 0 \\
\nabla q = -k \nabla + \tau_o 
\end{cases}$$  \hspace{1cm} (5)

Which is also represented in Fig. 1.

Figure 1: Transport law

We then take the gel constitutive behaviour into account (Eq. 1) in the mass conservation equation (Eq. 4), and assume that we are in a saturated region, that is the volume fraction occupied by the gel $\phi$ is equal to the solid porosity $\phi_s$.
Now we need to replace the volume flux by its expression as a function of the pressure.

We assume for now that we are in the flow regime (that is $\frac{\partial P}{\partial r} \leq -\tau_0$), and we will check this condition. Using the expression of the flux corresponding to that regime (Eq. 5), we obtain the following differential equation on the pressure:

$$\frac{\partial P}{\partial t} = \frac{K}{\phi_s} \left[ \frac{\partial P}{\partial r} \left( 1 + \frac{P}{K} \right) + k \frac{\partial P}{\partial r} \left( \frac{\partial P}{\partial r} + \tau_o \right) + \frac{2}{r} \left( \frac{\partial P}{\partial r} + \tau_o \right) \left( 1 + \frac{P}{K} \right) \right]$$

(7)

This differential equation is valid in the saturated region, between the radius $R_p$, where the pressure is prescribed equal to $P_{mp}$, and the end of the saturated region, at radius $R_s(t)$. For any radius $r > R_s(t)$, the concrete is not saturated. Therefore, the pressure is zero, since the gel has enough space. Hence, considering the transport law in Eq. 5, there is no flow. We can then consider that there is a zero pressure imposed at radius $R_s(t)$, which is unknown, or consider that this zero pressure is applied at a remote radius $R_\infty$, and that between $R_t(t)$ and $R_\infty$ the flux is zero. In any case, it leads to consider that for $R_s(t)$ the gel volume fraction in zero $\phi = 0$ and that there is a jump to $\phi_s$ when the gel front, which position is $R_s(t)$, passes. Finally we get:

$$\begin{cases}
\frac{\partial P}{\partial t} |_{t=0} = 0, & \frac{\partial P}{\partial r} |_{t=0} = 0, \\
\frac{\partial P}{\partial r} |_{t=0} = 0, & P(R_\infty, t) = P_{mp} \\
P(R_s(t), t) = 0 \\
P(r > R_p, t) = 0
\end{cases}$$

(8)

### 3.2. Dimensionless problem

Now let us make this system dimensionless, by setting:

$$\mu = \frac{r}{R_p}, \nu = \frac{t}{\tau}, p = \frac{P_{mp}}{\tau_0 R_p}, \Pi(\mu, \nu) = \frac{P(r, t)}{\tau_0 R_p}, \mu_s(\nu) = \frac{R_s(t)}{R_p}$$

(9)

Where the characteristic time $\tau$ is yet unknown. Eq. 7 then becomes:

$$\frac{R_p^2 \phi_s}{K} \frac{\partial \Pi}{\partial \nu} = \frac{\partial \Pi}{\partial \mu} \left( 1 + \eta \Pi \right) + \eta \frac{\partial \Pi}{\partial \mu} \left( \frac{\partial \Pi}{\partial \mu} + 1 \right) + \frac{2}{\mu} \left( \frac{\partial \Pi}{\partial \mu} + 1 \right) \left( 1 + \eta \Pi \right)$$

(10)

Where $\eta = \frac{\tau_0 R_p}{K}$ is a dimensionless parameter comparing the pressure threshold to the gel bulk modulus. We would like the characteristic time not to depend on the gel compressibility, and we know (looking at Eq. 4) that when gel is incompressible, the time
dependency disappears of this equation. This leads us to set \( \tau = \frac{\phi R_p}{k \tau_0} \). Doing so, the full system becomes (writing the main equation in a more compact form):

\[
\begin{align*}
R_p \kappa \phi \tau & = 0 \\
\phi \tau & - \phi \tau_0
\end{align*}
\]

3.3. Equilibrium

It can be studied looking at the equations obtained by making fluxes and time derivatives zero in Eq. 11. Since the evolution of the pressure profile is blocked when we reach the condition \( \tau = 1 \), the pressure gradient remains such that \( \frac{\partial \Pi}{\partial \mu} = -1 \). Then the final pressure state is a linear and satisfies the boundary conditions in pressure at \( \mu = 1 \). Therefore in the simple case where the porosity is uniform, the equilibrium dimensionless pressure writes:

\[
( ) = p ( 1 ) = (s)
\]

4. Resolution in the incompressible gel case

4.1. Non saturated case

If the porosity in a given point is not fully occupied by gel \( (\phi < \phi_s) \), no pressure build-up occurs \( P = 0 \). Except close to the interface with the zone which is saturated with fluid, if the vicinity of a point isn’t saturated, the pressure is zero close to this point, and so is the pressure gradient. Therefore, owing to the mass conservation (Eq. 4), the volume fraction of gel doesn’t vary. So since the concrete is initially filled with its interstitial solution, \( \phi_s = 0 \) until the gel front passes, which means that the volume fraction of gel jumps to reach the porosity \( \phi_s \).

4.2. Saturated region

The saturated region can also be divided in a region where flow occurs and a region with no flow.

4.2.1. Flowing regime

In this region where \( \frac{\partial P}{\partial r} \leq -\tau_0 \), the variation of porosity writes (from 4 and 5):

\[
\frac{\partial \phi(r,t)}{\partial r} = k \left[ \frac{\partial^2 P}{\partial r^2} + \frac{2}{r} \left( \frac{\partial P}{\partial r} + \tau_0 \right) \right]
\]

Since the gel fills the porosity of the porous medium which is assumed to be undeformable, its volume fraction is constant. that is we have the following ODE on the gel pressure:
\[
\frac{\partial^2 P}{\partial r^2} + 2 \left( \frac{\partial P}{r \partial r} + \tau_0 \right) = 0
\]  
(14)

Which can be found in its dimensionless form from Eq. 11 when \( \eta \to 0 \):

\[
\frac{\partial^2 \Pi}{\partial \mu^2} + 2 \left( \frac{\partial \Pi}{\mu \partial \mu} + 1 \right) = 0
\]  
(15)

This equation is valid when \( \mu \to +1 \) \( \eta \), and can be rewritten:

\[
\frac{\partial}{\partial \mu} \left[ \mu^2 \left( \frac{\partial \Pi}{\partial \mu} + 1 \right) \right] = 0
\]  
(16)

Then taking into account the boundary conditions \( \Pi(1, \nu) = p \) and \( \Pi(\mu_*(\nu), \nu) = 0 \), we obtain the pressure profile depending on the position of the front (see Fig. 2):

\[
\Pi(\mu, \nu) = \mu_*(\nu) - \mu + \left( p + 1 - \mu_*(\nu) \right) \frac{1 - \mu}{\mu_*}(\nu)
\]  
(17)

Which is valid when \( \mu_*(\nu) < p + 1 \).

Figure 2: Dimensionless pressure profile at a given time

4.2.2. No flow regime

When this condition is reached, which happens everywhere at the same time, since it does not depend on \( \mu \), the pressure profile is simply linear and consistent with the pressure gradient threshold:

\[
\Pi(\mu, \nu) = \mu_*(\nu) - \mu
\]  
(18)

At this point there is no more flow, so \( \mu_*(\nu) = \mu_*^\infty \), which we find using the first boundary condition: \( \mu_*^\infty = p + 1 \). The equilibrium pressure profile then writes:

\[
\Pi(\mu, \nu) = p + 1 - \mu
\]  
(19)

4.2.3. Determination of the front position

Now we want to study the evolution of the pressure and flux before the gel stops flowing in the porosity. Since the pressure distribution is known explicitly as a function of \( R_s(t) \) or \( \mu_*(\nu) \) in the dimensionless variables, it simply amounts to finding the evolution of this radius. The equation on \( \mu_*(\nu) \) can be found by mass balance. Here we write the mass balance at the front itself. We could also use a global mass balance, equating the
mass of gel which has entered up to a given time to the mass of gel present in the porous medium. However, integration difficulties would arise.

First, let us write the volume flux at the front with respect to the dimensionless variables defined in (9):

\[
q(R_p \mu_s(v)) = -k \tau_0 \left( 1 + \frac{\partial \Pi}{\partial \mu_s(v)} \right) = k \tau_0 \frac{1}{\mu^2} \frac{p + 1 - \mu_s(v)}{1 - \frac{1}{\mu_s(v)}}
\]  \hspace{1cm} (20)

Then thinking that when the front passes at a given radius, the volume fraction of gel jumps from zero to \( \phi_s \), and that the speed of the interface is equal to \( R_s'(t) \), this volume flux is also equal to:

\[
q(R_p \mu_s(v)) = R_s'(t) \phi_s
\]  \hspace{1cm} (21)

We then end up, equating these two expressions of the volume flux at the front, with a differential equation on the position of the front:

\[
\frac{\tau k \tau_0}{R_p \phi_s} \frac{p + 1 - \mu_s(v)}{1 - \frac{1}{\mu_s(v)}} = \mu_s'(v) \mu_s'(v)
\]  \hspace{1cm} (22)

Finally the time constant \( \tau \) is chosen equal to \( \frac{R_p \phi_s}{k \tau_0} \), consistently to the choice made in the general case in section. We then end up with the differential equation:

\[
\mu_s'(v) = \frac{p + 1 - \mu_s(v)}{\mu_s^2(v) - \mu_s(v)}
\]  \hspace{1cm} (23)

which is valid at all times except at \( v = 0 \) where \( \mu_s(v = 0) = 1 \), which is the initial condition in the integration. This equation can easily be solved numerically, provided that we avoid the singularity at time \( v = 0 \) by choosing \( \mu_s(0) \) very slightly greater than one. It doesn’t change the result at all and allows the equation to be solved easily. We also see that there is only one parameter in this equation: \( p \). The condition of validity of this equation (flow regime) writes:

\[
\mu_s(v) < p + 1
\]  \hspace{1cm} (24)

Which is interesting since the limit value of the dimensionless radius if the saturated region is also given by \( \mu_s(v) < p + 1 \). That means that since \( \mu_s(v = 0) = 1 \), any positive pressure will make at least a little bit of gel enter. Let us plot the dimensionless front position as a function of time (see Fig. 3). Here we chose to apply a dimensionless pressure \( p = 1 \), which explains the asymptotic value of the dimensionless front position \( (p + 1 = 2) \), since as can be seen on Eq. 23 and was mentionned in section 4, \( \mu_s^* = p + 1 \). The evolution is very fast at the beginning (there is a vertical asymptote at \( v = 0 \)), and it then slows down gently.
Once the dimensionless front position has been determined, it is possible to fully reconstruct the pressure field in time, using Eq. 17. We therefore plot the evolution of pressure with time and space (see Fig. 4). We see that the boundary condition \( p = 1 \) is well respected and that the asymptotic pressure profile is linear as was underlined before (see Eq. 18).

Therefore, we have a clear idea of how the gel enters the porosity when assumed incompressible. Let us now address the question of the amount of gel at equilibrium.

5. Equilibrium of fluid injection in an ITZ-surrounded cavity

5.1. Pressure distribution

Let us now assume that due to a high porosity zone surrounding the aggregate, which is called ITZ, the threshold becomes non homogeneous. In the ITZ the porosity will be \( \phi_s \), the threshold \( \tau_s \), while in the rest of the cement paste they remain equal to \( \phi_i \), \( \tau_i \). Let us study the equilibrium state. We have seen that at equilibrium, the pressure gradient is everywhere equal to the threshold. We define the ratio of the thresholds as \( \lambda = \frac{\tau_s}{\tau_i} \).

Assuming the ITZ is of thickness \( R_p \epsilon \), let us write in dimensionless variables de expressions of the pressure profile, in the case where \( \mu_s \leq 1 + \epsilon \):

\[
\Pi^\ast (\mu) = p - \lambda (\mu - 1)
\]

And consistently,

\[
\mu_s^\ast = \frac{p}{\lambda} + 1
\]

While in the case where \( \mu_s = 1 + \epsilon \) (which also means \( p \geq \lambda \epsilon \)),

\[
\Pi^\ast (\mu) = p - \lambda (\mu - 1), \text{ If } \mu \leq 1 + \epsilon
\]

\[
\Pi^\ast (\mu) = p - (\mu - 1) + \epsilon (1 - \lambda), \text{ If } \mu \geq 1 + \epsilon
\]

And,

\[
\mu_s^\ast = p + 1 + \epsilon (1 - \lambda)
\]

The pressure profile at equilibrium is represented in two cases on Fig. 5.
Figure 5: Dimensionless pressure profiles at equilibrium for a low pressure such that \( p^1 \leq \lambda e \) (line) and a high pressure such that \( p^2 \geq \lambda e \) (dotted line).

5.2. Injected mass

Let us call \( M(t) \) the injected mass at time \( t \), and define the dimensionless injected mass as:

\[
\tilde{M}(\nu) = \frac{M(\tau\nu)}{4\pi R_p^3 \rho_o} \tag{30}
\]

The injected mass is computed by integration of the local density:

\[
M(\tau\nu) = 4\pi \int_{R_p}^{R_p} \int_{h_{\nu}}^{h_{\nu}} \phi(r) \rho(r) r^2 dr = 4\pi \rho_o R_p^3 \int_{\mu_p}^{\mu_p} \phi(\mu) (1 + \eta \Pi(\mu, \nu)) \mu^2 d\mu \tag{31}
\]

From this expression, and using Eqs. 25-29, the dimensionless injected mass can be computed. It is useful to take into account that \( e << 1 \) (the ITZ thickness is much smaller than the aggregate size), and that \( p << 1 \) (the pressure magnitude is such that the distances reached by the gel in the paste (if there is no ITZ) are small compared to the aggregate size \( R_p \)). Using these two relations, but keeping the dimensionless compressibility parameter \( \eta \) finite, we write the first order expansion of the injected mass in dimensionless pressure and thickness. It writes:

\[
\tilde{M}^\infty = 3\phi_a \frac{p}{\lambda} (1 + \eta p), \text{ If } \mu_p^\infty \leq 1 + e \tag{32}
\]

\[
\tilde{M} \approx 3 \phi_a (1 + \rho) + 3 \eta (\rho - (1 + \phi) (1 + \rho + \phi \overline{1})), \text{ If } \phi \leq 1 + e \tag{33}
\]

The terms proportional to \( \eta \) are all of second order in \( p \) and \( e \). However since the bulk modulus of the gel in ASR is not well known, we make no assumption about it. Finally, considering that the porosity of the cement paste \( \phi_a \) is much smaller than that of the ITZ, it suggests that it is a good approximation to take the following law for the mass of fluid that escapes the porosity to move into the fluid:

\[
\tilde{M}^\infty = 3\phi_a \min( e, \frac{p}{\lambda}) (1 + \eta p) \tag{34}
\]

This law expresses that the volume of gel that flows into the cement paste is first proportional to the pressure and then almost constant. If the compressibility of the gel is not neglected, the mass is first quadratic and then linear.

6. Discussion

Let us now briefly discuss the meaning of our result.
6.1. Relation of our result to ASR modelling

Our main result is the expression of the injected mass given in Eq. 34. Let us introduce a chemical time constant $\tau_c$ which represents the characteristic time for the production of the gel (taking into account of the kinetics of the chemical reaction and the transport time of the chemical species to the reaction site). Since ASR is very slow, it seems a good approximation to assume $\frac{\tau}{\tau_c} \ll 1$. In this case, we can also consider that we are always at equilibrium and that the pressure profiles labelled $\infty$ apply at all times. Therefore, the pressure $P_{\text{imp}}$ could be computed as the result as a total gel mass balance instead of being imposed, as long as it is an increasing function of time. This is basically what is done in ASR models developed by authors already cited (Bažant and Steffens 2000; Multon et al. 2010) and also the authors of this paper (Charpin and Ehrlacher 2012; Charpin 2013). We see that the most important parameter is the product $\phi_a e$ which gives the total ITZ volume accessible to the gel.

6.2. Effect of the parameters

We have introduced various geometrical and mechanical parameters, such as the aggregate size $R_p$, the pressure thresholds $\tau_a$ and $\tau_s$, the ITZ thickness $e$, the bulk modulus of the gel $K$, the permeability $k$ and the porosities $\phi_a$ and $\phi_s$. As stated previously, the important parameter is the product $\phi_a e$. Also, the characteristic time $\tau = \frac{R_p \phi_s}{k \tau_s}$ needs to be estimated, in order to be compared to the speed of creation of the products, as mentioned in the previous paragraph.

6.3. Limit of the description

ASR is generally considered to induce damage in the aggregates and in the cement paste, depending on the type of aggregates. Of course, our model which assumes spherical symmetry will not be meaningful anymore at the appearance of a crack. However, it would help determining the pressure build-up leading to the appearance of the crack. After the appearance of the crack, the pressure will drop, which we can assume will stop the gel progression in the porous cement paste. In energy-based fracture models as presented by the authors (Charpin and Ehrlacher 2012; Charpin 2013) this is of paramount importance.

Also, the threshold Darcy law we chose to represent the behaviour if the fluid should be questioned and motivated. There is a lot of work about the upscaling of the properties of fluids from the bulk to flow properties in porous medium (Sochi 2010), but we chose not to work on this very complicated issue in order to build a simple model.

7. Conclusion

In this article we deal with the ideal problem of the ingress of a threshold fluid in the porous medium surrounding a cavity. We address this problem in the case where the fluid is compressible and give the equations which would have to be solved numerically to have the full solution to this problem. We solve for the pressure profile in the case where the fluid is incompressible, which allows us to plot the evolution of the dimensionless front position and dimensionless pressure in this simple case.

Finally we show that at equilibrium, when an ITZ is present, it is interesting to consider
simple filling laws for the ITZ (mass proportional to the pressure before the ITZ is full and then constant), which is frequently done in mechanical modelling of ASR.

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Figure 5