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Long-Term Deformation and Failure of Concrete and Shale Structures

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ABSTRACT

Long-Term Deformation and Failure of Concrete and Shale Structures

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This thesis deals with modeling long-term deformation and failure of concrete and shale structures. First concrete structures are studied. For concrete structures, design lifetime of over a hundred years is required. Shrinkage, swelling, creep and Alkali-silica reaction are significant parts of the long-term deformation of concrete and are studied here. The aim is developing a comprehensive, physically based and computationally inexpensive model that be able to predict the deformation of concrete structures in all environmental conditions from hours after casting till several hundred years after that. This comprehensive model is lacking in literature and is necessary for designing sustainable structures.

The long-term deformations of concrete structures highly depend on hydration reaction evolution and drying of concrete. Thus, predictive and computationally inexpensive models for the hydration reaction and drying process of concrete structures are required. These models are lacking in the literature. Therefore, first, a new hydration model is developed which is able to predict the hydration evolution of concrete structures for hundred-year lifespan and beyond. Next, a new method is proposed that is able to predict the humidity
evolution of concrete structures during the drying process. Computationally, these models are sufficiently inexpensive to be used in finite element simulating of concrete structures. These models later are used to predict long-term deformations.

To predict autogenous shrinkage and swelling, a new paradigm is proposed. The hydration process causes permanent volume expansion of cement paste as a whole due to the growth of C-S-H shells around anhydrous cement grains. In addition, a new thermodynamic formulation of unsaturated poromechanics with capillary and adsorption is proposed. Later, to predict creep, microprestress-solidification theory (MPS) is modified. The original MPS theory predicts incorrectly the diffusion size effect on drying creep and the delay of drying creep behind drying shrinkage. Presented here is an extension named XMPS that overcomes both problems and also improves a few other features of the model response. To this end, different nanoscale and macroscale viscosities are distinguished. Finally, to predict the deformation and damage induced by ASR, a new diffusion-based and creep-based chemo-mechanical model is developed. Comparisons with the existing relevant experimental evidence validate all the proposed models.

In the last chapters, shale deformation is studied. Shale is the main constituents of the unconventional oil reservoirs. Hydraulic Fracturing technology, aka fracking, is the technology used to extract oil from these unconventional reservoirs. This technology has become highly developed and astonishingly successful recently and made USA energy independent. However, a consistent formulation of the associated fracture mechanics that would not conflict with some observations is still unavailable. The main issue is the significantly higher permeability of reservoirs respect to intact shale. This discrepancy currently is attributed to widely open natural fractures. In this study, first, we show these
natural fractures should be closed due to creep and calcification and later we show the branching of hydraulic cracks due to the presence of weak layers is the main reason for significantly higher permeability of reservoirs.
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CHAPTER 1

Introduction

A porous medium (or a porous material) is a material containing pores (voids). The skeletal portion of the material is often called the matrix or frame. The pores are typically filled with a fluid (liquid or gas). The skeletal material is usually solid.

Many natural substances such as rocks and soil (e.g., aquifers, petroleum reservoirs), zeolites, biological tissues (e.g., bones, wood, cork), and man-made materials such as cements and ceramics can be considered as porous media. Many of their important properties can only be rationalized by considering them to be porous media.

The concept of porous media is used in many areas of applied science and engineering: filtration, mechanics (acoustics, geomechanics, soil mechanics, rock mechanics), engineering (petroleum engineering, bio-remediation, construction engineering), geosciences (hydrogeology, petroleum geology, geophysics), biology and biophysics, material science, etc.

In this work, we study the long-term deformation and failure of the two very important and widely used porous media: Concrete and shale.

1.1. Long-Term Deformation and Failure of Concrete Structures

In this thesis, first the long-term deformation of concrete is analyzed. Concrete is used more than any other man-made material in the world. As of 2006, about 7.5 billion cubic meters of concrete are made each year, that is more than one cubic meter for every person on Earth [180]. Therefore, even some small improvements in the concrete
composition, and in structural analysis and design, may have considerable effect, not only economically but also on the environmental aspects and sustainability of infrastructure. For this end, we should be able to understand and predict the associate deformations of concrete structures and the factors that may cause significant damage and even failure of concrete made structures.

Concrete deforms in an extremely complex way. Like normal materials it has some short-term deformations that are usually dependent on external loads and some long term ones that can be either dependent or independent on external loads. These long-term deformations are significant for concrete structures since they can cause a huge amount of damage and even failure of structures.

In literature, several models can be found for predicting long-term deformation of concrete and some of them have incorporated in different design codes. However, these models have one or several of the following issues: First, Most of the currently available models are simplistic and may result in significant error in predicting the deformation; Second, these models usually work well for some specific environmental condition but result in significant error in other conditions; Third, these models are predicting the average behavior of cross-section. Unfortunately, the fact that, in case of environmental exposure, the cross-section is in a highly non-uniform state of stress and humidity, with nonlinear stress-strain behavior, cracking and damage, makes the prediction much more complicated and predicting the average behavior is not sufficient; Fourth, some of the models in literature are able to predict the long-term deformation of concrete with good accuracy but are computationally too expensive to be used in modeling real structures.
It should be mentioned that using currently available models to design structures can result in major disasters and failure of structures. An example of these disasters happened on the KB Bridge in Palau in 1996. This bridge suffered grossly excessive creep deflections, which reached 1.61 m (compared to design camber), and a prestress loss of about 50% \[60\]. This disastrous experience led to the creation of the RILEM Committee TC-MDC (Multi-Decade Creep). The Palau disaster led to the creation of RILEM Committee TC-MDC (Multi-Decade Creep, chaired by Bažant). An arduous search led to a collection of multi-decade deflection data of 69 large-span prestressed bridges from around the world, which suffered excessive or near-excessive deflections resulting in bridge closing or costly retrofit \[60, 31\]. The collection confirmed that all the design codes and recommendations severely underestimate the multi-decade creep and led to the development of a general creep and shrinkage prediction model, called model B4\[43\] (the fourth in a sequence of models developed at Northwestern since 1978). Although B4 model is a comprehensive model that is able to predict creep and shrinkage more accurate than other design codes, it has some issues in predicting autogenous shrinkage, drying creep and self-desiccation. In addition, like other design codes, it predicts the average behavior of the structure.

Considering all the above issues and dangers, developing a better model seems necessary. The proper solid mechanics approach is to formulate a constitutive law for a point of the homogenizing continuum. This requires the structure to be analyzed by 3D finite elements. Computationally, this is, of course, more demanding but has by now ceased to be an obstacle, thanks to the enormous increase in computer power. Developing such a model is the aim of this study. Note that the long-term deformation of concrete includes several different parts. In this study, we try to propose a comprehensive model that be
able to predict the most significant parts of these long-term deformations including creep, shrinkage and ASR-induced expansion.

Long-term deformations of concrete highly depend on hydration evolution and drying process of concrete. As a result, it is necessary to have good models to predict hydration evolution and drying process. However, it seems other works in literature have overlooked the complexity of predicting these two phenomena. Hydration reaction and drying process not only affect significantly almost all long-term deformations of concrete but also significantly affect each other. Hydration reaction has an effect on the drying of concrete due to self-desiccation and can decrease the relative humidity of concrete significantly (up to 0.65 in presence of silica-fume). In addition, hydration reaction changes the pore structure of concrete which changes the permeability and thus affect drying. On the other hand, drying control the rate of hydration reaction and as the material dries up hydration slows down and in low relative humidity values, it almost stops. Furthermore, when drying happens the relative humidity of the specimen is not uniform and varies in different points. Thus, the rate of hydration in the specimen is not uniform in exposed structures.

All the aforementioned facts show that the hydration reaction and drying process are completely dependent and can not be modeled separately. Therefore, we need a hydration model that be formulated based on relative humidity and have a drying model which consider the effect of self-desiccation and change of pore structure due to hydration. Unfortunately, both of these models are lacking in the literature and thus using currently available models for hydration and drying can cause a considerable error. Therefore, the first chapters of this study are dedicated to developing new models for hydration reaction
and drying of concrete. These models will be used in later chapters to predict long-term deformation of concrete structures.

The first long-term deformation that will be analyzed in this study is shrinkage. Total shrinkage of concrete includes two part: 1. autogenous shrinkage and 2. drying shrinkage. In older studies, the self-desiccation and autogenous shrinkage were generally ignored since they were negligible in old concretes with high water-cement ratios or no admixtures, or both. Recently, though, the trend toward high performance concretes raises the importance of the evolution of hydration for predicting the creep, shrinkage and self-desiccation, especially long-term.

As known since 1887, the volume of cement hydration products is slightly smaller than the original volume of cement and water (chemical shrinkage). Nevertheless, this does not indicate that the hydration reaction results in contraction of the concrete and cement paste. In this study, a new paradigm is used which considers the opposite to be true for the entire lifetime of porous cement paste as a whole. The hydration process causes permanent volume expansion of the porous cement paste as a whole due to the growth of C-S-H shells around anhydrous cement grains which forces the neighbors apart, while the volume reduction of hydration products contributes to porosity. Additional expansion can happen due to the growth of ettringite and portlandite crystals. On the material scale, the expansion always dominates over the contraction, i.e., the hydration per se is, in the bulk, always and permanently expansive, while the source of all of the observed shrinkage, both autogenous and drying, is the compressive elastic or viscoelastic strain in the solid skeleton caused by a decrease of chemical potential of pore water, along with the associated decrease in pore relative humidity.
The second part of long-term deformation that is studied is the creep of concrete. The creep of concrete, which originates from the calcium silicate hydrates (C-S-H) in the hardened Portland cement paste, is fundamentally different from the creep of metals and polymers. Unlike the creep of metals, it occurs at all stress levels. The creep can cause the irreversible deformations that can seriously compromise the structural integrity, serviceability and ultimately the sustainability of concrete buildings and can cause huge disasters.

The established model for point-wise constitutive law of concrete exhibiting shrinkage and creep with aging has been Bažant’s (1997) microprestress solidification (MPS) theory [30]. Although the MPS model can lead to good results on sealed conditions, some researches have shown the predicted results for the cases that structure is exposed to drying are not good enough. Therefore, the aim of this study is to modify MPS model in the way that be able to predict drying creep correctly. In this thesis, a new extension named XMPS is presented that overcomes both problems and also improves a few other features of the model response. To this end, different nanoscale and macroscale viscosities are distinguished. The aforementioned incorrect predictions are overcome by the dependence of the macroscale viscosity on the rate of pore humidity change, which is a new feature inspired by recent molecular dynamics (MD) simulations of a molecular layer of water moving between two parallel sliding calcium-silicate-hydrate (C-S-H) sheets.

The final long-term deformation that will be discussed here is ASR-induced expansion. The excessive expansion and disintegration of some concrete made of cements of a relatively high alkali content and of certain silica-containing aggregates can be explained
by the so-called alkali–silica reaction (ASR). The alkali-silica reaction (ASR) afflicts mineral aggregates in concrete if they contain imperfectly crystalline silica. The reaction produces a gel that can imbibe an enormous amount of water. The swelling that results, typically after several months to many years or even decades, is often causing severe strength degradation and fracturing in concrete structures. Since drying arrests the ASR, the worst damage usually occurs in massive structures such as dams, large bridges, and nuclear power plant structures. To prevent ASR-induced damage is one important goal of sustainable design.

The objective of this work is to develop a comprehensive continuum model capable of capturing the main physical aspects of ASR damage, including 1) the delay of ASR due to diffusion of ASR gel into the pores and expanding cracks within and very near aggregate grains; 2) fracturing of the solid framework of concrete as a two-phase medium caused by fluid expansion in the pores and cracks; 3) oriented character of damage; 4) mitigation by creep (with chemical aging) of the stresses created by gel expansion 5) effect of alkali content on ASR induced damage; 6) effect of temperature on ASR kinetics and ASR-induced expansion; and 7) control of the ASR process by variation of pore humidity due to self-desiccation or external drying.

1.2. Long-Term Deformation and Failure of shale Structures and Their Effects on Hydraulic Fracturing

In the last two chapters of this study, the deformation of shale layers is studied. Shale is the main constituent of unconventional oil reservoirs. The permeability of shale is very low thus in order to have an economically profitable production of oil and gas a process
named hydraulic fracturing is used. Fracking is shorthand for hydraulic fracturing, a type of drilling that has been used commercially for 65 years. Today, advanced hydraulic fracturing, employing cutting-edge technologies, is mostly responsible for surging U.S. oil and natural gas production. Safe hydraulic fracturing is the biggest single reason America is having an energy revolution right now, one that has changed the U.S. energy picture from scarcity to abundance. Fracking is letting the U.S. tap vast oil and natural gas reserves that previously were locked away in shale and other tight-rock formations. Up to 95 percent of natural gas wells drilled in the next decade will require hydraulic fracturing. Hydraulic fracturing also is being used to stimulate new production from older wells. Currently, the most knowledge regarding fracking is empirical and there is not much solid scientific understanding about what is happening down in the shale (3 km below the surface) which makes fracking feasible. Therefore, it is necessary to have a model that can simulate hydraulic fracturing and can help the industry to understand the process better and perform fracking in a more optimized way. It should be noted that, currently, in the fracking industry, they can just extract about 5% of gas in shale and make a small improvement in this process can help U.S. Energy to become more independent.

The existing commercial programs for simulation of hydraulic fracturing (aka fracking, or frac) of gas (or oil) shale predict parallel vertical cracks to spread in vertical parallel planes, with no lateral branching. These cracks emanate from the perforation clusters on the horizontal wellbore casing, typically spaced 10 m apart or more. For such a large spacing, the rate of gas production observed at the wellhead can be explained only upon making the hypothesis that the large-scale (or regional) permeability of shale is (even at 3km depth) about 10,000 times higher than the gas permeability of shale measured in the
lab on drilled (non-dried) shale cores under confining pressures corresponding to shale at the depth of about 3km. In literature, this high permeability is attributed to widely open natural fractures. In this study, the feasibility of having widely open natural fractures is studied. Especially, we study the possibility of closure of these fractures due to primary and secondary creep over the life of shale (million years).

In the last chapter of this thesis, a new physical model to simulate the hydraulic fracturing process is developed. The aim is understanding the crack growth and possibility of the branching of hydraulic cracks. Especially, the effect of weak layers on the growth of cracks is studied. This effect has usually neglected in other models.
CHAPTER 2

Cement Hydration from Hours to a Century Controlled by Diffusion through Barrier Shells of C-S-H

The hydration of cement is an extremely complex reaction involving chemo-physical phenomena spanning from a nanometer to micrometers in length and from seconds to decades in time. Aside from creep, the hydration is a crucial process for heat generation, strength development, self-desiccation, and autogenous shrinkage. Developing a realistic model is essential for understanding and control of all these phenomena. Although a few good models for cement hydration exist, they have some limitations. They do not take into account the complete range of variation of pore relative humidity and temperature and apply over durations limited from up a few months to up to about a year. The best ones are computationally too intensive for use in finite element programs. However, recent tests of autogenous shrinkage and swelling in water imply that the hydration may continue, at a decaying rate, for decades, provided that a not too low relative pore humidity persists for a long time, as expected for the cores of thick concrete structural members. Therefore, and because design lifetimes of over a hundred years are required for large concrete structures, in this chapter a new hydration model for a hundred-year lifespan is developed. Here in opposite to the most of available models in the literature, hydration is considered as long-lived phenomena which can continue for decades or centuries as long as its humidity remains above a critical relative humidity. This critical relative humidity,
Based on experimental results, can be assumed around 0.75. However, in presence of some admixtures like silica-fume this critical humidity can be much less. Finally, it should be mentioned that in the developed model, relative humidity is considered as the driving force of hydration process thus this model can be used for any environmental condition and is not restrained to sealed or immersed condition.

2.1. Introduction

Bridges, super-tall buildings and other large concrete structures are supposed to be designed for lifetimes in excess of hundred years. However, much shorter lifetimes have often been experienced. This is documented by a Northwestern University database [45, 149] of 71 large-span prestressed bridges that developed seriously excessive deflections with concomitant damage. It was shown that the major cause was the design based on obsolete and inadequate prediction models for multi-decade creep [40, 60].

Although a significant progress toward developing a more realistic model has been made [26, 266], one aspect that needs a better mathematical model is the aging of concrete due to hydration. Since the creep is strongly affected by aging, and the aging is caused by solidification due to cement hydration, what is especially needed is a model that can predict the evolution of hydration for a hundred-year lifetime, and do so in a simple enough computationally effective way. To this end, we refine and extend the model presented in 2015 at ConCreep-10 see [23].

It is often thought that cement hydration is not a long-lived phenomenon. But that is true only if concrete dries up. In the cores of massive walls, high pore humidity may persist for decades even in structures exposed to dry environment. What makes the hydration
process long-lived and progressively slower is that, after the first day, the anhydrous cement grains become enveloped in contiguous shells of cement hydrate. The shells are highly impermeable and allow only a very slow transport of water toward the anhydrous grain interface.

Therefore, the basic idea of the present model (whose outline was recently presented in ConCreep-10 proceedings [23]) is that the evolution of hydration is controlled by water diffusion through barrier shells of hydrated cement surrounding the anhydrous remnants of cement grains. Although contiguous barrier shells exist only after the first day of hydration, it will be convenient and computationally effective to introduce a hypothetical equivalent barrier shell model even for the first day.

The hydration of the cement is an extremely complex reaction involving chemo-physical phenomena on spanning from a nanometer to micrometers in length and from seconds to decades in time. Aside from creep, the hydration is a crucial process for heat generation, strength development, self desiccation and autogenous shrinkage. Developing a realistic model is essential for understanding and control of all these phenomena. The literature is large. The main works include, e.g., Jennings and Johnson [157]; Bentz and Garboczi [68]; Van Breugel [263] (whose model is known as HYMOSTRUC); Navi and Pignat [199]; Bentz [66] (whose model is known as CEMHYD3D); Maekawa et al. [184]; and Lin and Meyer [179].

Although some of the existing models are able to predict short- and mid-term hydration with sufficient accuracy, they consider either no physics or only the physical processes during the first few hours after mixing. Besides they are computationally too demanding for use in finite element programs for structural analysis. Also, the existing hydration
models do not calculate the decrease of pore relative humidity caused by hydration, i.e., the self-desiccation, although this phenomenon is critical for understanding and modeling of autogenous shrinkage and drying creep, especially long-term [43, 30, 61, 211]. In older studies, the self-desiccation and autogenous shrinkage were generally ignored since they were negligible in old concretes with high water-cement ratios or no admixtures, or both. Recently, though, the trend toward high performance concretes raises the importance of the evolution of hydration for predicting the creep, shrinkage and self-desiccation, especially long-term.

Therefore, the present study aims to develop a new and complete hydration model. The goal is not only to predict the evolution of cement hydration with heat generation, but also to provide underpinning in the physics of hydration and achieve a model that would be credible for predicting century-long hydration with its effect on the pore humidity evolution. To this end, the model is based on the water diffusion through the aforementioned C-S-H barriers and conveniently considers the pore humidity (or relative vapor pressure) as the driving force controlling the hydration rate. Simplicity, computational efficacy and usability in finite element program is also an important goal.

The pore relative humidity (or just humidity) drives shrinkage, greatly influences creep and facilitates the alkali-silica reaction (ASR). As shown in Fig. 2.1, a humidity decrease results from both external drying and self-desiccation, the latter being caused by hydration. This figure illustrates how the two shrinkage driving forces interact. At drying exposure, the drying front spreads at a rapidly decaying rate into a slab, as described by a nonlinear diffusion equation [28]. For thick slabs, the core can remain unaffected by external drying for a very long time. This makes it clear that the self-desiccation, driven
by hydration, can be the cause of a large pore humidity drop and play for a long time a
significant role. These are situations where the evolution of hydration degree over months
and years, even many decades, needs to be realistically modeled.

There are two recent observation that lead to the conclusion that the hydration and the
consequent self-desiccation must be long-lived, proceeding at higher pore humidities (at
decaying rate) for years, even decades (cf. Fig. 1 in [23]): Both the terminal autogenous
shrinkage and the swelling under water immersion proceed logarithmically up to at least
ten years. From this one must conclude that 1) The hydration, too, proceeds, at not
too low humidity, for years, probably even decades, and 2) the hydration reaction on the
micrometer level of cement paste must be expansive, even though on the nanolevel it is
contractive, as known for more than a century [23]. The latter conclusion was explained by the crystal growth pressure of growing and contacting barrier shells of C-S-H, which is more than offset by contraction caused by a decrease of chemical potential of nanopore water related to increased capillary tension. This multi-year or multi-decade evolution cannot be described by the existing hydration models because they predict the hydration to terminate within a few months and not more than a year.

2.2. Transport Through a Spherical C-S-H Shell

Fig. 2.2 illustrates the hydration reaction stages from setting time up to several years.

Because of complexity of the hydration reaction, simplifying assumptions are necessary. After an initial period $\Delta t_1$ of about 4 to 24 hours, a complete spherical barrier shell of C-S-H surrounding the remnant of anhydrous cement grain must exist, and is considered in the present analysis to persist for decades in duration. For simplicity, a fictitious ‘equivalent shell’ is here assumed to exist even during $\Delta t_1$, beginning with the time of set ($t = 0$).

In reality, the initial hydration, which is very fast, creates separate growing nano-scale globules of C-S-H clustered around the cement grain, which fuse at the end of $\Delta t_1$ into a continuous shell. Since the initial rate of hydration around the separate nano-globules is faster than predicted for the barrier shell, the inward diffusion of water through the shell toward the reaction interface of C-S-H with the remnant of anhydrous cement grain must initially (during $\Delta t_1$) be characterized by an increased effective hydraulic permeability,
Figure 2.2. The schematics of a five-stage idealized thought model of cement hydration that combines transport and boundary nucleation growth mechanisms. (a) formation of a semi-impermeable layer around cement particles during the dormant period, (b) a symmetric nucleation of a constant number of C-S-H particles on the surface of cement particles, (c) an isotropic and self-similar growth of C-S-H nuclei with spatially constant but time varying growth rate, (d) overlapping of growing C-S-H nuclei and the coverage of the entire surface with hydrates which terminates the nucleation-growth process, (e) the uniform growth of C-S-H particles that is controlled chiefly by the inward transport of water and the outward diffusion of ions to the solution.

The gradual decrease of effective $k_h$ as a function of the hydration degree reaches the actual permeability at the end of $\Delta t_1$ (see the next section, 2.3).

If a continuous barrier shell exists (hydration stages (d) and (e) in Fig. 2.2), the hydration kinetics is controlled by radial inward water diffusion through the porous C-S-H barrier, which is here assumed to occur uniformly over the shell. Let the shell thickness be $z - a$, where $a$ is the decreasing radius of anhydrous cement grain particle and $z$ is the
growing outer radius of the shell surface (Fig. 2.3). Assuming spherical symmetry, the velocity of the inward radial diffusion of water may be assumed to follow the Darcy law:

\[ v_w(r) = k_h \frac{dp_v}{dr} \]  

(2.1)

where \( r \) = radial coordinate and \( k_h \) = hydraulic permeability (also called conductivity or filtration coefficient). It is instructive to express the aforementioned transport formulation in terms of the pore relative humidity \( h \) (often called ‘humidity’, for the sake of brevity);

\[ h = \frac{p_v}{p_{sat}} \]  

(2.2)

where \( p_v \) = partial pressure of water vapor and \( p_{sat} \) = saturation vapor pressure, which greatly increases with \( T \). Although, fundamentally, the diffusion of water through the shell is driven by the gradient of chemical potential of pore water, \( \mu \), it will be convenient to replace \( \mu \) with the equivalent humidity \( h \), even though water vapor exists neither in the C-H-S shell nor at the interface with the anhydrous cement. So we define:

\[ h = e^{(\mu_0 - \mu M/RT)} \]  

(2.3)

where \( \mu_0 \) = chemical potential of water at saturation, and \( M \) = molecular weight of water = 18.02 g/mol. Inside the C-S-H shell, \( \mu \) is the chemical potential of the adsorbed water (free or hindered) in the nanopores. Eq. 2.1 now takes the form:

\[ v_w = B \frac{dh}{dr} , \quad B = \frac{k_0 \rho_0 p_{sat}}{\eta_0} \exp \left( \frac{-E}{RT} \right) \]  

(2.4)
where $B$ is the effective permeability (dimension $m^2/s$). Based on assuming incompressibility of diffusing water, the radial discharge, $Q_w$, or volume flow rate, is uniform for all radial directions and is

$$Q_w = A(r)v_w(r) = 4\pi r^2 B \frac{dh(r)}{dr}$$

which is a first-order ordinary differential equation. Its solution reads,

$$h(r) = -\frac{Q_w}{4\pi r B} + C$$

where $C$ is the integration constant, which must be determined from the boundary conditions.

The first boundary condition at the interface $r = a$ of C-S-H with the remnant of anhydrous cement grain is,

$$h(a) = h_c$$

where $h_c$ is the equivalent humidity at the interface of C-S-H shell with the anhydrous cement. As an approximation, probably a good one, we may consider that $h_c = 0.75$ for $T = 20^\circ C$, which is believed to be the humidity below which the hydration reaction does not proceed. Note that this value of $h_c$ might be different for concretes with some special admixtures, such as the silica fume (SF); see Bouny[13] who showed in 1999 that, for SF, $h_c$ can be as low as 0.60).
The second boundary condition, at \( r = z \), is \( h(z) = h_p \) = pore relative humidity. Substituting both boundary conditions into Eq. 2.6 we get:

\[
Q_w = 4\pi a B \frac{\langle h_p - h_c \rangle}{1 - \frac{a}{z}}, \quad C = \frac{h_p - h_c}{1 - \frac{a}{z}}
\]

where \( \langle x \rangle = \max(x, 0) \) serves to give \( Q_w = 0 \) when \( h_p < h_c \). Then, inserting this into Eq. 2.6 we obtain the relative humidity profile in terms of dimensionless quantities:

\[
h(r) = \frac{\left( r - h_p \right) - \langle h_p - h_c \rangle}{\left( \frac{r}{a} - \frac{r}{z} \right)}
\]

### 2.3. Effective Permeability Model

In the initial hydration period of one day or less, two stages of different kinetics can be distinguished (for a comprehensive discussion, see, e.g., [83]). The first stage is a short ‘dormant’ period right after the time of set, having the duration of a few hours. Second comes the stage of nucleation and growth of nano-globules of C-S-H. They are necessarily disconnected. The reason: since the C-S-H molecules have an effective diameter of 5 to 50 nm, a continuous shell, obviously, cannot form until the volume of all the C-S-H globules becomes sufficient to fill a continuous shell of that same thickness (Fig 2.3). The hydration degree \( \alpha \) at which this occurs is called the critical hydration degree, \( \alpha_c \). It is typically attained within the first day, an empirical equation for \( \alpha_c \) will be proposed). For \( \alpha > \alpha_c \), a continuous barrier shell of C-S-H exists and grows.
Based on the one-dimensional transport model in Eq. 2.8, the water discharge for one cement particle is,

\[
Q^1_t = 4\pi a_t z_t B_{\text{eff}}(\alpha, h_p) \frac{h_p - h_c}{z_t - a_t}
\]

where, for brevity, \(z_t = z(t), a_t = a(t)\), and \(B_{\text{eff}}\) = effective permeability, which is a function of the hydration degree and the inter-particle pore humidity. Assuming both functions to be independent, we can write,

\[
B_{\text{eff}} = B_0 f_0(h_p) f_4(\alpha)
\]

where \(B_0\) is an unknown constant. In the present simulations we consider \(B_0 = 1.0 \cdot 10^{-11}\text{m}^2/\text{day}\) for alite specimens and \(B_0 = 7.32 \cdot 10^{-12}\text{m}^2/\text{day}\) for cement specimens.
Function $f_4(\alpha)$ describes the dependence of $B$ on the hydration degree, and is assumed as follows:

\[ f_4(\alpha) = \gamma e^{-\gamma}, \quad \gamma = \left( \frac{\alpha}{\alpha_{max}} \right)^m \quad \text{for } \alpha \leq \alpha^* \]  
\[ f_4(\alpha) = (\beta/\alpha_s)^m e^{(\beta/\alpha_c)^m}, \quad \beta = \alpha - \alpha^* + \alpha^* \alpha_s / \alpha_{max} \quad \text{for } \alpha > \alpha^* \]  

Here $\alpha_{max}$ = hydration degree at which the permeability reaches its maximum, and $\alpha_{max} = \alpha_c/2$ where $\alpha_c$ is the critical hydration degree, defined here as the hydration degree at which the C-S-H barrier around the cement particle gets completed (typically about 24 hours.). Constants $\alpha^*$, $m$ and $\alpha_s$ are empirical and are considered as $\alpha^* = 1.5\alpha_{max}$, $m = 2$ for alite samples and $m = 1.8$ for cement samples, and $\alpha_s = 0.3$. Note that $\alpha - \alpha^* + \alpha^* \alpha_s / \alpha_{max}$ is used in Eq. 2.12b instead of $\alpha$, for $f_4$ to be continuous.

The expression for $f_4(\alpha)$ gives an increasing water discharge rate until the hydration degree reaches $\alpha_{max}$, and a decreasing rate afterwards. The start of $f_4(\alpha)$ from a small value can be explained by the metastable barrier hypothesis \[250, 154, 155, 129, 82, 83\] or the slow dissolution step hypothesis \[15, 16, 127, 105, 128, 83\]. The gradual decrease of $f_4(\alpha)$ after $\alpha_{max}$ is explained by the filling of capillary pores with hydration products \[214, 139\].

The number of cement particles, $n_g$, per unit volume of cement, is obtained from the volume of cement and the particle size, $a_0$:

\[ n_g = \frac{V^c}{\frac{4}{3}\pi a_0^3} \]
We calculate finite increments to track the evolution of state variables throughout the hydration process. The incremental, $dV^g$, and total, $V^g_{t+dt}$, volumes of C-S-H gel produced during $dt$ can be calculated as,

\begin{align}
(2.14a) & \quad V^c_{t+dt} = V^c_t + dV^c_t = V^c_t - n_g Q^1_t \zeta_{cw} dt \\
(2.14b) & \quad V^g_{t+dt} = V^g_t + dV^g_t = V^g_t + n_g Q^1_t \zeta_{gw} dt \\
(2.14c) & \quad V^{CH}_{t+dt} = V^{CH}_t + dV^{CH}_t = V^{CH}_t + n_g Q^1_t \zeta_{CHw} dt
\end{align}

where $V_t = V(t)$, etc., and $\zeta_{cw}$, $\zeta_{gw}$ and $\zeta_{CHw}$ are, respectively, the volumes of the cement consumed, the C-S-H gel produced and the portlandite per unit volume of discharged water.

To find the humidity changes in inter-particle capillary porosity, we rely on the increment of saturation degree, $S^{ip}$. To relate the saturation degree to the relative humidity, we use the desorption isotherm. To this end, we begin with calculating the total inter-particle water content, $V^w_{ip}$, and its increment. Noting that the increment of $V^w_{ip}$ must be equal to the incremental total water that diffuses through C-S-H gel, we have

\begin{equation}
(2.15) \quad V^w_{ip} = \phi^{ip} S^{ip}, \quad dV^w_{ip} = d\phi^{ip} S^{ip} + \phi^{ip} dS^{ip} = -n_g Q^{tot} dt
\end{equation}

The desorption isotherm, which relates the water saturation degree to the pore relative humidity, may be written as,

\begin{equation}
(2.16) \quad (1 - h_p) = K_h (1 - S^{ip}), \quad dh_p = K_h dS^{ip}
\end{equation}
where $K_h$ is the isotherm slope. The slope depends highly on the water-cement ratio, curing conditions and hydration degree of the cement paste. Here we limit attention to humidity levels higher than 75% and assume the isotherm slope to be constant, i.e., $K_h(w/c) = 1.2 - w/c$. Combining eqs. 2.15 and 2.16 we derive the humidity increment, 

$$dh_t^p = K_h \left( -\frac{n_q Q^t_{tot} dt - d\phi^{ip}_t S^{ip}_t}{\phi^{ip}_t} \right)$$

where $d\phi^{ip}_t$ is the inter-particle porosity increment (a decrement) calculated as 

$$d\phi^{ip}_t = -(dV^p_t + dV^{CH}_t + dV^c_t)$$

### 2.4. Predicted Hydration Curves

Now we use the proposed method to predict the hydration curves under different control parameters. Fig. 2.4 presents the effect of particle size distribution on the hydration degree and the rate of the heat of hydration for alite systems with the mean particle sizes of 13, 18, 38 and 82 µm which, respectively, correspond to effective particle sizes of 12, 17, 34 and 60 µm. We calibrate the model for the 13 µm particle size only, and the rest are predictions.

The calibration parameters are $\alpha_0 = 0.011$ and $\alpha_{max} = 0.21$ and the time of set was considered as 2 hours. Fig. 2.5 shows the effect of water-cement ratio on the hydration degree of OPC pastes. For the tests of Danielson [106] (Fig. 2.5a), the effective permeability model is calibrated to $w/c = 0.3$, and the calibration parameters are $\alpha_0 = 0.07$ and $\alpha_{max} = 0.21$. For the tests of Bentz [67] (Fig. 2.5b), the model is calibrated for $w/c = 0.35$ and $\alpha_0 = 0.06$ and $\alpha_{max} = 0.26$. 
Figure 2.4. Examining the predictive power of the effective permeability model vs. experimental measurements of Costoya [123] on alite phase. a) Hydration degree, b-e) The rate of heat of hydration for different particle sizes.

Figure 2.5. Prediction of the hydration degree of OPC for different water-to-cement ratios using effective permeability model against experimental measurement of a) Danielson [106] and b) Bentz [67].

Fig. 2.6 shows the effects of hydration temperature and cement type on the hydration degree. The activation energy and effective particle sizes for cements of Type I to Type III in Fig. 2.6 a-c are, respectively, 4.5*10^4, 4.5*10^4 and 5.0*10^4 J/mol and 12.5, 14 and 9 µm.
Figure 2.6. Prediction of the hydration degree of different cement types at different temperatures against experimental measurements of Lerch and Ford [176]. a) cement type-I, b) cement type-II and cement type-III.

[179] The calibration parameters for Type I cement with $T = 24^\circ C$ are $\alpha_0 = 0.07$ and $\alpha_{\text{max}} = 0.24$. All the other results are predicted. Note that, through all the simulations, the Blaine fineness of cement equal to $350m^2/kg$ was considered to correspond to particle size $13\mu m$.

Finally note that $a_{\text{eff}}$ is different from the average particle size, $a_m$. These two should not be used interchangeably. Here we use the effective size rather than the average size. For further illustration, we use a set of poly-disperse alite systems with the volume probability distribution function given in Fig. 2.7(a). First we calibrate the present effective permeability model with an alite system in which the average and effective particle sizes are almost identical, calibration parameters are the same with other Costoya’s experiments. Subsequently we use these parameters to predict the hydration degree of the aforementioned alite system (see Fig. 2.7). Clearly the model with the average particle size deviates from the experimental data while the model with the effective particle size fits these data perfectly.
Figure 2.7. Effective particle size method. a) The volume probability distribution function of an elite system measured by Costoya [123]. b) Calculating the hydration degree using effective permeability model with both the average size (solid blue) and the effective size (solid red) and their comparison with experiment [123].
CHAPTER 3

Moisture Diffusion in Unsaturated Self-Desiccating Concrete with Humidity-Dependent Permeability and Nonlinear Sorption Isotherm

By now it is generally accepted that correct calculation of the spatial distribution and time evolution of the pore humidity in concrete structures is necessary for realistic prediction of creep, shrinkage and thermal dilatation, along with their effects on deflections, crack formation and various degradation processes \[42, 37, 219\]. Relative humidity in concrete can change due to several phenomena including: self-desiccation, drying and temperature change. The nonlinear diffusion model for the drying of concrete previously developed at Northwestern by Bažant. and Najjar (1972) and embedded in some design codes. In this chapter, the Bažant.-Najjar model is improved and calibrated on the basis of recent more extensive experimental data from the literature as well as theoretical considerations. The improvements include: a new equation for the dependence of the self-desiccation rate on pore humidity and hydration degree; an updated equation for the decrease of moisture permeability at decreasing pore humidity, new equations are presented to predict the permeability and diffusivity parameters from the water-cement and aggregate-cement ratios, hydration degree and the type of concrete; and to capture the nonlinearity of the sorption isotherm as a function of pore humidity, and the dependence on the water-cement ratio. In this chapter, a new model to predict the humidity evolution
due to self-desiccation and drying process is presented and the humidity change due to temperature change later will be discussed in chapter 5.

3.1. Introduction

The relative humidity in the pores has a major effect on the moisture permeability, the rate of hydration (or aging), and the rate of strength-degrading chemical reactions such as the alkali-silica reaction (ASR). It plays a big role in the assessments of durability, fire resistance and radiation shielding. It matters for the age-old question of uplift in dams, and it must be considered in formulating the stress-strain law for concrete.

The studies of nuclear reactors structures in the 1960s led to the discovery (Bažant and Najjar 1971, 1972) that the diffusion equation for concrete is highly nonlinear because the permeability, as well as diffusivity, drops by more than an order of magnitude as the relative humidity, $h$, in the pores decreases. This causes that the linear diffusion theory is unable to fit the measurements of pore relative humidity, which is for example documented by the poor optimum fits of the experimental data in Fig. 3.1.

The Bažant-Najjar (BN) nonlinear diffusion model worked quite well for concretes half a century ago, and has been embedded in various recommendations and design codes, e.g., in the Model Code of fib (Fédération internationale de béton, 2013). Recently, though, it became clear that a generalization and improvement of the BN model is necessary to meet several broader objectives:
Figure 3.1. The best possible fits of mid-cylinder evolutions and transverse distributions of relative humidity in pores achievable with linear diffusion theory (characterized by constant permeability and diffusivity). Humidity data measured by (a) Kim and Lee (1999); (b) Abrams and Monfore (1965); (c) Abrams and Orals (1965); and (d) Hanson (1968). $y =$ distance from drying surface, and $t' =$ drying exposure time.

(1) For modern concretes, the foremost objective is to take into account the selfdesiccation, which drives the autogenous shrinkage. The selfdesiccation was negligible in old concretes but became significant in modern high-strength and high-performance concretes. The causes are two: 1) a low water-cement ratio ($w/c$),
and 2) admixtures such as silica fume and blast furnace slag \cite{14,162}. Baroghel-Bouny (2006) showed that, in sealed specimens of high-strength concrete with a high content of silica fume, the self-desiccation can decrease the relative humidity in the pores to as low as 65%. Such a humidity decrease may be beneficial because most of the deteriorative reactions in concrete stop or drastically slow down when the relative humidity in pores drops below 80%. For instance, the destructive alkali-silica reaction virtually stops at \( h < 75\% \) \cite{54,220}. During the last two decades, several self-desiccation models were proposed \cite{66,264,151}, but they turned out to fit poorly long time data, to be complicated by a number of empirical parameters requiring experimental calibrations for different concretes, or to be computationally too expensive for use in finite element analysis. In addition, the existing models have not been conceived as part of a general model for water transport in concrete. Such a model must combine self-desiccation with a simultaneous exposure to drying environment, and is the main goal of this study.

(2) The second objective is to use the currently existing database, significantly extended since 1972, to recalibrate the decrease of permeability with decreasing \( h \). As will be seen, better fits of test data can be obtained when the permeability decrease with \( h \) is more gradual than in the original BN model and continues even for \( h < 50\% \).

(3) The third objective is to improve the desorption isotherm. Its slope in the BN model was considered to be constant for \( h \) greater than about 30%, but recent tests \cite{203,201,270} showed that this is not true for high \( w/c \). Models to capture it exist \cite{270,104} but appear to be unnecessarily complicated, inaccurate
and computationally demanding. It will be sufficiently accurate and simpler to consider the isotherm to consist of two straight-line segments, the upper one being much steeper than the lower one and having a length depending on the type of concrete, particularly the $w/c$. The steepness and length of the initial straight segment of the isotherm near $h = 1$ are particularly important for calculating the selfdesiccation.

(4) The fourth objective is to propose a simple approximate formula for predicting the four parameters of the equation giving the moisture permeability as a function of $h$. This is needed to enable realistic calculation of the pore humidity field in structure, which must be known for predicting autogenous shrinkage, drying shrinkage, creep, rate of aging, and various deteriorative reactions such as the ASR. Further this is needed to extract more useful information from the existing creep and shrinkage data, for example from the worldwide database of about 4000 creep and shrinkage tests assembled at Northwestern [45, 150].

In the literature, the autogenous shrinkage and the drying shrinkage due to environmental exposure have generally been regarded as separate additive phenomena, the former caused directly by a volume change in chemical reaction. But were it true, it would be impossible to explain, e.g., that 1) the lowest, rather than highest, $w/c$ leads to the biggest autogenous shrinkage, or that, 2) under water immersion, the concrete swells. From such arguments it was concluded [24] that:

a) contrary to prevailing opinion, the hydration reaction per se is always expansive, beginning as soon as the solid nano-skeleton of C-S-H is formed (i.e., after
Figure 3.2. Schematic evolution of profiles of relative humidity (RH) caused by self-desiccation and by drying in the right half of a thin specimen: (a) uniform profile prior to drying exposure, (b) soon after that, (c) later, and (d) very late after drying exposure; and by drying in the right half of a thick specimen: (e) uniform profile prior to drying exposure, (f) soon after that, (g) later, and (h) very late after drying exposure.

The first day of hydration). Note that swelling can be due to several hydration reaction related phenomena like ettringite formation, portlandite formation, crystallization pressure, C-S-H shells push each other, disjoining pressure and nono-scale creep.

b) just like the drying shrinkage (caused by external drying), the autogenous shrinkage, too, is caused by a decrease of pore humidity (or of the corresponding chemical potential of water)\cite{148, 183, 130, 136, 181}. Therefore, both must follow the same law.
When external drying occurs, the selfdesiccation cannot be modeled independently. Rather, it must be considered as coupled. For clarification, consider the right half of a concrete slab of width $2L$, exposed to drying at environmental humidity $h_{env}$ that is below the maximum possible humidity decrease due to selfdesiccation. All the four sides of the slab are assumed to be sealed, so that the drying would be unidirectional.

Before exposure to drying, the relative humidity in the pores decreases uniformly due to selfdesiccation(Fig. 3.2a and e). In this study, the initial humidity value minus the humidity decrease due to selfdesiccation is called the selfdesiccation humidity. After drying exposure, the humidity profile of drying propagates into the slab, while the selfdesiccation humidity decreases only in that portion of cross section where its relative humidity has not yet reached the relative humidity in which hydration stops, $h_{st}$ (Fig. 3.2b,c,e and f). Eventually the drying front reaches the slab center (Fig. 3.2c,d and h). After that, the humidity due to external drying separates from the selfdesiccation humidity and decreases slower than the drying humidity profile, which causes a growing separation of the profiles of selfdesiccation and of external drying (Fig. 3.2d and h).

The area between the aforementioned two profiles, multiplied by the inverse isotherm slope (and weighted for axisymmetry), represents the amount of water lost to the environment, and the area above the upper profile, multiplied and weighted similarly, represents the amount of water consumed by the hydration reaction. When $h_{env}$ is not smaller than the minimum possible humidity reachable by selfdesiccation, $h_{st}$, the stages in Fig. 3.2c, d and h, are never reached.
3.2. Moisture Transport

Thermodynamic equilibrium of moisture in concrete is achieved when the chemical potential of water, $\mu$ (i.e., the Gibbs free energy per unit mass), is the same in all the phases of water and throughout the structure volume. Therefore, in principle, the mass flux of water, $j_w$, should be proportional to $\text{grad} \mu$. Such a diffusion model, however, seems inconvenient and apparently has never been developed.

Until the 1960s, one approach was to consider the flux of moisture to obey Fick’s law and to be proportional to $\text{grad} w_e$ where $w_e$ is the evaporable water mass per unit volume of concrete. In 1971, Bažant and Najjar (1971) proposed and in 1972 [28] elaborated a model in which the pore relative humidity, $h$, is considered as the primary unknown, whose gradient drives the diffusion (and is proportional to both $\text{grad} \mu$ and $\text{grad} p_v$ times a factor dependent on $h$, with $p_v$ being the vapor pressure).

The use of $\text{grad} h$ has three advantages:

(1) It is simpler, especially when the selfdesiccation is mild, for dealing with the effect of hydration, which acts as a sink withdrawing a lot of evaporable water from the pores;

(2) it is more direct for formulating the environmental boundary condition; and

(3) it can be verified and calibrated more directly, thanks to the recent development of good pore humidity gauges [200].
The Bažant-Najjar (BN) model, which has been embodied in the fib Model Code 2010 (Fédération internationale de béton, 2013), directly postulates that, under uniform temperature, the total moisture flux is:

\begin{equation}
\dot{j}_w = -c_p(h) \nabla h
\end{equation}

where $c_p$ is the moisture permeability (dimension kg/m·s), and $j_w = \text{mass flux of water (kg /m}^2\cdot\text{s)}$.

To ensure water mass conservation, the rate of change of moisture content, $\dot{w}_t$, must balance the influx of water mass, i.e.,

\begin{equation}
\dot{w}_t = -\nabla \cdot j_w + \dot{w}_s
\end{equation}

where $\dot{w}_s = \text{water mass that has been withdrawn from the pores and combined chemically by the hydration process; } \dot{w}_s \text{ represents a distributed sink and is the cause of selfdesiccation}$.

Under local isothermal equilibrium conditions, the pore humidity rate is a function of the rate of specific evaporable water content, $w_t$ (kg per m$^3$ of concrete). This function differs between drying and wetting conditions, due to capillary and hindered adsorption hysteresis. In the literature, the functions describing the dependence of $h$ on $w_t$ for drying and wetting are called the desorption and resorption isotherms. Only the desorption isotherm is considered here, and is formulated as:

\begin{equation}
\dot{h} = k(\alpha, h) \dot{w}_t
\end{equation}
where \( k(\alpha, h) \) (dimension \( \text{m}^3/\text{kg} \)) is the reciprocal moisture capacity (i.e., the inverse slope of the isotherm), whose evolution depends on the pore relative humidity and hydration degree, \( \alpha \) (characterizing the age of concrete); and \( k \) varies with \( h \), more so for concretes with high \( w/c \) (for detail, see Sec. Sorption Isotherm).

Incorporating \( 3.1 \), \( 3.2 \) and \( 3.3 \) gives the governing moisture diffusion equation for concrete:

\[
\frac{\partial h}{\partial t} = k(\alpha, h) \nabla \cdot (c_p \nabla h) + \frac{\partial h_s}{\partial t}
\]

where the last term on the right-hand side is a distributed sink representing the self-desiccation. It can be calculated using Rahimi-Aghdam et al.’s (2017) hydration model, and it can continue for many years. Same as Bažant-Najjar (1972) model, the dependence of moisture permeability \( c_p \) on \( h \) may again be expressed as follows (see Fig. 3.3a):

\[
c_p(h, \alpha) = c_1 \left( \beta + \frac{1 - \beta}{1 + \left( \frac{1-h}{1-h_c} \right)^r} \right)
\]

although the values of parameters \( c_1, \beta, h_c \) and \( r \), are now somewhat different, as obtained by the fitting of more extensive experimental data.

The data needed to calibrate Eq. (3.5) should ideally include the time evolution of relative humidity in the pores and of the humidity profiles throughout the cross section at subsequent times, for various specimen sizes, various environmental humidities and various ages at exposure to drying atmosphere. Such comprehensive data, unfortunately, do not usually exist for one concrete, and the parameters change significantly from one concrete to another. To be able to extract information from data for different concretes,
the equations for the dependence of unknown material parameters in Eq. (3.5) had to be judiciously assumed and then empirically calibrated.

The basic parameter is $c_1$, i.e., the permeability at saturation ($h = 1$). It is mainly a function of the pore size distribution. Two factors that affect the distribution most are the water-cement ratio, $w/c$, and the hydration degree, $\alpha$. The fitting test data led to the following empirical relation:

\begin{equation}
(3.6) \quad c_1 = 60[1 + 12(w/c - 0.17)^2] \alpha/\alpha_u
\end{equation}

The second important parameter is the transitional humidity, $h_c$, at which the steep permeability drop is centered. When $w/c$ increases, the pore size distribution shifts towards larger pores. As the drying starts, the larger pores get emptied first (since larger pores empty at a higher humidity). Therefore, for a higher $w/c$, for which the pores are bigger, the permeability begins to decrease at a higher relative humidity. The hydration reaction causes the pore sizes to decrease as a function of age or, more precisely, the
Figure 3.4. Dependence of permeability on relative humidity extracted from experimental data from: (a) Sørensen et al. (1979); and (b) Nilsson (1980).

hydration degree. The data fitting led to the following empirical relation:

\[
    h_c = 0.77 + 0.22 (w/c - 0.17)^{1/2} + 0.15 \left( \frac{\alpha_u}{\alpha} - 1 \right) \quad \text{but } h_c < 0.99
\]

It should be noted that the curing condition can also affect the value of \( h_c \), while Eq. 3.7 is calibrated for the sealed curing condition. Especially, this value should be modified if the specimen is cured in fog room or immersed in water. This is due to the fact that in the immersed condition, for the same hydration degree (and approximately the same pore structure), the relative humidity is higher. In this study, for simplicity, for water immersion, the \( h_c \) value is increased by the same amount as the \( h \) increase at the start of drying, compared to the sealed condition. For instance, if, at a certain drying time, the relative humidity at sealed condition is 0.96 and for water immersion is 1, the value of \( h_c \) is also increased 0.04 (1 - 0.96). This modification is more pronounced for lower \( w/c \) and is almost negligible for high \( w/c \).
Parameter $\beta$ gives the maximum relative drop of permeability as $h$ decreases. Based on more limited data, Bažant and Najjar (1972) assumed its value as 0.05, which was reached as $h$ dropped to approximately 0.7. Later experiments, though, indicated that permeability decreases further as $h$ drops below 0.7 (see Fig. 3.4). To describe it, $\beta$ is now changed to a variable parameter defined as follows:

$$\beta = \frac{c_f}{c_1}$$  \hspace{1cm} (3.8)

$$c_f^0 = 60[1 + 12(w/c - 0.17)^2] \frac{\alpha}{\alpha_u}$$ \hspace{1cm} (3.9)

$$c_f = \begin{cases} 
  c_f^0 & (h > h_s) \\
  0.1c_f^0 + 0.9c_f^0(h/h_s)^4 & (h < h_s)
\end{cases}$$  \hspace{1cm} (3.10)

where $\alpha_u$ is the ultimate hydration degree in sealed concrete, which is a function of $w/c$; it is estimated as

$$\alpha_u = 0.46 + 0.95(w/c - 0.17)^{0.6} \text{ but } \alpha_u < 1$$  \hspace{1cm} (3.11)

The last parameter to define is $r$; it determines how steeply the permeability decreases with $h$. Analysis of the data available in 1972 indicated exponent $r$ to be between 6 and 12, which gave a rather steep permeability drop (see Fig. 3.3). However, based on newer and more extensive data, $r = 2$, i.e., the permeability drop is much smoother. Fig. 3.4 compares the old and new permeability equations.

The predictions of pore humidity and drying shrinkage are quite sensitive to the shape of permeability law, Eq. (3.5). To document it, Fig. 3.5a,b presents the humidity profiles and shrinkage strain evolutions, in which two of the four parameters of permeability law
are changed in correspondence to the values commonly used in the literature. As seen, it makes a large difference.

Figure 3.5. Bažant-Najjar model for estimating humidity evolution and shrinkage using two different sets of calibration parameters. (a) Predicted relative humidity, and (b) predicted shrinkage.

### 3.3. Sorption Isotherm

The sorption isotherm is defined as the dependence of specific evaporable water content, \( w_e \) (kg per m\(^3\) of concrete, at constant temperature) on the relative pore humidity, \( h \). Drying depends on the desorption isotherm. Because of pronounced capillary hysteresis, the resorption (or adsorption) isotherm, which governs wetting, lies significantly below the desorption isotherm. Here we focus on drying and seek a simple equation for the desorption isotherm. To be applicable to concretes of different porosities, the isotherm is described in a dimensionless form, by the saturation degree; \( S = w_e/w_s \), as a function of \( h \) (\( w_s \) is the \( w_e \) at \( h = 1 \)).

The desorption isotherm is nonlinear \([203, 201, 270, 42]\), and highly so when \( h \) is near 0 and near 1. Its shape depends the concrete type. The most important parameters are
the water-cement ratio, $w/c$, and the age. Some tests show that certain admixtures affect the desorption isotherm significantly. Especially, adding silica fume fills the nanopores and thus makes the matrix more uniform, which in turn makes the isotherm slope near $h = 1$ less steep \[14, 162\]. Various methods to measure the desorption isotherm have been proposed, but either they are too complicated to use or necessitate several calibration parameters hard to determine.

Based on numerous experimental studies, the desorption isotherm may be approximated by a linear asymptote dropping from $h = 1$ with a steep slope, $m_1$, followed by a long linear segment of milder slope $m_2$ (Fig. 3.6a), extending down to about $h = 50\%$. 

Figure 3.6. Desorption isotherm. (a) The considered simplified desorption isotherm, (b) Experimental vs. simulated desorption isotherm. Lines = simulated results; points = experimental results. (experiments by Nilsson 2002.)
The proposed isotherm is a smooth transition between two linear asymptotes (Fig. 3-6a):

\[
(3.12) \quad dS = \frac{dh}{k(h, \alpha)}
\]

\[
(3.13) \quad \frac{1}{k(h, \alpha)} = m_2 + \frac{m_1 - m_2}{1 + \left(\frac{1-h}{1-h^*}\right)^2}
\]

where \(k(h, \alpha)\) is the inverse slope of the desorption isotherm,

and \(h^*\) is the intersection point of the two linear asymptotes. These parameters can be empirically estimated using experimental desorption isotherms. To do so, we use the tests of Nilsson (2002), who reports the desorption isotherm for various \(w/c\) ratios at different ages. Their fitting by the present model yields the following empirical relations:

\[
(3.14) \quad m_1 = \frac{f_\alpha}{c_{SF}} \left[2.4 + (5.26w/c - 0.68)^{1.5}\right]
\]

\[
(3.15) \quad m_2 = c_{SF}^{0.5} \times 1.18 \times (w/c)^{0.4}
\]

\[
(3.16) \quad h^* = 1 - f c_{SF} \frac{0.03}{(w/c)^2}
\]

\[
(3.17) \quad f_\alpha = \frac{\alpha_u - \alpha_0}{\alpha - \alpha_0}
\]

Parameter \(c_{SF}\) accounts for the silica fume. In its absence, \(c_{SF} = 1\). More generally, for a finite specific content \(\xi_{SF}\) of silica fume (mass of silica fume / mass of cement):

\[
(3.18) \quad c_{SF} = 1 + 2\xi_{SF}
\]

Fig. 3-6b compares the foregoing equations (solid curves) with Nilsson’s (2002) test data. As seen, the agreement is satisfactory. The effect of temperature on the isotherm
reported in some studies \[187, 216] is minor, except at high temperatures, and is here neglected.

### 3.4. Selfdesiccation

According to RILEM TC 196-ICC, the selfdesiccation is defined as the reduction in the internal relative humidity of sealed concrete. It is explained by removal of water for hydration and probably leads mainly to enlargement of already existing vapor filled pore space rather than cavitation of new vapor bubbles within liquid water. The volume and size of the empty pores depends chiefly on the water-cement ratio \((w/c)\), the degree of hydration, the particle size distribution of cement, and the type of admixtures. For lower \(w/c\) and finer cements, the microstructure is denser and the average pore size is smaller. A smaller pore size intensifies selfdesiccation, i.e., causes a higher rate and a lower final value of relative humidity, \(h_{st}\). The reason is that, in smaller pores, the meniscus curvature is higher and thus the pressure, given by the Laplace equation, is higher, which makes the desorption isotherm at high relative humidity values less steep. The decrease of pore size also explains why adding fine admixtures, such as silica fume, intensifies the selfdesiccation.

To calculate the selfdesiccation correctly, one should begin with the amount of water consumed by hydration. The ordinary portland cement (OPC) consists of various phases such as alite \((C_3S)\), belite \((C_2S)\), calcium aluminate \((C_3A)\), tetracalcium aluminoferrite \((C_4AF)\) and gypsum, as well as minor other phases (we use cement chemistry notation in which C, S, A and F stand, respectively, for CaO, SiO\(_2\), Al\(_2\)O\(_3\) and Fe\(_2\)O\(_3\); cf. Taylor, 1997). \(C_3S\) and \(C_2S\) are two major components of OPC and their ratio depends on the
cement type. To quantify the amount of water consumed by hydration, we consider, for simplicity, only C\textsubscript{3}S and C\textsubscript{2}S and we assume the cement to be composed of 80% C\textsubscript{3}S and 20% C\textsubscript{2}S. Chemically, hydration reaction of C\textsubscript{2}S and C\textsubscript{3}S is summarized as follows: \[218\]:

\begin{align*}
C_2S + 2.1 H &\rightarrow C_{1.7}SH_{1.8} + 0.3 CH \\
C_3S + 3.1 H &\rightarrow C_{1.7}SH_{1.8} + 1.3 CH
\end{align*}

(3.19)

where C\textsubscript{1.7}SH\textsubscript{1.8} and H constitute the typical C-S-H and water found in OPC pastes. Table 3.1 summarizes the molar volumes and densities of different components in the hydration reactions. Note that, in analyzing the aforementioned hydration reactions, the C-S-H pores have been assumed to be empty, and so it is necessary to include additional water that is trapped in the nanopores of C-S-H.

Beginning in 2000, Jennings, Constantinides and Ulm \[100, 156, 258\] established that the hydration reaction produces two types of C-S-H: low density C-S-H (LD), with porosity 36%, and high density C-S-H (HD) with porosity 26%. Jennings and Thomas (2000) showed that, for different w/c values, the ratio between these two C-S-H types varies. For instance, their model (JT model) predicted that, for w/c = 0.45, the proportions are 50% HD and 50% LD and, for w/c = 0.25, 80% HD and 20% LD. Therefore, the average porosity of C-S-H, \(\phi_{gp}\), depends mainly on w/c. Here, for simplicity, we assume a linear relation limited by lower and upper bounds, as follows:

\begin{align*}
0.27 < \phi_{gp} = 0.28 + 0.20(w/c - 0.3) < 0.35
\end{align*}

(3.20)

The lower and upper bounds on \(\phi_{gp}\) are introduced because it does not suffice to consider only the LD or HD phases.
Table 3.1. The molar volume and density of calcium-silicate phases present in the cement paste.

<table>
<thead>
<tr>
<th>Phase</th>
<th>Molar volume (cm$^3$/mol)</th>
<th>Density (g/cm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_3S$</td>
<td>72.9</td>
<td>3.15</td>
</tr>
<tr>
<td>$C_2S$</td>
<td>52.7</td>
<td>3.28</td>
</tr>
<tr>
<td>$H_2O$</td>
<td>18.0</td>
<td>1.0</td>
</tr>
<tr>
<td>C-S-H</td>
<td>110.1</td>
<td>2.05</td>
</tr>
<tr>
<td>CH</td>
<td>33.1</td>
<td>2.24</td>
</tr>
</tbody>
</table>

Having quantified the hydration relation and calculated the porosity of C-S-H, we can calculate the total volume of water, $\xi_{wc}$, per unit volume of cement, that is consumed by hydration to produce a C-S-H gel with the empty pores, $\xi_{bwc}$, and the volume of water that fills the gel pores, $\xi_{fwc}$:

\[
\xi_{wc} = \xi_{bwc} + \xi_{fwc} \tag{3.21}
\]

\[
\xi_{bwc} = w_{C_2S}\xi_{bwc}^{C_2S} + w_{C_3S}\xi_{bwc}^{C_3S} \tag{3.22}
\]

\[
\xi_{fwc} = \phi_{gp}\xi_{gc}S_{gp} \tag{3.23}
\]

where $\xi_{gc} = \text{volume of C-S-H gel produced (per unit volume of consumed cement)}$; $S_{gp} = \text{saturation degree of gel pores}$; $w_{C_2S}$ and $w_{C_3S} = \text{volume ratio of C}_2\text{S and C}_3\text{S respectively}$; $\xi_{bwc}^{C_2S}$ and $\xi_{bwc}^{C_3S} = \text{volume of water that is consumed to hydrate a unit volume of C}_2\text{S and C}_3\text{S, respectively, and to produce the C-S-H gel with empty pores}$.

For the typical case of 80% of cement consisting of the C$_3$S and 20% C$_2$S, we have

\[
\xi_{bwc} = 0.2(37.8/52.7) + 0.8(55.8/72.9) = 0.755
\]

\[
\xi_{gc} = 0.2(110/52.7) + 0.8(110/72.9) = 0.487
\]
Since the gel pores are much smaller than the capillary pores, they have been in the literature always considered as saturated. But Rahimi-Aghdam et al. (2017) show that this cannot be true and that at least some of the gel pores must be considered as unsaturated. Since gel pores are smaller than capillary pores their average saturation degree should be higher than normal capillary pores. Here, to calculate the average saturation degree of gel pores, we assume 33% of gel pores to have the same saturation degree as the capillary pores, and 67% the same as the nanopores (which, at high relative humidities, are always saturated). Accordingly, for a high relative humidity, we assume the saturation degree of gel pores to be:

\[(3.24) \quad S_{gp} = 0.67 + 0.33S_{cp}\]

**3.5. Hydration Model**

Although several experimental studies [140, 161] demonstrated a significant effect of aging on concrete permeability, this effect has usually been omitted from the equations for drying. This omission may be acceptable when the environmental exposure begins at a late age and high hydration degree. But it can cause a considerable error for concretes that begin to dry at an early age.

Therefore, the effect of the age at drying exposure is here taken into account. Although several hydration models took the age effect into account [158, 66, 263, 179], they were, unfortunately, too complicated and computationally demanding for use in finite element
structural analysis. Also, the previous hydration models applied only to concrete that is sealed or immersed in water.

The only model that is computationally undemanding and usable for all environmental conditions and ages appears to be the new model of Rahimi-Aghdam et al. (2017) which has been explained in Chapter 2. The only modification is considering variable slope for isotherm instead of constant slope.

Fig. 3.7 shows the predicted evolution of hydration degree using the presently modified version of Rahimi-Aghdam et al.'s (2017) hydration model. As seen, the model predicts the hydration degree well, both short-term and long-term. In addition, this model, unlike previous models, predicts the hydration reaction to be a long-lived phenomenon decaying roughly logarithmically (even for decades if $h$ does not drop). This agrees with the long-term selfdesiccation experiments.

Fig. 3.8 demonstrates the experimental vs. simulated comparisons for the relative humidity change due to hydration reaction in sealed specimens. The results agree again with the experiments quite well. Especially, by virtue of the varying slope of the desorption isotherm, they predict correctly the initial relative humidity, which is important for autogenous shrinkage and swelling. As seen, the calculated relative humidity change due to hydration depends strongly on the $w/c$ and is greater for concretes with a low water content. On the other hand, the selfdesiccation of concretes with a high $w/c$ at early ages is predicted to be mild, as expected.

Note that the foregoing selfdesiccation curves are true predictions; no parameter had to be calibrated. Also, considering various C-S-H gel porosities for concretes with different $w/c$ helped significantly in matching the observed selfdesiccation.
As the previous figures show, the proposed hydration model predicts well both the hydration degree and the self-desiccation. Since the present model is time consuming to code, and since often precise hydration predictions are not necessary for obtaining the correct humidity profiles, simplified empirical relations are here proposed and calibrated.
using the experimental results of Bentz (2006), as follows:

\[ (3.25) \quad d\alpha = c_a(\alpha_u - \alpha)^2(h - h_f)dt \]

\[ (3.26) \quad h_s = h_s^f \left( \frac{\alpha - \alpha_{set}}{\alpha_u - \alpha_{set}} \right)^{n_{wc}} \]

where \( n_{wc} = 25(w/c - 0.1) \); \( h_s^f = 0.5(w/c)^{-0.26} - 0.53 \); \( \alpha_u = 0.46 + 0.95(w/c - 0.17)^{0.6} \) \( \ll 1 \)

Here \( h_f \) is the relative humidity at which the hydration reaction stops; for normal concretes it can be set as \( h_f = 0.75 \); \( \alpha_{set} \) is the hydration degree at the time of set, which can be calculated as \( \alpha_{set} = 0.05 + 0.2(w/c - 0.3) \) (according to the hydration model, for normal temperature and normal cement). Note that these empirical relations are calibrated for concretes without admixtures, whose presence changes the rates of hydration degree and of selfdesiccation significantly.

The proposed simple equations can be integrated numerically by any method and suffice for capturing the effect of relative humidity on the rate of hydration. Fig. 3.9 shows that these equations predict correctly the evolutions of hydration degree and self-desiccation. However, they do not suffice for predicting the autogenous shrinkage. The reason is that the autogenous shrinkage is highly sensitive to an initial drop of relative humidity, and that a small error in predicting the selfdesiccation and hydration can cause a large error in predicting the autogenous shrinkage\[56, 225]\]. To avoid major errors, the complete hydration model must be used for that purpose.
Figure 3.9. Predicted hydration degree and self-dissipation using simplified hydration model: (a) Predicted vs. experimental hydration degree for experiment by Bentz (2006) [67]; and predicted vs. experimental self-dissipation for experiments by (b) Jiang et al. (2006), and (c) Kim and Lee (2004). Lines = simulation results; points = experimental results.

3.6. Boundary Conditions

Unless the specimen or structure is very large, the surface moisture transmission between the concrete and environment cannot be considered as instantaneous. The relative humidities in the environment and in the concrete pores at surface are different. In terms of relative humidity, the boundary condition reads [28]:

\[ j_w = \eta (h_c - h_{env}) \]  

where \( j_w \) is the moisture flux across the surface and \( \eta \) (dimension kg/m\(^2\)s) represents the surface emissivity. Various values \( \eta \) are found in the literature. For instance, Bažant and Najjar (1972) suggested \( 85 \times 10^{-6} \) (kg/m\(^2\)s) for indoor (or still) air and \( 350 \times 10^{-6} \) (kg/m\(^2\)s) for outdoor (moving) air. Ali and Urgessa (2014) used \( 6 \times 10^{-6} \) (kg/m\(^2\)s) for indoors and \( 9 \times 10^{-6} \) (kg/m\(^2\)s) for an isolated space. West and Holmes (2005) used a boundary condition of slightly different form: \( C(h) \frac{\partial h}{\partial r} = f_s (h - h_{cavity}) \). Here the
surface factor, $f_s$, was approximately considered as $10^{-8} - 10^{-7}$ m/s. This is roughly equivalent to the evaporation rate of $1.1 \times 10^{-6} - 11 \times 10^{-6}$ (kg/m$^2$s).

Parameter $\eta$ has a rather uncertain value. Besides, it adds complexity and hardly leads to better predictions. Therefore, Bažant and Najjar (1972) proposed replacing Eq. (3.28) for the boundary condition with a certain equivalent extra surface thickness, $\delta h_{eq}$, of an imagined added layer of concrete on whose surface the pore humidity equals the environmental value. Comparing analytical and experimental results, they adopted the value $\delta h_{eq} \approx 0.75$ mm. This method, also, was used to calculate the shape factors for concrete shrinkage and drying creep. For simplicity, $\delta h_{eq}$ is also used here, but with a different value of $\delta h_{eq}$, equal to about 4 mm, as estimated from the present calculations. The error of this approximation is found to have little effect and decreases as drying continues.

3.7. Comparison of Numerical and Experimental Results

The numerical results of the present model are first compared to the humidity profiles from the classical tests originally used for calibration by Bažant and Najjar (1972); see Fig. 3.10 and Fig. 3.11, which show a rather close agreement. The properties of the concrete used in these tests are given in Table 1. Note that, in Fig. 3.11, the humidity profile shows a decreasing humidity near the center. These experiments were performed under unidirectional drying (two parallel faces exposed and 4 faces sealed). The decrease of relative humidity seen near the center was probably caused by breakage of the seal near the center.
### Table 3.2. Material parameters for the analyzed experiments

<table>
<thead>
<tr>
<th>Figure</th>
<th>Authors</th>
<th>$w/c$</th>
<th>$a/c$</th>
<th>Shape</th>
<th>Thickness or diameter</th>
<th>Exposure time, $t'$</th>
<th>Env. humidity, $h_{env}$</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.10</td>
<td>Hanson (1968)</td>
<td>0.66</td>
<td>6.95</td>
<td>Cylinder</td>
<td>6 in</td>
<td>7 days</td>
<td>0.5</td>
<td>Elgin sand and gravel</td>
</tr>
<tr>
<td>3.11</td>
<td>Abrams and Monfore (1965)</td>
<td>0.45</td>
<td>4.36</td>
<td>Slab</td>
<td>12 in</td>
<td>7 days</td>
<td>0.1</td>
<td>7 bags of cem. per cu. yard</td>
</tr>
<tr>
<td>3.12</td>
<td>Kim and Lee (1999)</td>
<td>0.28, 0.4, 0.68</td>
<td>3.15, 4.14, 5.6</td>
<td>Slab</td>
<td>20 cm</td>
<td>3 days and 28 days</td>
<td>0.6</td>
<td>moist cured</td>
</tr>
<tr>
<td>3.13</td>
<td>Lauren et al. (2002)</td>
<td>0.66</td>
<td>6.95</td>
<td>Slab</td>
<td>7 cm</td>
<td>2 days</td>
<td>0.55</td>
<td>small aggregates</td>
</tr>
<tr>
<td>3.14</td>
<td>Grasely et al. (2004)</td>
<td>0.44</td>
<td>0.44</td>
<td>Slab</td>
<td>20 cm</td>
<td>1 day</td>
<td>0.5</td>
<td>Silica fume has added</td>
</tr>
</tbody>
</table>

![Figure 3.10. Relative humidity profiles in the cross section for different drying times; all experimental results (dots in parts a,b and c) by Hanson (1968). Lines = simulation results; points = experimental results.](image)

Next consider the more recent comprehensive experiments of Kim and Lee (1999). Concrete specimens were exposed to drying at the ages of 3 days and 28 days. Fig. 3.12 demonstrates again a satisfactory agreement with the pore humidity profiles for different depths below the drying surface, $y$, and for both exposure times. The specimens were
cured under water immersion before drying exposure, and thus the relative humidity prior to drying exposure was higher than in the sealed specimens. The equation that is proposed in this study for calculating $h_c$ (Eq. 3.7) was calibrated for the sealed condition. But in the case of curing under water immersion it needs to be modified. Here, for simplicity, the value of $h_c$ in Eq. (3.7) was simply increased by an amount equal to the humidity difference between the sealed and immersed conditions. This change was negligible for drying that began 3 days after mixing, but for drying that began after 28 days, the initial increase of $h_c$ at 28 days was about 2% for the finite elements near the surface.

All the preceding simulations of experimental data used the permeability law (Eqs. 3.5) with the same coefficient values. But permeability varies widely, depending on the aggregate size and type, cement type, casting technique, etc. It was clearly quite different in some reported experiments, as seen from the time evolution of pore humidity in, e.g., Laurens et al.’s (2002) experiments. Fortunately, to fit other data it is not necessary to change all the coefficient values. Mere scaling of the permeability law is sufficient. The
Figure 3.12. Evolution of relative humidity vs. drying time for different depths below the drying surface, $y$: (a-c) specimens immersed in water for 3 days and then exposed to drying; and (d-f) specimens immersed in water for 28 days and then exposed to drying. Lines are simulated results; points are experimental results. (Kim and Lee 1999)

Figure 3.13. The profiles of relative humidity over the cross-section for different drying times. Lines = simulated results; points = experimental results. (Experiments by Laurens et al. 2002.)

data of Laurens et al. (2002) can be closely matched if the permeability is reduced by 35% compared to the preceding simulations; see Fig. 3.13.
Furthermore, consider pore humidity predictions for concretes with silica fume (SF) as admixture, which has a major effect on permeability [162, 14]. The reason is refining the pore structure due to filling the pores and pozzollanic effect [69]. Refining pores causes the pore humidity to drop faster and to lower values. In addition, SF reacts with portlandite, producing more C-S-H and a denser structure, which result in a faster decrease of pore relative humidity. Fig. 3.14a compares the selfdesiccation for concretes with and without the SF. As it can be seen, the SF effect on the selfdesiccation is predicted correctly.

The SF effect gets modified in specimens exposed to the environment. This is evident from the plots of predicted vs. measured relative humidity in Fig. 3.14. They show again a good agreement. Some experimenters [162, e.g.] studied also admixtures other than the SF. But their effect is found to be very small, and is here neglected.

Finally, the validity of the simplified hydration model for exposed specimens needs to be checked. Fig. 3.9 showed that this model is able to predict closely the selfdesiccation
and hydration degree in sealed concrete. Now we check it for exposed concrete. As seen in Fig. 3.15, the simplified hydration model yields almost the same predictions as the full model, except for a small error during the initial days. The reason for this error is the slope change of the desorption isotherm due to the drying process. This change cannot be captured by the simplified linear isotherm. Overall, though, the predictions are quite good, and thus the present simplified hydration relation appears to be satisfactory.

### 3.8. Summary and Conclusions

1. An improved equation for the dependence of selfdesiccation rate on pore humidity and hydration degree is here formulated and calibrated by experimental data.

2. The 1972 Bažant-Najjar model for the strong decrease of moisture permeability with decreasing pore humidity is improved and calibrated by recent experimental data.
(3) Equations to predict the four parameters for the permeability variation from the material composition, water-cement ratio and hydration degree are formulated and calibrated by experimental data.

(4) The desorption isotherm as a function of pore relative humidity is improved by a nonlinear isotherm with a steep top segment whose slope and length depend on the water-cement ratio, according to experimental calibration.
CHAPTER 4

Predicting Autogenous Shrinkage and Swelling of Concrete Structures

A physically based model for autogenous shrinkage and swelling of portland cement paste is necessary for computation of long-time hygrothermal effects in concrete structures. Autogenous shrinkage and swelling of concrete are highly related to each other and both depend on relative humidity and hydration evolution of concrete. These two phenomena usually happen simultaneously in concrete samples. Therefore, to model these two phenomena correctly, one should consider a single model that can predict both. Unfortunately, this approach hasn’t been used by currently available models in literature and usually, they propose separate models to predict autogenous shrinkage and swelling. In this chapter, we propose a new physics-based model that is able to predict both swelling and autogenous shrinkage. In this new model, we use the new approach in which the hydration process causes permanent volume expansion of the porous cement paste as a whole due to the growth of C-S-H shells around anhydrous cement grains which forces the neighbors apart, while the volume reduction of hydration products contributes to porosity. Additional expansion can happen due to the growth of ettringite and portlandite crystals. On the material scale, the expansion always dominates over the contraction, i.e., the hydration per se is, in the bulk, always and permanently expansive, while the source of all of the observed shrinkage, both autogenous and drying, is the compressive elastic
or viscoelastic strain in the solid skeleton caused by a decrease of chemical potential of pore water, along with the associated decrease in pore relative humidity.

4.1. Introduction and Objective

It is usually required that concrete structures such as bridges be designed for a lifespan of at least hundred years. But durability problems often shorten the lifespan drastically \cite{41, 60, 31}. Although studied for more than a century, they are not yet understood satisfactorily. The long time volume changes of concrete are a particular problem.

The difficulty is to find a model that agrees with all the basic experimentally observed phenomena and, in particular, captures the interaction of creep, shrinkage, pore humidity changes, water transport, and cement hydration. The general form of such a model was proposed in Bažant et al. (2015) and was developed in more detail in Bažant et al. (2018). In this study, this previous model is refined, put on a solid thermodynamic basis, and made comprehensive so that it would agree with all the basic types of experiments reported in the literature. They include:

1. Autogenous shrinkage tests of long duration.
2. Long-time drying shrinkage tests for specimens of various sizes, in which the autogenous shrinkage is under way in the core until the drying front arrives.
3. Long-time tests of swelling under water and in fog room, for specimens of various sizes.
5. Tests of sorption isotherms.
(6) Abuhaikal et al.’s test of a specimen under constant saturation and constant pore pressure.

A selection of some of these tests, e.g., drying and autogenous shrinkage, can be described by diverse models. But a model that fits them all ought to be virtually unique. Its formulation is our objective.

The analysis that follows recognizes that data fitting must employ realistic models for: 1) creep, 2) hydration, 3) moisture diffusion, and 4) cracking damage due to tensile stresses, e.g., in drying shrinkage (for 2 and 3, see Appendices I and II).

4.2. Some Basic Phenomena

A serious problem with most models has been that they assume the autogenous shrinkage, drying shrinkage and swelling to terminate with a horizontal asymptote. This erroneous assumption has been caused by scarcity of long-time tests and by the widespread deplorable habit to plot the observed data in a linear time scale, which cannot reveal the long term trend. There exist, nevertheless, a few test data whose logarithmic scale plots document that no asymptotic bound is approached even after 30 years (Figs. 4.1a and 4.1b). The present analysis shows that the autogenous shrinkage and swelling evolve logarithmically for decades, and probably even for centuries. What is the physical source of such behavior?

To answer this vital question, consider first the autogenous shrinkage, which is, in modern concretes with low water-cement ratios and certain admixtures, much more intense than it used to be decades earlier. Following Bažant et al. (2015, 2018), its primary cause must be the decrease of pore relative humidity $h$ during self-desiccation, which is
physically no different from a decrease of $h$ due to external drying. It is inconceivable that a pore shrinkage would be caused by pore humidity drop due to external drying and not due to selfdesiccation.

According to RILEM TC 196-ICC [171], the selfdesiccation is defined as the “reduction of the relative humidity, $h$, in a sealed system when empty pores are generated ”. However, the wording of this definition is unrealistic and misleading. Empty pores are unlikely to be ‘generated’.

Vapor bubbles must form in liquid water at the time of set, produced by precipitation of dissolved gases or triggered by various dissolved ions. But they must immediately coalesce into large sparse bubbles of small surface curvature in the biggest pores [182]. Later on, generation of new vapor bubbles is not necessary, and is even very unlikely, due to the high resistance of liquid water to cavitation. What is doubtless happening is that the existing pore vapor space expands at decreasing $h$ as the existing capillary menisci increase their curvature and gradually recede into narrower and narrower pores; see Fig. 4.2. This is how selfdesiccation mechanism should be explained.
Consequently, the main source of the long-term autogenous shrinkage is the long-lived hydration reaction (for its prediction, see Appendix I). It was a widely held belief that the hydration stops after about one year. However, that can be correct only for specimens thin enough to dry up uniformly within a year. In the cores of a massive wall exposed to drying, high enough $h$ may persist for decades, even for centuries.

What causes the hydration process to be long-lived and progressively slower is that, after the first day, the anhydrous cement grains become enveloped in contiguous shells of cement hydrate. These shells have very low permeability, and the transport of water toward the anhydrous grain interface is very slow. In addition, as these shells gradually thicken, the pore relative humidity, $h$, as well as the chemical potential of water, $\mu$,
decreases, causing the hydration to slow down further (which experimentally illustrated by \[215, 160, 210, 269\] and mathematically described by Rahimi-Aghdam et al., 2017). If \( h \) in the pores is not below about 0.8, the hydration reaction proceeds logarithmically for many decades, probably even for centuries. This causes similar long-time evolution of autogenous shrinkage.

Locally, at a point of macroscale homogenizing continuum, all the phases of water must be in thermodynamic equilibrium. This means that the chemical potential \( \mu \) in all the phases must be the same. Therefore, a decrease of pore humidity \( h \) must produce tensile stress changes in all the phases of water (i.e., the vapor, liquid, and free and hindered adsorbed water). These stress changes must be balanced by compressive stress changes in the solid skeleton, which in turn must cause some compressive elastic deformation and creep in the solid skeleton of cement paste.

When immersed in water, most concretes swell. There exist data showing that the swelling is also a long-lived, multi-decade, phenomenon. Specimens immersed for a decade continue to expand logarithmically, with no bound in sight \[78\]. In these specimens, the pores near the surface must be saturated, \( h = 1 \), and those in the core must undergo self-desiccation until the water saturation front arrives. Since these specimens were not thick, the relative humidity was near saturation in all pores. Therefore, expansion cannot be driven by a change of pore humidity, which was almost constant throughout the experiment. The remaining reasonable candidate to drive the swelling is the expansiveness of hydration. Because it is inconceivable for the hydration to be both expansive in water immersion and contractive without immersion, the swelling must be driven by chemical expansion during hydration.
Indeed, if the hydration were contractive, the swelling in water immersion would not be physically explicable. But if it is expansive, both the swelling and the (autogenous and drying shrinkage) become explicable. Thus, in contrast to traditional thinking, we have a new paradigm (proposed in 2015 in \([25, 57]\)): The cement hydration is \textit{permanently expansive} in terms of apparent volume (solid with pores) although the absolute volume of hydration products is smaller than initial reactants (chemical shrinkage).

There is another phenomenon that probably also prolongs shrinkage, whether autogenous or drying—nanoscale viscoelasticity (Bažant et al. 2015) of the solid skeleton of the hardened cement paste. The increase of capillary tension, the increase of solid surface tension on nanoscale globules of hydrated cement, and the decrease of disjoining pressure of hindered adsorbed water in nanopores (<3 mm thick), as produced by pore humidity decrease, during selfdesiccation included, must be balanced by compressive stress changes in the solid skeleton; see Fig. 4.3 (all of these stress changes are proportional to \(\rho_l(RT/M)\ln h\), as defined later; cf. \([42, 25]\)).

Fortunately, all the three sources of pressure depend on the relative humidity, \(h\), in a similar way, as follows:

\[
(4.1) \quad p_i = C \ln h
\]

where \(i = \text{capillary tension, solid surface tension or disjoining pressure}\). Therefore, it is logical to consider \(p_l\) to depend on the relative humidity similarly:

\[
(4.2) \quad p_l = C_w \ln h
\]
where $C_w$ is a coefficient here considered as empirical. Therefore, $h$ may be used as a parameter controlling all these internal forces.

*Note:* In the writers’ opinion, which is not shared by some experts and has not been quantitively verified by anyone, the tensile stress changes in the nanopores caused by a drop of disjoining pressure are probably most important. The reason is that the nanopore water is a major part of water content and fills the nanopores completely, which the larger capillary pores are unsaturated. The disjoining stress changes must be expected to produce viscoelastic compressive strain in the skeleton, extending shrinkage in the long term, much beyond the drying process termination. But these stress changes may be expected to produce smaller volumetric strain than an externally applied pressure since they act only on a reduced cross section area.

### 4.3. Expansiveness of the Hydration Reaction in Hardened Cement Paste

Already in 1887, Le Chatelier showed that the cement hydration reaction is always contractive, i.e., the volume of the cement gel produced by hydration is always smaller
than the sum of the original volumes of anhydrous cement and water. Later this conclusion was supported by Powers and others. However, this conclusion is valid only in terms of absolute volume, not bulk volume (apparent volume). In the bulk, with a sufficient pore volume growth and constant relative humidity in the pores, the porous hydrated cement must always expand, even if the volume of the solid phase in the material decreases (for analogy, imagine a truss in which the bars are growing longer while the cross sections are shrinking even more to reduce the bar weight).

In hardened portland cement paste, the growth of two mutually contacting C-S-H shells around neighboring anhydrous cement grains must push the neighbors apart; Fig. 4.4. This must cause volume expansion of the porous skeleton of cement paste during hydration. The compression force within each pair of contacting neighbors, akin to the crystal growth pressure, must be balanced by overall tension in the solid skeleton of cement paste. Like hydration, the overall tension evolves for decades, and so does the swelling. Thus the long-term hydration causes not only autogenous shrinkage, via selfdesiccation, but also swelling. Furthermore, the long-time loading of the solid skeleton by pore pressure must, of course, produce not only elastic deformation but also creep.

Several studies suggested a different source of swelling—the growth of ettringite crystals. There are two reasons why this could be only one minor contribution and not the basic source of the long-term expansion: 1) Ettringite crystals do not form with a delay of many years. Rather, they dissolve at an early age and thus cannot affect the long-term swelling. 2) The long-term swelling can be seen in any type of cement regardless of the amount of C₃A phase. Therefore, even if we would admit the ettringite
crystal growth to be the cause of swelling in the initial days, it could not explain the swelling in the long term.

The final conceivable source of long-term swelling of concrete is the alkali-silica reaction (ASR). With a typical delay of 20–50 years, it can, of course, engender considerable swelling, but only if the alkali-content of the cement paste is high enough, if the aggregates are reactive, and if the ore humidity remains sufficiently high [54, 220]. This is not the case in normal laboratory experiments.

4.4. Thermodynamics of Unsaturated Poromechanics and Biot Coefficient

For normal saturated elastic porous materials, the volumetric part of the constitutive equations is written as [109]:

\begin{align}
\epsilon &= \frac{\sigma + bp}{K} = \frac{\sigma_{ef}}{K} \\
-\zeta &= \frac{c\sigma + p}{R} \\
\text{with } \frac{b}{K} &= \frac{c}{R}
\end{align}

where $\epsilon$ = volumetric strain, $\sigma_{ef}$ = effective stress, $\zeta$ = relative expansion of fluid phase (measured in terms of liquid water); $\sigma$ = volumetric stress (positive for tension), $p$ = pore pressure (positive for compression), $K$ = bulk elastic modulus of the dry solid skeleton, $R$ = elastic modulus of fluid phase characterizing its compressibility, $b$ = Biot coefficient, $c$ = Skempton coefficient. Eq. (4.5) is a symmetry relation required by the existence of thermodynamic potential, $\Psi$. 
For porous materials fully saturated by liquid water, Biot and Willis [71] proposed the estimate \( b = 1 - K/K_s \) where \( K_s \) = bulk modulus of the material forming the solid skeleton; for cement paste, \( K_s \) = bulk modulus of the C-S-H gel, and \( K_s \approx 45 \) GPa [201]. It must be stressed, however, that this estimate of \( b \) cannot be applied to the cement paste and other partly saturated media. The reason is that, due to selfdesiccation beginning right after the set, the hardened cement paste is always unsaturated, containing water vapor, liquid water, adsorbed water and air (usually, the water vapor and air may be combined as a single gas phase).

To deal with the unsaturated case, Coussy et al. (2004) ignored the adsorbed water and considered the Helmholtz free energy. Here we take the adsorbed water into account and find it clearer to use the Gibbs free energy, \( \Psi \) (per unit volume of the porous material):

\[
\Psi = \frac{\sigma^2}{2K} + \varphi(1 + \chi\epsilon)[S_l \rho_l + \Gamma_a(h) + (1 - S_l)\rho_v]\mu(h)
\]

where \( \rho_l = \rho_l\mu, \rho_v = \frac{p_v}{RT}, \mu = \frac{RT}{M} \ln h \)

\[
\Gamma_a(h) = \theta(h)\Gamma_1, \quad \theta(h) = \frac{1}{1 - h} - \frac{1}{1 - h + c_T h}
\]

Here \( \theta(h) \) is the well-known BET isotherm of gas adsorption in multimolecular layers [80, 79, 42, 270] (derived mathematically by Edward Teller); \( c_T = c_0 e^{\Delta Q_0/RT}, c_0 = \) constant, \( \Delta Q_0 \) = latent heat of adsorption minus latent heat of liquefaction; \( \Gamma_a(h) \) = mass of free adsorbed water at pore humidity \( h \) per unit pore surface area (\( \approx \rho_l \times \) effective thickness of adsorption layer); \( \Gamma_1 = \) constant = value of \( \Gamma_a \) for a full monomolecular layer; \( R = \) universal gas constant, \( \varphi = \) porosity, \( T = \) absolute temperature, \( M = \) molecular weight of water; \( \rho_l = \) pressure in liquid (capillary) water; \( S_l = \) liquid water saturation degree
≈ volume fraction of liquid water per unit volume of porous material; \( h = p_v/p_{sat}(T) \) = relative vapor pressure, or humidity, in the pores; \( \rho_l, \rho_v = \) mass densities of liquid water and water vapor (\( \rho_l \approx \) constant, in the case of unsaturated material); \( \chi = \) parameter introduced as an empirical coefficient (close to 1) such that \( \chi \) represent the relative change of pore volume. Equations (4.7) are based on the ideal gas equation for water vapor and on the Laplace and Kelvin equations of capillarity [42, e.g.].

Eq. (4.8) is contingent upon the assumption that the surface area of the adsorbed water layers exposed to water vapor is constant. This is not really true, even if there is no change in pore volume due to hydration. The reason is that much of the pore volume in hydrated cement has a width less than 10 molecular layers of water (2.67 nm), and the width of many pores is <0.5 nm. The adsorbed water layer in such pores completely fills the pore and is not exposed to vapor (while developing disjoining pressure [42]). As the drying penetrates into narrower and narrower pores, more of the surface area gets in contact with vapor and becomes available for free (unhindered) adsorption; see the schematic of a wedge pore in Fig. 4.2. This phenomenon is neglected in the BET isotherm, Eq. (4.8). Important though it might be, it is beyond the scope of this study.

The advantage of using the Gibbs, rather than Helmholtz, potential is that the Gibbs free energy per unit mass of the pore water, called the chemical potential, \( \mu \), must be the same in all the phases of water (at the same point of homogenizing macro-continuum) [42 e.g.]. For our purposes, we can neglect \( \chi \) compared to 1 and \( \rho_v \) compared to \( \rho_l \) (for
high temperatures, though, it could not be neglected). Then we have

\[
\Psi = \Psi(\sigma, p_l) = \frac{\sigma^2}{2K} + \varphi S(h) p_l(h) \tag{4.9}
\]

where

\[
S(h) = S_l(h) + \frac{\Gamma_1}{\varphi \rho_l(h)} \theta(h) \tag{4.10}
\]

Here \( S(h) \) = effective volume expansion of the combined liquid and adsorbed phases measured in terms of the liquid phase, such that \( S(h) p_l(h) \) give the work of the combined liquid and adsorbed water phases per unit pore volume within the material, while stress \( \sigma \) works on volumetric strain \( \epsilon \); \( \Gamma_1/\varphi \) is the mass of monomolecular adsorption layer per unit volume of the pores. Eq. (4.7) shows that capillary pressure \( p_l \) is controllable as a function of \( h \). Since the independent thermodynamic variables in Gibbs free energy \( \Psi \) are \( \sigma \) and \( p_l \), the first variation of \( \Psi(\sigma, p_l) \) is

\[
\delta \Psi = \frac{\partial \Psi}{\partial \sigma} \delta \sigma + \frac{\partial \Psi}{\partial p_l} \delta p_l \tag{4.11}
\]

\[
= \epsilon \delta \sigma - \zeta \delta p_l \tag{4.12}
\]

or

\[
\delta \Psi = \frac{\sigma}{K} \delta \sigma + \varphi S(h) \delta p_l \tag{4.13}
\]

(if \( \delta \) is replaced by \( d \), these equations represent the total, or exact, differential of \( \Psi \)). Note that the last term in the last equation can be neither \( \varphi \delta(S p_l) \) nor \( \delta(v_f S p_l) \), since neither \( S \) nor \( \varphi \) do any work, although they are variable (due to drying or hydration).
According to the Taylor series expansion, the second variation of $\Psi$ may be written as

\begin{equation}
\delta^2 \Psi = \frac{\delta^2 \sigma}{K} + 2 \frac{\delta \sigma \delta p_l}{H} + \frac{\delta^2 p_l}{R}
\end{equation}

where $K, H, R$ are the poromechanical stiffness moduli of the material, and the symmetry, manifested in the equality of the cross-diagonal coefficient $H$, is required by the existence of Gibbs potential. It follows that the elastic volumetric constitutive equation of the unsaturated elastic porous material may be written as

\begin{align}
\epsilon &= \frac{\sigma}{K} + \frac{p_l}{H} = \frac{\sigma + bp_l}{K} \\
-\zeta &= \frac{\sigma}{H} + \frac{p_l}{R} = \frac{c\sigma + p_l}{R}
\end{align}

Here $b = \text{Biot coefficient}$ and $c = \text{Skempton coefficient}$,

\begin{equation}
b = \frac{K}{H}, \quad c = \frac{R}{H}
\end{equation}

According Eqs. (4.16)–(4.17),

\begin{equation}
\frac{1}{K} = \left[ \frac{\partial \epsilon}{\partial \sigma} \right]_{p_l}, \quad \frac{1}{H} = \left[ \frac{\partial \epsilon}{\partial p_l} \right]_{\sigma} = - \left[ \frac{\partial \zeta}{\partial \sigma} \right]_{p_l}, \quad \frac{1}{R} = - \left[ \frac{\partial \zeta}{\partial p_l} \right]_{\sigma}
\end{equation}

Therefore, the definitions of the Biot and Skempton coefficients are

\begin{equation}
b = \left[ \frac{\partial \sigma}{\partial p_l} \right]_\epsilon, \quad c = \left[ \frac{\partial p_l}{\partial \sigma} \right]_\zeta
\end{equation}
If the porous material undergoes inelastic deformations or change of pore volume due to hydration or microcracking, $\Psi$ must be considered as a small increment of Gibbs free energy per unit volume, and a similar derivation can then show that Eqs. (4.16)–(4.17) must be replaced by the incremental volumetric constitutive relation

\[
\delta \epsilon = \frac{\delta \sigma}{K} + \frac{\delta p_l}{H} = \frac{\delta \sigma + b \delta p_l}{K} \tag{4.21}
\]

\[
-\delta \zeta = \frac{\delta \sigma}{H} + \frac{\delta p_l}{R} = \frac{c \delta \sigma + \delta p_l}{R} \tag{4.22}
\]

The equivalent pore pressure, $\pi$, in unsaturated material must be defined as a stress variable that works on the pore volume $\varphi$ when it is imagined to expand by $d\varphi$. This work is $\pi d\varphi$ and the complementary work is $\varphi d\pi$. So, the expression of Gibbs free energy increment may be written as

\[
d\Psi = \frac{\sigma}{K} d\sigma + \varphi d\pi \tag{4.23}
\]

Comparing this to Eq. (4.13), we conclude that

\[
d\pi = S \ d\rho_l \text{ or } d\pi = S(h) \frac{\rho_l RT}{M} \frac{dh}{h} \tag{4.24}
\]

\[
d\sigma_{ef} = d\sigma + b d\pi \tag{4.25}
\]

which is the increment of the total stress acting on the solid skeleton.

To allow using thermodynamics, the foregoing derivation had to treat $K$ and $H$ as material constants, which also implied $b$ to be a constant. Thus the potential, $\Psi$, applies only to a short time interval in which the progress of hydration is negligible, and is different in subsequent intervals. For an interval with negligible advance of hydration, integration
Figure 4.4. Contacting C-S-H shells, surrounding diminishing C₃S grains, push each other apart as they grow during hydration.

of the last equation gives,

\( \sigma_{ef} = \sigma + b\pi \)  

(4.26)

The Biot coefficient (as well as \( K \) and \( H \)) must obviously change if the pore volume \( \varphi \) decreases due to hydration or increases due to microcracking. Since Eq. (4.26) must hold true for any porosity \( \varphi \) and any \( b \), differentiation of the last equation furnishes the final general expression for the effective stress increments:

\[ d\sigma_{ef} = d\sigma + b\,d\pi + \pi\,db \]  

(4.27)

This means that the volume change of an elastic porous solid is \( d(\sigma_{ef}/K) \). The added term \( \pi\,db \) is physically supported by the fact that, if the pore volume increases due to, e.g., microcracking, the pore fluid must penetrate the additional pore space \( \varphi\Delta b \) and exert on the pore boundary additional pressure \( \pi\Delta b \), which is proportional to the added pore volume (because pressure \( \times \) volume change = work). This fact is graphically illustrated in Fig. 4.5.
Figure 4.5. The increase of Biot coefficient due to damage.

Note that, for a big change of the Biot coefficient, the term $\pi db$ is quite significant. This is the case at the early age of concrete after casting. Unfortunately, the term $\pi db$ seems to have been neglected in all the literature. Neglecting this term for a saturated soil with an almost constant Biot coefficient is acceptable, but causes a large error in predicting the deformation of young concrete. This error is discussed in more detail later, at Eq. (4.37). Note that there is similarity with the variation of Biot coefficient on hydraulic cracks in shale, which was intuitively explained and modeled in Chau et al. (2016).

The adsorption part, Eq. (4.8), has no effect on the present data fitting, because the humidities in the selfdesiccation and swelling experiments are not low enough. But it would matter for various applications.

Note also that the Gibbs free energy, $\Psi$, cannot be used when the solid skeleton of the material undergoes strain softening, because the response becomes non-unique. In that case, it may be preferable to use the Helmholtz free energy $\Phi$, which is related to $\Psi$ by the Legendre transformation:

\begin{equation}
\Phi(\epsilon, \zeta) = \sigma \epsilon - p_l \zeta - \Psi(\sigma, p_l)
\end{equation}
In practice, though, this is unimportant. One can simply deal with the solid part as if loaded by $\sigma_{ef}$.

Since concrete is not elastic but viscoelastic, the expression for the volumetric viscoelastic strain of the porous solid must be generalized as:

\begin{equation}
\epsilon(t) = \int_{0}^{t} f_{h}J(t, t', h, T)d\sigma_{ef}(t')
\end{equation}

where $t'$ is the time at which stress increments $d\sigma_{ef}(t')$ are applied, and $J$ is the compliance function for creep plus elastic deformation. It should be noted that creep compliance has been considered to be a function of relative humidity, $h$, and temperature, $T$, since several studies showed viscosity of concrete depends significantly on relative humidity and temperature.

### 4.5. Equations Governing Both Shrinkage and Swelling

As established at the dawn of cement research by Le Chatelier and confirmed by Powers and others, the cement hydration reaction is always contractive, i.e., the volume of the cement gel produced by hydration is always smaller than the sum of the original volumes of anhydrous cement and water. So how can the hardened cement paste swell? The reason is porosity. The growth of adjacent C-S-H shells around the cement grains pushes the adjacent shells apart, producing crystallization pressure $^{25}$. This is what causes the porous solid to expand (Fig. 4.4).

Until now we have disregarded the effect of swelling due to the hydration process. To this end, we can consider an additional pressure (akin to crystal growth pressure) that is induced by the macro-expansive hydration process, $\sigma_{hyd}$. For simplicity, we can assume it...
to depend linearly on the hydration degree (for its calculation, see Appendix I). Adding this pressure, we can rewrite Eq. (4.27) as:

\[
\begin{align*}
\frac{d\sigma_{ef}}{d\alpha} &= d\sigma + \pi db + b d\pi \\
&= d\sigma + \pi db + b \left( C_{w} S_{w} \frac{dh}{h} + C_{hyd} d\alpha \right)
\end{align*}
\]  

(4.31)

where \( C_{hyd} \) is a calibration parameter.

So far we have shown how the pressure should be modified for unsaturated media. However, pressure is not the only parameter that needs modification. Recently, Bažant et al. (2018) showed, for unsaturated medium, that the Biot coefficient depends not only on the relative stiffness of matrix to solid constituents, but also on the relative humidity of pores. Fortunately, at high relative humidity values \((h > 0.8)\) the dependence of Biot coefficient on relative humidity is negligible. Therefore, the traditional definition of Biot coefficient for simulating autogenous shrinkage and swelling when the pore humidity stays high leads to no significant error and is used here. To calculate the Biot coefficient, the effect of aging on the bulk modulus of concrete needs to be determined first. For simplicity, we consider the stiffness of cement paste to be related to the hydration degree linearly:

\[
K(\alpha) = K_u \left( \frac{\alpha - \alpha_{set}}{\alpha_u - \alpha_{set}} \right)
\]

(4.32)

The Biot coefficient depends on the bulk modulus \( K \) and can be calculated as

\[
b(\alpha) = 1 - \frac{K(\alpha)}{K_S}
\]

(4.33)
Figure 4.6. Decrease of stiffness, or $E$-modulus, at increasing water-cement ratio.

Finally, we must introduce a realistic equation for the creep compliance, $J$, of solid skeleton (dry material) loaded by pore water pressure.

This compliance should be different from the creep compliance of the cement paste as a whole (which includes water in capillary pores and the load-bearing adsorbed water in nanopores). For lack of deeper information, we assume a simple power function:

$$J(t, t', h, T) = \frac{\alpha_u}{\alpha} \left(c_0 + \beta_\eta(T, h)c_1(t - t')^n\right)$$

where $c_0$ corresponds to the instantaneous response; constant $c_1$ and exponent $n$ are two calibration parameters; $\alpha = $ hydration degree and $\alpha_u = $ ultimate hydration degree which is function of $w/c$. Note that factor $\alpha_u/\alpha$ is to consider the effect of aging. The effect of this factor is significant at early ages. Finally, $\beta_\eta(T, h)$ is a function to consider the change of viscosity due to the change of relative humidity or temperature. In this study, the same relation as the one in XMPS model [219] has used to calculate the $f_h$.

$$\beta_\eta(T, h) = \exp\left[\frac{Q_\eta}{R} \left(\frac{1}{T_0} - \frac{1}{T(t)}\right)\right] \left(p_0 + \frac{1 - p_0}{1 + \left(\frac{1-h}{1-h^*}\right)^n}\right)$$

Now we try to predict the autogenous shrinkage for cement pastes with different water-cement ratios, $w/c$. A change in $w/c$ changes not only the amount of selfdesiccation but
also the strength, as well as stiffness, i.e., Young’s modulus $E$. Therefore, to predict the autogenous shrinkage correctly, we need a model that can predict the stiffness, $E$, for different cement pastes correctly. In the literature, there are several empirical models for predicting the stiffness of concrete from its strength $f'_C$, but they are not suitable for the hardened cement paste. Therefore, we propose the following simple, empirically calibrated, equation that can predict the stiffness of cement paste for different $w/c$ (Figure 4.6):

$$E = A - B\frac{w}{c} \quad (A = 52 \text{ GPa}, \ B = 62 \text{ GPa})$$

The Poisson ratio for cement pastes with different $w/c$ is considered the same, $\nu = 0.2$.

### 4.6. Prediction of Autogenous Shrinkage

To check how good the predictions of the present model are, we begin with the experiments of Baroghel-Bouny. et al. (2006), in which the autogenous shrinkage of cement pastes was measured for different water-cement ratios. Fig. 4.7a compares the calculated...
curves with the measured points. As seen, the agreement is good. Note that the model was calibrated only by matching of the measured bulk modulus of the cement paste, and only for $w/c = 0.4$. The rest is the prediction. Also, it should be mentioned that due to considering hydration to be expansive, the model was able to predict initial swelling in cement pastes with high $w/c$ correctly.

Next, consider the experiments of Jiang et al. (2006). This experiment is almost similar to experiment by Baroghel-Bouny et al. (2006). The only main differences are having cement with higher percentage of C$_3$A and use of high range water reducer for cement pastes with low water cement ratio. However as autogenous values shows, the autogenous shrinkage values of this experiment is higher than the ones for Baroghel-Bouny et al. (2006). Specially, for the cases in which water reducer has used ($w/c = 0.2$ and 0.3) the autogenous shrinkage is significantly larger. This is due to the fact that water reducer admixtures reduce the viscosity thus increase the creep compliance \[44\]. In addition, the bigger autogenous shrinkage value for the cases in which water reducer hasn’t used is due to greater amount of C$_3$A. C$_3$A hydration process consumes significantly larger amount of water thus increases the selfdesiccation. To consider the effect of greater amount of C$_3$A, the value of $\xi_{bw}$ was modified from 0.755 to 0.76. Note that all other calibration parameters are the same. As shown in Fig. 4.7b, the predicted results are in good agreement with the experimental ones.
4.7. Combined Autogenous Shrinkage and Swelling

Let us now analyze the ability of the model to predict deformations when both swelling and autogenous shrinkage happen simultaneously. This is the case for all swelling experiments. The core undergoes autogenous shrinkage until the front of wetting arrives from the exposed surface (if the environmental humidity is not too high). Only the region between the surface and the wetting front experiences swelling. Therefore, the evolution of deformation and its final value are size dependent. As the volume-surface ratio increases the parts that undergo swelling play a smaller role and the autogenous shrinkage begins to dominate. The opposite happens for thin specimens, with low volume-surface ratios, in which the swelling dominates.

Mizayawa and Monteiro (1996) illustrate this phenomenon by experiment. They tested mortar prisms with $w/c = 0.30$ and different sizes. On the first day, the samples were sealed and then placed in fog room. The bottom and top of the test specimens were sealed to ensure two-dimensional (rather than three-dimensional) diffusion. In the literature, there seems to be no model that could predict these experiments well.

Fig. 4.8a compares the predicted versus experimental results for three different specimen sizes. As it can be seen, the predicted results are in good agreement with the experimental ones. In addition, the present model is able to predict the size effect in swelling. It should also be noted that these authors started measuring deformation at one day. This way, they ignored a significant part of autogenous shrinkage.

Fig. 4.8b shows the predicted results starting from the time of set. As seen, the difference is very significant. Considering all these complexities, it appears that the finite
element simulation is the only reliable method for predicting swelling. All the prediction formulae have significant errors.

4.8. Importance of Considering the Variation of Biot Coefficient

To illustrate the significant effect of variation of the Biot coefficient, a recent highly interesting and original experiment of Abuhaikal and Ulm [4] must be considered. In their experiment, the specimen was kept constantly saturated while both the external load (compressive) and the pore pressure were kept nearly the same, at $P \approx -1$ MPa; Fig. 4.9a. Their experiment ran from the time of set and the concrete was hydrating during the experiment. Therefore, the Biot coefficient must have been decreasing as the matrix was gaining strength; Fig. 4.9b. Since the material is hydrating and the masses of constituents are changing, the Biot relation can be used only at constant hydration degree $\alpha$, i.e.,

\begin{equation}
\sigma = \sigma_{solid}(\alpha) - b(\alpha)P
\end{equation}
Since \( P = -\sigma = 1 \text{MPa} \) is maintained, the stress (in MPa) in the solid part should be calculated as,

\[
\sigma_{\text{solid}}(\alpha) = -1 + b(\alpha)
\]

The stress evolution in the solid part, \( \sigma_{\text{solid}} \), is portrayed schematically in Fig. 4.9c. As the figure shows, the stress in the solid part, \( \sigma_{\text{solid}} \), is always compressive and increases in magnitude. This stress can cause significant deformation, especially when the creep due to this stress is taken into account.

The total strain, \( \epsilon_{\text{tot}} \), in Abuhaikal et al.’s experiment should be divided into two parts: 1) the normal shrinkage and swelling of young concrete, \( \epsilon_{\text{swell}} \), and 2) the strain due to compressive load on the solid constituent, \( \epsilon_{\text{load}} \). To calculate the strain increment due to the stress in solid, \( d\epsilon_{\text{load}} \), the strain due to \( \sigma_{\text{solid}} \) needs to be divided into two parts.

The first part is the strain at constant hydration degree due to the increments in loads, and the second part is the strain at constant loads due to the change of the properties of hydrating concrete, i.e.,

\[
d\epsilon_{\text{load}} = d\epsilon_{1,\alpha} + d\epsilon_{2,\alpha}
\]

As mentioned, at constant hydration degree, both the external load and the pressure are kept constant, and so \( d\epsilon_{1,\alpha} = 0 \). However, the second part, \( d\epsilon_{2,\alpha} \), is not zero and can be calculated as,

\[
d\epsilon_{2,\alpha} = J \cdot d\sigma_{\text{solid, load}} = J \cdot db
\]
Figure 4.9. Schematic predicted responses in Abuheikal et al. (2018) test. 
a) Applied forces; b) evolution of Biot coefficient; c) schematic evolution of stress in solid skeleton; 
d) strain due to swelling, summed with e) strain due to stress in solid skeleton, to obtain f) total strain.

where $J$ is the creep compliance value for the given time (including the elastic strain).

Finally, $\varepsilon_{\text{load}}$ can be obtained as,

$$
\varepsilon_{\text{load}}(t) = \int_0^t J(\alpha, t') \cdot \text{d}b(t')
$$

The effect of $\varepsilon_{\text{load}}(t)$ is not negligible. But it has, unfortunately, been neglected by Abuheikal et al. (2018), which led them to attribute the shrinkage to eigenstresses in concrete.
4.9. Conclusions

(1) The present analysis and comparisons with tests confirm a new paradigm on the porous material scale, the hydration reaction is *always and permanently expansive* (Bažant et al., 2015, 2017), even though it is contractive on the nanoscale. All shrinkage is caused by a decrease of pore humidity (it is possible that limited periods of early expansiveness of hydration might have been suspected by earlier researchers).

(2) Recognizing the expansiveness property leads to a grand unification of models for drying shrinkage, autogenous shrinkage and swelling at water immersion, and especially their long-time, multi-decade and century-long, evolution.

(3) Consequently, these seemingly diverse phenomena can all be predicted from one and the same material model, even for decades-long (and probably century-long) duration.
(4) When used in computer-aided design, this grand unification should help to design for century-long durability, a goal that has rarely been achieved for large concrete bridges and other structures [41].

(5) Selfdesiccation is explicable without assuming any cavitation in liquid water. It is explained by gradual recession of existing capillary menisci into narrower and narrower pores.

(6) Without a realistic model for long-term selfdesiccation, autogenous shrinkage, swelling and moisture diffusion, the present test data could not be fitted, in their entirety, by one-and-the same model.

(7) The importance of considering the Biot coefficient to decrease with increasing degree of hydration and to increase with increasing cracking damage is demonstrated.

(8) The new thermodynamic formulations of poromechanics with capillarity and adsorption, based on the Gibbs energy potential, lends the present model general applicability although the fitting of the present data, not encompassing low enough humidities, can be fitted without the adsorption part.

(9) The recently developed computationally efficient hydration model [221] using local continuum variables to bypass Bentz’s simulation of a three-dimensional system of hydrating particles makes feasible finite element analysis of shrinkage or swelling with hydration degree evolving differently at each integration point.

(10) The present introduction of a nonlinear desorption isotherm improves shrinkage and swelling predictions.
CHAPTER 5

Extended Microprestress-Solidification Theory for Long-Term Creep with Diffusion Size Effect in Concrete at Variable Environment

The current design procedures in structural engineering still require the design codes and recommendations to predict the creep and shrinkage for the whole cross-section of a beam or slab. Unfortunately, the fact that, in case of environmental exposure, the cross-section is in a highly non-uniform state of stress and humidity, with nonlinear stress-strain behavior, cracking and damage, makes the prediction much more complicated. The established model for point-wise constitutive law of concrete exhibiting shrinkage and creep with aging is Bažant.’s (1997) microprestress solidification (MPS) theory. Although this model has several advantageous respect to other models, it has issues in predicting drying creep correctly. Therefore, it is necessary to modify this model to be able to predict the creep of structures correctly in all environmental conditions. In this chapter, we present a method to modify the MPS model. The modified model is denoted XMPS model and in opposition to the original MPS model, considers different nano- and macro-scale viscosities. In XMPS model the macro-scale viscosity depends on the rate of pore humidity change, which is a new feature inspired by previous molecular dynamics (MD) simulations of a molecular layer of water moving between two parallel sliding C-S-H sheets.
5.1. Introduction

Until recently, the microprestress-solidification (MPS) theory [39, 38] appeared to give satisfactory predictions of the creep of concrete, long-term creep included, both without and with, simultaneous drying and temperature changes. In 2014, however, simulations of P. Havlásek at Northwestern University (in collaboration with M. Jirásek in Prague and with Z.P. Bažant) identified incorrect predictions of the effect of cross section size on the additional creep due to drying, and an excessive delay, behind drying shrinkage, of the additional creep induced by drying. In this paper (whose preliminary version was posted as ArXiv1805.05469), both deficiencies are here rectified by the extended microprestress-solidification theory (XMPS). Several other amendments are also introduced to improve on previous amendments proposed by Jirásek and Havlásek (2014). Extensive verification by a broad range of test data is an essential objective of this paper, due to its paramount importance for a complex material such as concrete.

The solidification theory separates viscoelasticity of the solid constituent, the cement gel, from the chemical aging of the hardened cement paste caused by solidification of gel particles and characterized by the growth of volume fraction of hydration products. This separation permits considering the viscoelastic constituent as non-aging, and thus greatly simplifies mathematical formulation. The gradual decrease of compliance with the age at loading is explained by the growth of the volume fraction of a non-aging constituent, the cement gel or C-S-H (calcium silicate hydrate) [52, 53, 42].

The solidification, however, cannot explain the marked decrease of creep viscosities continuing even after the hydration progress becomes feeble. Neither can it explain the
drying creep effect (aka the Pickett effect) and the transitional thermal creep. To explain these phenomena, the concept of microprestress was conceived, resulting in the microprestress-solidification (MPS) theory [39].

The microprestress characterizes self-equilibrated stresses at the nano-scale level. These stresses stretch and break the interatomic bonds resisting the slip of parallel C-S-H sheets and of adjacent C-S-H nano-globules, which is believed to be the main mechanism of creep in concrete. The microprestress is considered to be the result of disjoining pressures across the nanopores filled by adsorbed water layers. Micropresstress cannot be appreciably affected by the applied load, and may be imagined as a the effect of a strong but very soft prestressed spring compressed within a very stiff frame. The microprestress is initially produced by incompatible volume changes in the microstructure during hydration. It then relaxes but later again builds up when changes of moisture content and temperature create thermodynamic imbalance between the chemical potentials of vapor, liquid water and adsorbed water in the nano-pores of cement gel.

The XMPS improves the modeling of drying creep, particularly the dependence of macro-scale viscosity on the rate of microprestress. This improvement has been inspired by recent molecular dynamics (MD) simulations of Vandamme et al. [265] and Sinko et al. [243, 242] at Northwestern. They showed that the viscosity of creep, associated with the rate of relative slip of parallel planar walls or sheets of C-S-H, is greatly diminished by the presence of a water layer between the walls, and that, furthermore, the effective viscosity of slip between the solid surfaces decreases when the water layer moves. A useful finding is that the direction of movement of the water layer does not matter, which means that drying and wetting should have a similar effect. While suspected long ago [19, 21, 22],
these facts were not reflected in the original MPS model. They happen to have been the cause of error in diffusion size effect on drying creep, and also in delay of the drying creep effect after shrinkage.

5.2. Microprestress-Solidification Theory (MPS)

Within the service stress range (but with certain exceptions for unloading and simultaneous drying), the concrete creep law can be considered to be linear in stress and to follow the principle of superposition in time. Therefore, the creep is fully characterized by the uniaxial compliance function $J(t, t')$, representing the strain in time $t$ caused by a unit sustained uniaxial stress applied at age $t'$. The triaxial generalization need not be discussed here since it is well known how to obtain it under the assumption that the material is isotropic (e.g., Bažant (1972), (1982), Bažant and Jirásek (2018)).

In absence of significant plastic and viscoplastic strains that may arise at very high confining pressures, the normal strain of concrete can be decomposed as follows (Fig. 5.1)

\[(5.1)\]
\[
\epsilon = \epsilon_a + \epsilon_v + \epsilon_f + \epsilon_{sh} + \epsilon_T 
\]

where $\epsilon_a =$ instantaneous strain; $\epsilon_v =$ viscoelastic strain; $\epsilon_f =$ flow strain (purely viscous strain); $\epsilon_{sh} =$ shrinkage strain and $\epsilon_T =$thermal strain. The instantaneous strain, which is the strain appearing immediately after applying uniaxial stress $\sigma$, may be written as

\[(5.2)\]
\[
\epsilon_a = q_1 \sigma
\]

Since the retardation spectrum of concrete creep extends smoothly to load durations $\hat{t} \ll 10^{-4}$ s, it is convenient to define the instantaneous compliance as an asymptotic
extrapolation of short-time creep curves for near-zero load duration (i.e., for \( \hat{t} \to 0 \)). Such an extrapolation has the advantage that the age effect on the true mean instantaneous compliance happens to be negligible (the evidence for this fact is quite scattered but deviations are not systematic). This property was demonstrated by Bažant and Osman (1976) and Bažant and Baweja (1995, 1995b) by considering the measured compliances for load durations \( t \) ranging from 0.3 s to 1 week. They obtained optimum fits of the compliance values measured for different \( t' \) using a smooth formula of the type \( J(t, t') = q_1 + c \hat{t}^n \), where \( \hat{t} = t - t' = \) load duration. Then, by optimizing the fit of the data for various loading ages \( t' \), they obtained \( q_1 \), and found that the \( q_1 \) values for various \( t' \) were nearly the same (see also [42, Fig.3.5]).

Therefore, similar to B3 and B4 models [33, 44], the instantaneous compliance (i.e., its asymptotic value for \( \hat{t} \to 0 \)) is here considered as age independent, which brings about a significant simplification. Introducing an empirical factor \( p_1 \) depending on the cement
type, the instantaneous compliance $q_1$ is expressed as:

$$q_1 = \frac{p_1}{E_{28}}$$

where $E_{28}$ is the conventional elastic modulus at age 28 days (which, according to model B4, corresponds to the loading duration of about 0.001 day, or 1.44 min., while in previous model B3 it was 0.01 day).

The viscoelastic strain $\epsilon_v$, which originates in the solid gel of C-S-H, may be described by a relation of the same type as in the B3 and B4 models [33, 44, 42 Eq. 3.11];

$$\epsilon_v = \sigma \{ q_2 Q(t, t') + q_3 \ln \left[ 1 + (t - t')^n \right] \}$$

Function $Q(t, t')$ was derived in a differential form by asymptotic matching in 1988 by Bažant and Prasannan [52, 42 Eq.3.10]. Its integration leads to a binomial integral that cannot be expressed in closed form. But, in numerical structural analysis in time steps, the integral is not needed, and is even useless if the pore humidity or temperature varies. If $Q(t, t')$ is desired, it can be easily evaluated numerically.

Function $Q(t, t')$ is age dependent and its value decreases as concrete ages. Previous studies neglected the dependence of $Q(t, t')$ on the growing degree of hydration, $\alpha(t)$, and simply considered it as a function of loading time $t'$. This degree matters when the temperature or pore relative humidity, $h$, varies. The hydration reaction speeds up as the temperature increases and slows down as $h$ decreases. So does $\alpha(t)$. In traditional normal concretes (without silica fume), the hydration at room temperature virtually stops when $h < 0.75$. In modern concretes with low $w/c$ and silica fume, the pore humidity in sealed
specimens can drop as low as 0.65, because of self-desiccation. Besides, external drying causes non-uniform humidity profiles evolving in time.

To capture these effects, the actual time $t$ needs to be replaced by the equivalent time $\theta$ that is a function of hydration degree. To calculate $\theta$, a relation of the same type as in the solidification theory is used, and is calibrated using the experimental results on the hydration reaction reported in 2006 by Bentz:

$$\theta(\alpha) = \left[ \frac{0.28}{w/c} \left( \frac{\alpha_u}{\alpha} - 1 \right) \right]^{-4/3}$$

where $\alpha = \alpha(t)$ is a function of time, $w/c$ = water-cement ratio of concrete mix (by weight); $\alpha_u$ = ultimate hydration degree in sealed condition (which depends on $w/c$); and

$$\alpha_u = 0.4 + 1.45(w/c - 0.17)^{0.8}$$

Using the equivalent time, one can modify the expression for the evolution of $Q(t, t')$ by replacing the actual time $t$ with the corresponding equivalent time $\theta$:

$$\frac{dQ(t, t')}{dt} = \left( \frac{\lambda_0}{\theta[\alpha(t)]} \right)^m \frac{n\zeta^{n-1}}{\lambda_0(1 + \zeta^n)}$$

where $\lambda_0 = 1$ day, $m = 0.5$, $n = 0.1$, and $\zeta = t - t'$. This relation needs to be integrated to find $Q(t, t')$:

$$Q(t, t') = \int_{t'}^{t} \left( \frac{\lambda_0}{\theta[\alpha(t)]} \right)^m \frac{n\zeta^{n-1}}{\lambda_0(1 + \zeta^n)} dt$$

The integral $Q(t, t')$ in Eq. 5.8 cannot be expressed in a closed form, but its numerical evaluation is fast. An approximate asymptotic matching formula for $Q(t, t')$ was developed
by asymptotic matching in 1988 by Bažant and Prasannan [52, Eq. 20]. Appendix C
gives its generalization to the equivalent time. This explicit formula is very accurate and
its use reduces the demand for computer time.

Note that the shrinkage strain $\epsilon_{sh}$ is here understood as a point-wise eigenstrain,
whereas in the B3 and B4 models [33, 44] it represents the average shrinkage of the
whole cross section of a long beam or slab. In addition, contrary to model B4, the au-
togenous shrinkage is here not separated from the drying shrinkage of the cross section,
since both are caused by pore humidity drop [221]. The shrinkage strain is approximately
proportional to the relative humidity change, whether caused by external drying or self-
desiccation [42]. Therefore, in step-by-step analysis, at each integration point of each
finite element, the rate of shrinkage strain may be calculated as

\begin{equation} \tag{5.9}
\dot{\epsilon}_{sh} = k_0 \frac{\alpha_u - \alpha_0}{\alpha - \alpha_0} \dot{h}
\end{equation}

where $\dot{h} = \frac{dh}{dt} = \text{rate of humidity change}$, $k_0 = \text{empirical constant}$, and $\alpha_0 = 0.9\alpha_{set}$.
Likewise, the thermal strain rate reads:

\begin{equation} \tag{5.10}
\dot{\epsilon}_T = k_T \dot{T}
\end{equation}

where $\dot{T} = \frac{dT}{dt} = \text{rate of temperature change}$ and $k_T = \text{empirical constant}$.

To complete the creep law, one finally needs the rate of flow strain $\epsilon_f$, which is discussed
next.
5.3. Evolution of Flow Strain and Microprestress

The flow strain is modeled by a viscous flow element coupled in series to the solidifying Kelvin chain, as schematized in Fig. 5.2. For the flow element portrayed, it is imagined that the bonds across the slip plane cross the nanopore filled by hindered adsorbed water. They are subjected to two kinds of stress: the macroscopic applied stress $\sigma$ causing shear slip, which acts in the figure horizontally, and the tensile microprestress $S$, which acts transversely (vertically in the figure). The rate of strain in the flow element is considered to be

$$
\dot{\varepsilon}_f = \frac{\sigma}{\eta_M}
$$

where $\eta_M$ is the macro-scale viscosity. In the original MPS model, for simplicity, the viscosity was assumed to be the same for both the nano-scale and macro-scale, and was in both cases assumed to be a function of microprestress, $S$.

This assumption was obviously a simplification since the macro-scale viscosity must depend on several other phenomena as well. In particular, it must depend on the flow of water within the pores—the meso-pores or capillary pores, and (mainly) the nano-pores. The adsorbed water flow along the nano-pores accelerates as the rate of pore humidity change increases.

Recently Sinko et al. (2018) conducted molecular dynamics (MD) simulations of the rate of slip between parallel C-S-H sheets loaded by constant shear stress and by transverse compression. An interstitial layer of water, several molecules thick, was inserted between the two sheets and forced to move between these two sheets to simulate the flow of water into or out of the nanopore. It was found that the presence of the interstitial water
layer accelerates the relative sliding of the C-S-H sheets subject to constant shear stress and, more importantly, that the rate of relative sliding of the C-S-H sheets accelerates if the interstitial water layer is made to move relative to the mean velocity of the two parallel C-S-H sheets. The explanation is that the movement of the water layer changes the activation energy landscape at the interface, causing the effective viscosity to depend strongly on the flow velocity. The direction of the flow, corresponding to drying or wetting, was found not to be important.

The conclusion is that the macro-scale viscosity depends not only on the micropre-stress, but also on the water flow through the pores, the velocity of which is determined mainly by the rate of pore humidity change. It should, of course, be kept in mind that the rate of humidity change is not the only phenomenon that can accelerate the flow along the
pores. Generally, the rate of any disruption of thermodynamic equilibrium has a similar effect.

Since any phenomenon that causes thermodynamic imbalance increases the micropre-
stress, one can imagine the rate of microprestress as a measure of the water flow rate in
the nanopore and the corresponding viscosity to depend only on the microprestress, \( S \).
In the rheological model of Fig. 5.2, this viscosity is captured by adding an extra dashpot
whose viscosity depends on the absolute value of microprestress rate, \( |\dot{S}| \). Introduction
of the absolute value \( |\dot{S}| \) is justified by the MD simulations of Sinko et al. (2018), since
they found the directions of the flow along a simulated nanopore to be unimportant. As
a result of these considerations, the macro-scale viscosity may be introduced in the form,

\[
\frac{1}{\eta_M} = \frac{1}{\eta_n(S)} + \frac{1}{\eta_{\text{flow}}} = aS + b|\dot{S}|
\]

where \( \eta_n \) is the nanoscale viscosity that is a function of the microprestress only, and
\( \eta_{\text{flow}} \) represents the decrease in viscosity due to the water flow or the rate of any other
phenomenon causing thermodynamic imbalance in nanopores. More generally one could
consider \( \frac{1}{\eta_M} = aS^p + b|\dot{S}|^p \) but data fitting indicates no need for such complication.

The viscosity in Eq. 5.12 must increase with the age or, more precisely, with the degree
of hydration, \( \alpha \), which is a fact confirmed by many experiments [124]. So, it is reasonable
to consider both parameters \( a \) and \( b \) in Eq. 5.12 to depend on \( \alpha \). For simplicity, a linear
dependence may be assumed:

\[
a = a_0 \frac{\alpha_u}{\alpha}, \quad b = b_0 \frac{\alpha_u}{\alpha}
\]
where \( a_0 \) and \( b_0 \) are two empirical constants. Combining Eq. 5.12 and 5.13, we can write

\[
\frac{1}{\eta_n} = \frac{\alpha_u}{\alpha} a_0 S, \quad \frac{1}{\eta_M} = \frac{\alpha_u}{\alpha} \left( a_0 S + b_0 |\dot{S}| \right)
\]

The next important issue the evolution of microprestress. The microprestress, \( S \), is imagined to characterize the average of normal stresses acting across the slip planes with hindered adsorbed water layers between them. The disjoining pressure in these layers, and thus also the microprestress, is considered to develop first during the initial hardening of cement paste. During the initial rapid hydration, the microprestress builds up mainly as a result of crystal growth pressures and localized volume changes close to the nanopores. Therefore, during the initial days of fast hydration, \( S \) depends mainly on the hydration degree, \( \alpha \), and is calculated simply as

\[
S = S_0 = c_0 q_4 \text{ for } \alpha < \alpha_0
\]

where \( c_0 \) is an empirical constant; \( q_4 \) is the creep law parameter in models B3 and B4; \( S_0 \) is the initial microprestress, and \( \alpha_0 \) is the hydration degree prior to which the hydration reaction has the dominant control of microprestress. The value \( \alpha_0 = 0.6\alpha_u \) works well.

Later, after the volume changes due to hydration have almost ceased, the changes of microprestress are controlled mainly by the changes in the disjoining pressure, which responds with negligible delay to the changes in the capillary tension and surface tension at the same location. According to Bažant et al. [39, 38], the evolution of microprestress can be assumed to obey a Maxwell-type rheological model with variable viscosity \( \eta_n(S) \).
and stiffness $C_s$.

\[
\frac{\dot{S}(t)}{C_s} + \frac{S(t)}{\eta_n(S)} = \frac{\dot{s}(t)}{C_s}
\]

where $\dot{s}(t)/C_s$ is the time rate of Maxwell model strain due to any phenomena that may cause thermodynamic imbalance in the microstructure \[42\]. These phenomena are analyzed next.

### 5.4. Temperature and Humidity Effects

The main phenomena affecting $\dot{s}(t)/C_s$ are: 1) the temperature change and 2) the humidity change. First the temperature. Its effect is complicated by interference of several physical mechanisms which can be described as follows,

1. A temperature increase accelerates the bond breakages and restorations.
2. The higher the temperature, the faster the chemical process of cement hydration, and thus the faster the aging of concrete.
3. The temperature change alters the capillary tension, crystal growth pressure, surface tension, and disjoining pressure, all of which can alter the microprestress and creep rate.
4. The temperature change alters the internal relative humidity, which either increases or decreases the creep rate.
5. The temperature increase alters the microstructure of C-S-H and of the weaker interfacial transition zone (ITZ), which usually weakens the concrete.

The relative pore humidity affects the creep rate and does so mainly in three ways:
: a) As the relative humidity decreases the viscosity increases and bond breakage decelerates.

: b) The higher the humidity, the faster is the chemical process of cement hydration and thus the aging of concrete, which reduces the creep rate.

: c) The evolution of humidity alters the capillary tension, crystal growth pressure, surface tension and disjoining pressure, which all change the microprestress.

To predict the creep rate correctly, one must consider the effect of all the aforementioned mechanisms. Except mechanisms 4 and 5 due to temperature change, they have already been considered in the original MPS model [39]. In addition, these two mechanisms are significant for the case of temperature varying during the experiment. Let us begin with considering the effect of temperature on the entire creep law. In models B3 and B4, this effect was considered using a temperature dependent time \( t_T \) instead of the actual time \( t \). Here we use the same idea to calculate the viscoelastic creep (the effect on flow term will be formulated separately);

\[
t_T = \int_0^t \beta_T(\tau) d\tau
\]

\[
\beta_T(t) = \exp \left[ \frac{Q_h}{R} \left( \frac{1}{T_0} - \frac{1}{T(t)} \right) \right]
\]

\[
\epsilon_v(T) = \sigma (q_2 Q(t_T, t'_T, t'_eff) + q_3 \ln [1 + (t_T - t'_T)^n])
\]

where \( T = \) absolute temperature; \( T_0 = \) reference temperature, chosen as \( T_0 = 293 \) K; \( R = \) universal gas constant; \( Q_h = \) activation energies for the hydration processes (whose values depend on the cement type); and \( t'_T = \) value of \( t_T \) at the time of loading.
The effect of the first temperature related mechanism on the flow strain rate is formulated by decreasing the viscosity, to reflect the acceleration of bond breakage, and by decreasing $C_s$, to reflect the acceleration of bond restorations. Both temperature effects can be described by an Arrhenius type equation. Since the first relative humidity related mechanism also changes the viscosity, the influences of both the temperature and relative humidity are considered simultaneously, as follows:

\[ \eta(T, h) = \eta_{T_0,\text{Sat}} / \beta_\eta(T, h) \]  \hspace{1cm} (5.20)

\[ \beta_\eta(T, h) = \exp \left[ \frac{Q_\eta}{R} \left( \frac{1}{T_0} - \frac{1}{T(t)} \right) \right] \left( p_0 + \frac{1 - p_0}{1 + (1 - h^*)^{n_h}} \right) \]  \hspace{1cm} (5.21)

\[ C_s(T) = C_{s_0} / \beta_{C_s}(T) \]  \hspace{1cm} (5.22)

\[ \beta_{C_s}(T) = \exp \left[ \frac{Q_C}{R} \left( \frac{1}{T_0} - \frac{1}{T(t)} \right) \right] \]  \hspace{1cm} (5.23)

where $Q_\eta, Q_C = activation$ energies for the viscosity change and $C_s$, while the following parameters are considered as empirical constants: $p_0 = 0.5, h^* = 0.75$ and $n_h = 2$. For simplicity, one may assume the same activation energy for both the macro- and nano-scale viscosities. In addition, since generally temperature increase accelerates creep, we assume the viscosity decrease to be dominant, and it is found reasonable to set $Q_C = Q_\eta/2$.

The effect of the second mechanism, driven by the changes of temperature and relative humidity, is already included through the acceleration of hydration reaction, and no other modification is needed. The third and fourth mechanisms caused by temperature change, and the third mechanism caused by relative humidity, modify the microprestress value through the changes of capillary tension, surface tension, and disjoining (or crystal growth) pressure.
All the aforementioned pressures have almost the same relation to the relative humidity, and all of them are all determined by changes in the chemical potential of pore water \((\mu = (RT/M\rho_l) \ln h, \rho_l = 1 \text{ g/cm}^3)\). At each location, \(\mu\) is the same in all the phases of pore water and its change represents the ultimate driving force of hygrothermal deformations. Therefore, and in conformity to the original MPS model (Bažant et al. 1997a,b), all these pressures are combined as one effective pressure, which can be written as

\[
(5.24) \quad p_{\text{eff}} = p_0 + C_p \frac{RT}{M} \ln (h(t, T))
\]

Here \(R\) = universal gas constant, \(M\) = molecular weight of water (in moles), and \(p_0\) and \(C_p\) are two empirical constants. Note that, to capture mechanism 4 due to temperature change, the humidity is here considered to be a function not only of time \(t\), as in the original MPS model, but also of temperature \(T\) (which is a mechanism considered in Diluzio and Cusatis [113] as well). The rate of effective pressure can then be written as,

\[
(5.25) \quad \dot{p}_{\text{eff}} = k_1 \left( \dot{T} \ln (h(t, T)) + \frac{T}{h} \frac{\partial h}{\partial t} + \frac{T}{h} \frac{\partial h}{\partial T} \frac{\dot{T}}{T} \right)
\]

where the last term describes the change of relative humidity due to the temperature change (Fig. 5.3a). In Eq. (5.25), \(\partial h / \partial T = \kappa\) = hygrothermal coefficient, introduced in 1970 by Bažant and in 1972 by Bažant and Najjar [28] (Fig. 13) and known to depend strongly on the relative humidity at which the temperature change happens; see Fig. 5.3b. Here \(\kappa\) is calculated based on the experimental results of Grasely and Lange in 2007.

The effect of the saturation degree on the effective pressure is not used here directly since the major effects on the microprestress are those of the disjoining pressure changes on the surface tension. They dominate in the smallest pores, which remain filled by water.
even at low $h$. This simplification, though, may cause appreciable errors at very low $h$.

Next we need to relate these changes to the microprestress change, which is, in turn, related to the effective pressure change. One can use the simple relation:

$$\dot{s} \propto \dot{p}_{eff}$$

The last mechanism that needs to be included is the effect of temperature change on the microstructure of C-S-H and particularly the ITZ \cite{228, 88, 121}. Several studies showed that elevating the temperature at early ages when the relative humidity is high alters the microstructure of C-S-H, especially in the ITZ, such that the average porosity of C-S-H increases. It has been previously shown that the microstructure has significant impact on the macroscopic (i.e., effective) material properties \cite{74, 75}. Therefore, the microstructure change weakens the concrete and can cause sudden increase of microprestress (due to
thermodynamic imbalance). One can formulate these effects as follows:

\[ q_{1,2,3}(T) = q_{1,2,3}(T_0) [1 + c_T (T - T_0)] \]  

\[ \dot{s} \propto f(h) \dot{T} \]

where Eq. 5.27 represents a weakening of microprestress and Eq. 5.28 represents its enhancement. Function \( f(h) \) introduces the fact that changing C-S-H microstructure is facilitated by a high relative humidity [121, 228]. Its simple, empirically calibrated, form is:

\[ f(h) = c_h h^r \]

where \( r \) is an empirical exponent which is set as \( r = 3 \).

The reality, though, is a bit more complicated. Fahmi et al. (1972) showed that the effect of temperature on material stiffness is irreversible and is important only if the temperature rises above the range of previously experienced temperatures. Temperature fluctuations within that range do not have much effect.

Having formulated the mechanisms contributing to the microprestress, we need to combine them. All these contributions are simply assumed to be independent and additive. Thus we get,

\[ \dot{s} = c_p \dot{p}_{eff} + f(h) \dot{T} = c_1 \left( \dot{T} \ln (h(t, T)) + \frac{T}{T} \frac{\partial h}{\partial t} + \frac{T}{h} \frac{\partial h}{\partial T} \frac{\partial T}{\partial t} \right) + c_h h^r \dot{T} \]
where the $c_p$ and $c_1 = k_1 c_p$. Consequently, the equation governing the relaxation of microprestress reads:

\[
\frac{\dot{S}}{C_S(T)} + \frac{S}{\eta_n(S,T,h)} = \frac{\dot{s}}{C_S(T)}
\]

Upon inserting Eqs. 5.20, 5.22 and 5.30 into Eq. 5.31, the equation for relaxation of microprestress becomes:

\[
\dot{S} + \frac{C_{T_0}}{C_S(T)} \frac{\beta_n(T,h)}{\beta C_s(T)} S = c_1 \left( \dot{T} \ln(h(t,T)) + \frac{T}{h} \frac{\partial h}{\partial t} + \frac{T}{h} \frac{\partial h}{\partial T} \frac{\partial T}{\partial t} \right) + c_h h^w \dot{T}
\]

where $\eta_{T_0, sat}(S) = 1/a S^{p_1}$, and $C_{T_0}$ is a constant. As already mentioned, we assume $p_1 = 1$, and so the microprestress relaxation equation can be simplified as:

\[
\dot{S} + a_S \frac{\beta_n(T,h)}{\beta C_s(T)} S^2 = c_1 \left( \dot{T} \ln(h(t,T)) + \frac{T}{h} \frac{\partial h}{\partial t} + \frac{T}{h} \frac{\partial h}{\partial T} \frac{\partial T}{\partial t} \right) + c_h h^w \dot{T}
\]

where $a_S = C_{T_0}/a$. Finally, using $p_1 = p_2 = 1$ as before, we obtain the rate of flow strain:

\[
\dot{\epsilon}_f = \frac{\sigma}{\eta_M(S,T,h)} = \sigma \left( a S + b |\dot{S}| \right) \beta_n(T,h)
\]

### 5.5. Water Transport Model, as Recently Refined

To predict the creep rate under the influence of variable humidity, a realistic model for water transport or drying is obviously important. Over the years, many were proposed. Some were transplants from other porous material, especially soil science, but did not work well since they ignored, or reflected poorly, two particular features of concrete: 1) in concrete, there is a major distributed sink of evaporable water due to hydration (continuing for years); and 2) in normal concrete, neither the vapor phase of water, nor
the liquid capillary phase, percolate, and a water molecule moving from one pore to the
next must pass through the hindered adsorbed phase filling the nanopores, which caries
load and behaves as part of the solid skeleton.

Because of gradual filling of pores by deposition of hydration products, the hydra-
tion sink affects the pore relative humidity much less than it does the specific content of
evaporable water. Recognizing this, Bažant and Najjar in 1972 adopted the pore relative
humidity as the primary variable (their model was incorporated into the Model Code of
fib (Fédération internationale de béton, 2013)). This model was improved by Cusatis and
Diluzio [111, 112], based on new experimental evidence. In 2018 it was improved more
substantially by Rahimi-Aghdam et al. [221], in three ways: 1) The humidity dependence of
permeability was separated from the diffusivity by using a more realistic nonlinear des-
orption isotherm; 2) the order-of-magnitude drop of permeability occurring at decreasing
pore humidity was made less steep than it originally was, and was extended below the
50% humidity; and 3) empirical formulae to estimate the permeability parameters from
concrete strength and composition were developed, to make possible realistic estimates
without experimental calibration of these parameters.

Like the Bažant Najjar model, the model of Rahimi-Aghdam et al. [221] postulates
that, at constant temperature, the total moisture flux $j_w$ is driven by the gradient of pore
relative humidity $h$, i.e.,

\[(5.35) \quad j_w = -c_p(h) \nabla h\]
where \( c_p \) is the function giving moisture permeability (kg/m.s). The condition of mass conservation of water reads:

\[
\dot{w}_{\text{tot}} = -\nabla \cdot j_w + \dot{w}_s
\]  

(5.36)

where \( \dot{w}_s = \) rate of water mass consumed by the chemical process of hydration, which is calculated from the rate of hydration degree, \( \dot{\alpha} \), according to Rahimi-Aghdam et al.’s 2017 model. Term \( \dot{w}_s \) represents a distributed sink, leads to self-desiccation, and is particularly important for modern concretes with low \( w/c \) or with silica fume, or both. The self-desiccation and the presence of anticlastic capillary menisci of negative total curvature causes that concrete pores never fully filled by liquid water, even when the pore vapor pressure greatly exceeds the saturation pressure \( p_{\text{sat}}(T) \) [42, p.824]. The hydration degree plays a great role in several equations (for a brief review, see Appendix A, and for details).

For \( 30\% \ll h \ll 100\% \), the desorption isotherm of pore water may be realistically simplified as follows:

\[
\dot{h} = k(\alpha, h) \dot{w}_t
\]  

(5.37)

where \( k(\alpha, h) \) (dimension m\(^3\)/kg) is the reciprocal moisture capacity (i.e., the inverse slope of the isotherm), and \( \alpha \) is the hydration degree, growing with concrete age. Incorporating 5.35, 5.36, and 5.37, one gets the governing equation of moisture diffusion in concrete:

\[
\frac{\partial h}{\partial t} = k(\alpha, h) \nabla \cdot (c_p \nabla h) + \frac{\partial h_s}{\partial t}
\]  

(5.38)
Table 5.1. Common model parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a_0)</td>
<td>0.005q_4</td>
</tr>
<tr>
<td>(c_h)</td>
<td>0.035</td>
</tr>
<tr>
<td>(Q_h/R)</td>
<td>1900</td>
</tr>
<tr>
<td>(C_{S_0}^{q_0})</td>
<td>1.6/q_4</td>
</tr>
<tr>
<td>(c_1)</td>
<td>22.5q_4</td>
</tr>
<tr>
<td>(c_T)</td>
<td>0.012</td>
</tr>
</tbody>
</table>

where the last term on the right-hand side represents the self-desiccation sink, which can be calculated using . The moisture permeability is calculated by an equation of the same form as in the original Bažant-Najjar model:

\[
(5.39) \quad c_p(h, \alpha) = c_1 \left( \beta + \frac{1 - \beta}{1 + \left( \frac{1-h}{1-h_c} \right)^r} \right)
\]

where \(c_1\), \(\beta\), \(h_c\) and \(r\) are four empirical parameters. The new model by Rahimi-Aghdam et al. (2018) provides equations to estimate these parameters from the properties of concrete mix \((a/c\) and \(w/c\)), which makes experimental calibration of these parameters for a given concrete unnecessary; see Appendix B for calculation of permeability.

5.6. Numerical Simulations and Validation by Test Data

To obtain a general model, it is important to fit the proposed model to all the main types of experimental data that exist (which is over a dozen). Many different models could fit a few types of data sets among them, but fitting all of them, with the same parameters, is the only way to get an unambiguous result. Here it is demonstrated for
several important data sets from the literature, dealing with various concretes under diverse environmental conditions.

The available data deal with the basic creep (defined as the creep at no moisture exchange), creep at different temperatures, creep and shrinkage under drying exposure, and transitional thermal creep after a sudden change of temperature. Table 1 summarizes the common parameters that were used for all experiments. In addition, the new model by Rahimi-Aghdam et al. [222] needs only the values of $w/c$ and $a/c$, which are usually reported by the experimenters. The remaining calibration parameters, which are specific to each experiment, are the parameters of long-term creep model, particularly parameters $q_2, q_3, q_4$ and $b_0$ of model B4 (adopted as standard recommendation of RILEM):

\begin{align*}
q_2 &= c_2p_2 \left( \frac{w/c}{0.38} \right)^3 \tag{5.40} \\
q_3 &= c_3p_3 \left( \frac{w/c}{0.38} \right)^{0.4} \left( \frac{a/c}{6} \right)^{-1.1} \tag{5.41} \\
q_3 &= c_4p_4 \left( \frac{w/c}{0.38} \right)^{2.45} \left( \frac{a/c}{6} \right)^{-0.9} \tag{5.42}
\end{align*}

where $c_2, c_3, c_4$ are three calibration parameters. Based on model B4 paper we set $p_2 = 0.0586$, $p_3 = 0.0393$ and $p_4 = 0.034$. Note that for $c_2 = c_3 = c_4$ the equations become the same as in model B4. The difference in optimum values is not surprising because model B4 was not formulated as a point-wise constitutive law. The last parameter, $b_0$, is simply calculated as:

\begin{equation}
b_0 = c_0q_4 \tag{5.43}
\end{equation}
where $c_b$ is a calibration parameter. Note that here most parameters are defined as functions of concrete properties, which reduces the number of unknown parameters to be calibrated by tests. Furthermore, the calibration parameters that are used do not change over a wide range. For instance, $c_2, c_3, c_4$ all vary between 0.6 and 1.5 and for crude estimates can be taken as 1. Thus the present XMPS model does not require more tedious calibration than the original MPS theory. It gives better results because the underlying phenomena are represented more realistically.

5.6.1. Tests of Bryant and Vadhanavikkit (1987)

As mentioned in the Introduction, the main impetus for developing the XMPS was that the original MPS theory predicted an excessive delay of, and a reverse size effect on, the additional creep due to drying, conflicting with the comprehensive tests of Bryant and Vadhanavikkit (1987). Bryant et al. used prismatic and slab specimens of different sizes, both sealed and drying. The concrete had $w/c = 0.47$ and $a/c = 5.1$. During the first two days after casting, the specimens were kept in sealed molds (with no moisture exchange) at temperature $T = 293.15$ K, and then were exposed for 6 days to an environment of the same temperature and 95% relative humidity (RH). Subsequently, groups of specimens were exposed to drying at 60% RH or sealed, and subjected either to no stress ($\sigma = 0$) or to applied compressive stress $\sigma = 7$ MPa parallel to the drying surface. The initial strain readings were taken on day 8, before the RH was lowered. All the sealed specimens were prisms $150 \times 150 \times 600$ mm. The drying specimens were prisms, of dimensions $D \times D \times 600$ mm, and slabs of the same thicknesses $D$, with sizes $D = 100, 150, 200, 300$ and 400 mm.
First we consider the prediction of creep at sealed condition, called the basic creep. Fig. 5.4a compares the experimental vs. simulated creep values of sealed prisms for different loading times. The calibration parameters are: $c_2 = 1$, $c_3 = 1$, $c_4 = 1.1$ and $c_b = 2.5$. As seen, the predictions are satisfactory.

Next we examine the predictions of creep of exposed specimens, prisms and slabs. For each slab, among the 6 faces, the 4 rim faces were sealed and only the two largest faces were exposed, which leads to unidirectional drying across the thickness.

Fig. 5.4b compares the experimental and simulated creep values of a drying slab with $D = 150\text{mm}$, loaded at different ages. As it can be seen, the predicted results are in good agreement. Note that here no extra calibration parameter is used to model the drying process, i.e., the permeability parameters are estimated from Eq. 5.39. It should be mentioned that the predictions usually show some initial error at short times. This error, can be due to several phenomena. Aside from a possible measurement error, these may include considering some short-time creep as instantaneous deformation; effect of
damage; error of model B4 in considering effect of loading time on viscosity; and finally error of the model itself.

After successful predictions of drying creep for the reference size, consider slabs of different sizes (thicknesses). Fig. 5.5a demonstrates correct predictions of the diffusion size effect in drying creep, which means that the drying creep in smaller specimens is faster and that the final value of drying creep is bigger. In this regard, note that Havlšek at Northwestern found the original MPS model to predict, incorrectly, the opposite—a lower final drying creep for smaller specimens, which contradicted the test results and is what motivated the development of XMPS.

Finally we consider the prisms in which only the bottom and top are sealed and the four long faces are exposed to $h = 0.6$. According to the simplification suggested in model B3, the prism can be approximated by an effective cylinder of the same volume/surface ratio, for which the diameter equals $1.09 \times$ the prism side. This approximation, though, did not yield very good results. Therefore, the specimen was simulated in two dimensions as a prism. Exploiting symmetries, only $1/4$ of the prism sufficed for analysis. The simulation results using the real prism achieved closer fits (Fig. 5.5b). Then, different diameters of effective cylinders were tried and, interestingly, the best approximation occurred when the diameter was almost equal to the prism side.

To illustrate the diffusion size effect on the drying part of creep more clearly, the basic creep part is now subtracted. Fig. 5.6 shows the drying part of creep for specimens with different sizes. As it can be seen, the diffusion size effect is significant and the XMPS theory is able to predict it.
Figure 5.5. Experimental vs. predicted results for slab and prisms with different sizes (experiments by Bryant and Vadhanavikkit (1987)); a) drying slabs (unidirectional drying) with different sizes, and b) drying prisms (4 faces exposed and 2 faces sealed) with different sizes.

Figure 5.6. Experimental vs. predicted results for the size effect in drying part of creep. Experiments by Bryant and Vadhanavikkit (1987). a) drying slabs (unidirectional drying) with different sizes, and b) drying prisms (4 faces exposed and 2 faces sealed) with different sizes.

Until now we have analyzed the diffusion size effect in drying creep but, of course, the drying shrinkage is size dependent as well. The cause is the differences in the rate of drying of samples with different size. Fig. 5.7 illustrates the predicted vs. simulated shrinkage.
Figure 5.7. a) Experimental vs. simulated shrinkage, b) Experimental vs. predicted results using the real loading time and effective loading time.

of specimens with different diameters. As seen, for smaller specimens the shrinkage rate is higher and the shrinkage value larger.

Finally, let us analyze the effect of considering in Eq. 5.5 the effective time instead of the real time. Fig. 5.7b shows the results obtained on the basis of both the real loading time and the effective loading time, for an experiment in which the loading and drying times are different. It is seen that better predictions are obtained with the effective loading time.

Note that all the material parameters of the present predictions are the same for all the simulations. This cannot be achieved with other existing models. The same is also true for the fitting of the data for other concretes which follows.

It should be mentioned that often (especially in bending) the cracking damage can make a significant contribution to the Picket effect [61], defined as a difference of the total creep of a drying specimen from the sum of the creep of an identical sealed specimen and of the shrinkage of a load-free identical companion specimen. The reason is that shrinkage
of a load-free specimen is diminished by cracking while in compressed creep specimens the effect of cracking is minimal or zero. This phenomenon is more important for thin specimens, for early ages and for flexure (used in the tests of [61], in which the cracking explained almost a half of the observed Pickett effect). However, in the tests of Bryant et al., the contribution of cracking to the Pickett effect was only about 2%, and thus was neglected in simulations. Nevertheless, the understanding of cracking contribution to the Pickett effect calls for deeper examination in future research.

5.6.2. Tests of Kommendant et al. (1976)

Kommendant, Polivka and Pirtz [170] measured creep for different ages $t'$ at loading and at different temperatures. Two almost identical concrete mixes were used. One mix (Berks) was characterized by $w/c = 0.381$ and $a/c = 4.34$. The second mix (York) was almost the same, with $w/c = 0.384$ and $a/c = 4.03$. The test specimens were cylinders $6\times16$ in. ($15.24\times40.46$ cm), sealed against moisture loss. All the specimens were cured at $23^\circ$C, and 5 days prior to loading the temperature started increasing at a constant rate of $13.33^\circ$C/day until the target value $43^\circ$C or $71^\circ$C was reached. Furthermore, for each temperature, several tests at different ages of loading were conducted. The calibration parameters were: $c_2 = 0.93, c_3 = 1.85, c_4 = 1.18$ and $c_b = 1.25$.

Fig. 5.8 shows the experimental vs. predicted creep values for the York mix at $T = 20^\circ$C. The results are in good agreement with the experimental ones. Note that although the specimens were sealed, the self-desiccation caused the relative humidity to decrease, which may have affected the microprestress value.
Next consider the experiments at $T = 43^\circ$C. Fig. 5.9a presents the predicted creep values for different load durations and for $T = 43^\circ$C. As can be seen, the predictions agree well with the test data well. Note that, in the modeling of the experiments at elevated temperatures, increased values of creep parameters $q_1, q_2, q_3$ were considered, due to the temperature induced compliance increase (Eq. 5.27). Finally we examine the ability of the model to predict the creep values for tests with different temperatures but the same loading time. Fig. 5.9b documents good prediction accuracy.

5.6.3. Tests of York et al. (1970)

York et al. (1970) conducted several basic creep and drying tests. The creep experiments were carried out on cylindrical specimens of $152 \times 406$ cm. The concrete properties were $w/c = 0.43$ and $a/c = 4.62$. Unfortunately, after about 300 days the specimens sealing failed. Therefore, only the data up to 300 days, shown by solid circles, are fitted, and the
Figure 5.9. Experimental vs. predicted results of Kommendant et al. (1976) experiment; a) different loading times at $T = 43 ^\circ C$, and b) different temperatures loaded at $t' = 28$ days.

Figure 5.10. Experimental vs. predicted results; a) experiment by York et al. (1970) for sealed samples with different temperatures, and b) Experiment by Russell and Corley (1978) for drying samples loaded at different ages.

subsequent data, shown as empty circles, are ignored. Fig. 5.10a shows the experimental vs. predicted results. As can be seen, until about $t=300$ days, the predictions are close enough. The calibration parameters are: $c_2 = 0.8, c_3 = 1.3, c_4 = 1.18$ and $c_b = 1.25$. Note that the same activation energies are here used for all concretes.
5.6.4. Tests of Russell and Corley (1978)

These drying creep tests included three ages of concrete ($t = 28, 180$ and $360$ days). The specimens were cylindrical, with the diameter of $15.2$ cm and height of $30.5$ cm; $w/c = 0.45$ and aggregate cement ratio $a/c = 3.95$ by weight), exposed to an environment with $h_{env} = 0.5$ and temperature $T = 23$. Drying began after 7 days of curing in a humidity chamber with $h_{env} = 1$. Fig. 5.10b compares the experimental vs. simulated results for different loading times. The predictions give a close enough agreement. The calibration parameters were: $c_2 = 1.1, c_3 = 1.1, c_4 = 1.1$ and $c_b = 1.25$.

5.6.5. Tests of Di Luzio et al. (2015)

This experimental study focused on basic and drying creep of the modern high-performance concrete used in large-span prestressed bridges. In this experimental investigation, cylindrical specimens (of diameter $150$ mm and height $360$ mm) were used with the environmental relative humidity of $50\%$ and the temperature of $20°C$. Basic and drying creep tests were conducted starting at the age of $2$ and $28$ days. The concrete properties were $w/c = 0.37$ and $a/c = 4$. The interesting aspect about these tests is that the loading started at the early age of only $2$ days. Fig. 5.11a shows the experimental vs. predicted results for basic creep. Fig. 5.11b illustrates the same for drying creep. As can be seen, the results are in good agreement with the test data and indicate that the XMPS theory is able to predict creep correctly even at early ages. The calibration parameters were: $c_2 = 1.35, c_3 = 1.1, c_4 = 1.33$ and $c_b = 1.25$. 
Figure 5.11. Experimental vs. predicted results for the experiments by Di Luzio et al. (2014); a) Sealed specimen loaded at different ages, and b) exposed specimen loaded at different ages.

5.6.6. Tests of L’Hermite et al. (1965)

The comprehensive laboratory study of L’Hermite et al. (1965) included several different types of creep tests. Only the fits of drying creep tests at different humidities are shown here, since good fits of other types of creep tests have already been demonstrated for other data sets. The environmental relative humidities were: $h_{env} = 1$, 0.75 and 0.5. The specimens were prisms 28 cm long, with cross section $7 \text{ cm} \times 7 \text{ cm}$. The concrete mix had $w/c = 0.45$ and $a/c = 3.95$. Fig. 5.12 shows the experimental vs. simulated results for different environmental relative humidity values. Again, the predictions agree well with the experiments. The calibration parameters were: $c_2 = 0.9$, $c_3 = 0.8$, $c_4 = 1.0$ and $c_b = 1.25$.

5.6.7. Tests of Fahmi et al. (1972)

In this test series [121], the temperature was increased during the test. This produced additional creep, called the transitional thermal creep, for both the sealed and drying
specimens. The specimens were hollow cylinders with inner diameter 12.7 cm, outer
diameter of 15.24 cm, and length of 101.6 cm. The concrete mix had $w/c = 0.45$ and
$a/c = 3.95$. The observed creep curves show upward jumps when the temperature is
raised. Fig. 5.13a compares the simulations with the data for basic creep and Fig. 5.13b
does the same for drying creep.

The predictions are satisfactory. To achieve it, it was important to consider all the
mechanisms by which the temperature can change creep rate. Especially, it was important
to consider the change of relative humidity in the pores, caused by the temperature rise.
Otherwise, the predicted jump for the drying condition would have been much bigger
than observed. In addition, it was important to consider the compliance increase due
temperature increase (30% compliance increase of sealed condition was reported). The
calibration parameters were: $c_2 = 0.73, c_3 = 1.2, c_4 = 0.8$ and $c_b = 1.25$. It should
be mentioned that these experiments were previously simulated by Bažant and Cusatis
Figure 5.13. Experimental vs. predicted results for the experiments by Fahmi et al. (1972); a) Specimen in fog room, and b) drying specimen.

5.7. Conclusions

(1) The extended microprestress-solidification theory (XMPS) eliminates two main drawbacks of the original version: 1) the reverse diffusion size effect on the part of creep due to drying, and 2) the excessive delay of the drying part of creep after the drying shrinkage. Both phenomena are now predicted correctly. Predictions of the diffusion size effect on shrinkage are also improved.

(2) Different nano- and macro-scale viscosities are now distinguished. The flow term in XMPS considered to be a function of the macroscale viscosity, which depends on the pore humidity rate, a feature that transpired from earlier MD simulations showing that the apparent viscosity in sliding of two parallel C-S-H sheet should depend on the velocity of molecular layer of water moving between these two sheets.

(3) In the XMPS, the age effect on creep at variable humidity and temperature histories is based not on an empirical effective age (or maturity) but on the
effective hydration time calculated from the growth of hydration degree at each point of the structure (or each integration point of a finite element program).

(4) The temperature change effect on pore relative humidity can be realistically described by a humidity dependent hygrothermic coefficient (introduced in \[27\]).

(5) Empirical formulae for estimating the parameters of the humidity dependence of permeability are developed. They mostly obviate the need for calibrating the permeability law by tests.

(6) The XMPS is fully compatible with the model B4. Unlike the previous version, all the material parameters can be estimated from model B4, making calibration unnecessary in most applications.
CHAPTER 6

Diffusion-Controlled and Creep-Mitigated ASR Damage via Microplane Model

Alkali-silica reaction (ASR) can cause serious expansion and cracking in concrete, resulting in major structural problems and sometimes necessitating demolition. In recent years, several large structures including dams, roads, nuclear power plants, and bridges were influenced by ASR and even completely demolished. Therefore, it is necessary to be able to predict the possible ASR induced damages and try to prevent them. In this chapter, first, the model of Bažant and Steffens for the diffusion controlled the kinetics of ASR is reviewed and used to calculate the rate of production of the ASR gel within the aggregate. Later, a diffusion- and creep-based chemo-mechanical model for calculating the evolution of damage caused in concrete and concrete structures by the alkali-silica reaction (ASR) is presented. Note that the ASR gel expansion in the aggregate and the ITZ causes fracturing damage in the surrounding concrete, which is analyzed by microplane model M7, into which the aging creep of a broad retardation spectrum is incorporated. The gel and the damaged concrete are macroscopically treated as a two-phase (solid-fluid) medium of non-standard type, because of load-bearing but mobile water in nanopores.

6.1. Introduction

The alkali-silica reaction (ASR) afflicts mineral aggregates in concrete if they contain imperfectly crystalline silica. The reaction produces a gel that can imbibe enormous
amount of water. The swelling that results, typically after several months to many years or even decades, is often causing severe strength degradation and fracturing in concrete structures. Since drying arrests the ASR, the worst damage usually occurs in massive structures such as dams, large bridges and nuclear power plant structures. To prevent the ASR damage is one important goal of sustainable design.

The damage due to alkali-silica reaction (ASR) (also called the alkali-aggregate reaction, AAR) was first identified by Stanton in 1942 [248]. Since that time a vast body of literature on this pernicious problem has been accumulated [10, 59, 62, 70, 98, 103, 118, 119, 145, 173, 191, 190, 194, 193, 195, 196, 208, 217, 26, 234, 248, 249, 251, 255, 192, e.g.]. Comprehensive literature reviews have recently been given by Saouma and Xi [234] and Pan et al. [208]. The chemical relations involved in ASR have been presented and discussed in [10, 59, 208, 233, 234]. A review relevant to mathematical modeling of the chemical reaction kinetics prior to 1999 appears in the introduction of [59]. An excellent recent review relevant to the ASR damage simulation is found in the introduction of Cusatis and Alnaggar’s paper [10]. This paper was also the first to provide a general structural analysis of a creep-mitigated ASR damage, in which Cusatis’ lattice discrete particle model (LDPM) was used. All of the ASR gel expansion was in that work assumed to occur in the interparticle contacts, with no delay due to gel diffusion.

The objective of this work is to develop a comprehensive continuum model capable of capturing the main physical aspects of ASR damage, including: 1) the delay of ASR due to diffusion of ASR gel into the pores and expanding cracks within and very near aggregate grains; 2) fracturing of the solid framework of concrete as a two-phase medium caused by fluid expansion in the pores and cracks; 3) oriented character of damage; 4)
mitigation by creep (with chemical aging) of the stresses created by gel expansion 5) effect of alkali content on ASR induced damage; 6) effect of temperature on ASR kinetics and ASR induced expansion; and 7) control of the ASR process by variation of pore humidity due to self-desiccation or external drying.

The ASR reaction occurs at various randomly located discrete sources inside the grain. Penetration of alkali from the cement mortar matrix will first activate the sources near the surface. The time to activate the deeper sources will grow with their depth. To describe the average behavior of many aggregate grains, we introduce a smeared continuum model in which water diffuses radially into a spherical aggregate grain. The ASR reaction is assumed to take place instantly at the spherical front of water penetration, which forms a spherical surface of decreasing radius $z$ (Fig. 6.1c). The reaction is slowed down by the diffusion of water through the aggregate piece, and particularly through the layer of ASR gel already formed. The radial diffusion of water through the barrier of the previously formed ASR gel controls the rate of ASR. This diffusion control captures the effect of grain
size on the reaction rate, typical of all diffusion phenomena. To describe the production of the ASR gel in mineral aggregates, the model of Bažant and Steffens (2000) is adopted. This model is considered as a given input, which predicts the gel mass $M(t)$ per unit area of aggregate surface (kg/m$^2$) extruded into the pores and developing cracks, in the aggregate or matrix, as a function of time $t$. The coupling with ASR gel penetration into the pores of the mortar surrounding the aggregate piece is treated as one-way, i.e., the evolution of gel mass $M(t)$ is considered to be independent of pressure $p(t)$ developed in the pores surrounding the aggregate piece. This is, of course, a simplification, although probably not a major one since the pore pressures sustainable by concrete are apparently not large enough to slow down the alkali-silica reaction appreciably.

6.2. Diffusion of ASR Gel into Pores in Aggregate and Concrete

The expulsion of gel from a gel source into adjacent pores is a mechanism that controls the rate of ASR. It may be treated as a diffusion flow obeying the Darcy law. The flow velocity $\dot{z} = b_D \nabla p$ where $\nabla$ denotes the gradient and $b_D =$ Darcy permeability (dimension m$^3$/s/kg). In detailed analysis, one could now proceed to a point-wise differential equation for the diffusion in coordinate $x$. However, in view of all the uncertainties and inevitable simplifications, it should suffice to replace $\dot{z}$ with the velocity $dx/dt$ of the diffusion front at distance $x$ from the gel source, and the pressure gradient with the average pressure gradient $\nabla p = p/x$. This leads to the simplified Darcy diffusion equation:

\[
\frac{dx}{dt} = b_D \frac{p}{x}
\]
Note that the pressure profile over the gel penetration zone \( x' \in (0, x) \) is doubtless nonlinear. What matters is the flux of mass into the pores at the penetration front, \( x = 0 \). Generally, it is a good approximation to consider the pressure distribution within \((0, x)\) to be a power law, proportional to \((x - x')^u\) (where \( u \) = positive constant). Then the pressure gradient at the front, \((x' \to x)\), is \( \nabla p = p/ux \). Thus Eq. (6.1) may be generalized as

\[
\frac{dx}{dt} = b'_D \frac{p}{x}, \quad b'_D = \frac{b_D}{u}
\]

So the form remains the same for every fixed profile of \( p \), and the fact that the permeability \( b_D \) changes to \( b'_D \) does not matter since its value is a fitting parameter anyway. Frequently considered is a parabolic profile, for which \( u = 2 \) (which corresponds to linear diffusion theory). One or several basic gel sources are imagined to develop in each aggregate grain with reactive silica. From each source, the gel diffuses in several directions (Fig. 6.3a) into pores, interfaces and micro-cracks in the aggregate grain, and into the ITZ (some of the
Figure 6.3. ASR gel transport; (a) ASR gel infiltration and produced global damage, and (b) One ideal gel infiltration path.

gel gets expelled into cracks in the surrounding cement paste but will not be harmful since it will calcify and thus will not imbibe water and swell). The distances \( x_n \) \((n = 1, 2, \ldots, n_t)\) from the numerous gel sources to the fronts of all these diffusion paths vary randomly (Fig. 6.2). On average, we can nevertheless assume \( n_f \) equivalent infiltration paths per unit volume of concrete;

\[
(6.3) \quad dv = a_f^2 b_D n_f^2 \frac{p}{v - v_0} \, dt
\]

Finally, upon denoting \( b/v_{ef} = a_f^2 b_D n_f^2/(v - v_0) \) we get,

\[
(6.4) \quad \frac{dv}{dt} = b \frac{p}{v_{ef}}
\]

where \( v_0 \) is the volume fraction of easily accessible and empty pores, \( a_f \) = average cross section area of infiltration path, \( p \) = average gel pressure, \( b \) = effective Darcy permeability (dimension m\(^2\)/N s or m s/kg), which is proportional to the actual Darcy permeability \( b_D \), and \( v_{ef} \) is the effective volume fraction. In Eq. (6.4), the ratio between
effective permeability and the effective volume fraction is the only unknown parameter to estimate, based on empirical data. The permeability, $b$, is sure to increase with the opening $\delta_c$ of the cracks. According to the crack band model [46, 51], $\delta_c \approx l_0 \epsilon''$ where $l_0$ is the material characteristic length and $\epsilon''$ is the inelastic part of average strain across the band (if it is tensile).

6.3. Unconditionally Stable Algorithm with Gel Pressure Relaxation at Constant Gel Mass During Time Step

Since the transport of ASR gel is considered in the average sense and the different infiltration paths are not treated individually, it is reasonable to assume the volume fraction of calcified gel to depend on the volume ratio of aggregate and cement content, $v_{a/c}$ and, for simplicity, the dependance to be linear. So, the calcified and pressurized pore volume fractions may be calculated as

$$(6.5) \quad v_{ca} = (1 - v_{a/c}) v_{fr} \quad v_p = v - v_{ca}$$

Another issue to address is the pressure profile of pressurized pore volume fraction $v_p$ (Fig. 6.3b). As already discussed, a zero pressure gradient is considered for the easily accessible volume. For the remaining part of $v_p$, we need to choose a reasonable pressure profile. A parabolic profile seems appropriate. Accordingly,

$$(6.6) \quad p = \kappa \frac{v_i - v}{v_0 + \frac{2}{3}v_p}$$

where $\kappa$ is the bulk modulus of the gel, which we consider to be approximately equal to that of water, and factor $2/3$ refers to the area under a parabola (it would be $1/2$ for a
linear profile). Also, since all $v$, $v_i$, and $v_p$ depends on the pressure (we use $v_i$ because the density is pressure dependent), the usual time-step integration algorithm will require very small time steps to remain stable. But ASR evolves slowly over decades.

In analogy with the unconditionally stable exponential algorithm for creep $^{20}$, $^{163}$, the volume fraction of fluid $v_i$ is held constant during the time step $dt$ and the pressure is allowed to relax due to diffusion. At the end of time step, a sudden increase of pressure is calculated. By some mathematical calculation we have,

$$\frac{dp}{dt} + \frac{p}{\tau_p} = 0$$

Here $\tau_p$ plays the role of characteristic time. The advantage of this exponential formula is that, for arbitrarily long time steps, the numerical integration can never yield unbounded $p$. In other words, the computation of pressure is unconditionally stable. This is important for predicting the ASR effects for hundred-year lifetimes. Typically, after an initial period of short steps, the pressure variation becomes slower and slower. The step duration can be increased in geometric progression even if the step duration becomes many months long.

### 6.4. Two-Phase Medium for Loading of Concrete by Pressure in Pores and Cracks

It has been standard to treat porous solids with fluid in the pores and microcracks as the Biot two-phase medium, exemplified by saturated sand. Its volumetric equilibrium relation reads $S_V = \sigma_V - \alpha p$, in which $\sigma_V$ = volumetric stress in the solid phase, calculated from the constitutive model for the solid (the solid phase is the concrete, consisting of
aggregate and mortar); \( S_V = \) total volumetric stress in the two-phase medium, which is used in finite element analysis to calculate the nodal forces; and \( \alpha = \) Biot coefficient approximately equal to the porosity but usually defined such that \( 1 - \alpha \) represents the bulk moduli ratio of the solid without and with the pore fluid \([95]\). Using effective porosity \( \phi \) instead of Biot coefficient \( \alpha \), we may write the incremental volumetric and tensorial two-phase equilibrium relations as follows:

\[
\Delta S_V = \Delta ((1 - \phi)\sigma_V) - \Delta (\phi p) \tag{6.8}
\]
\[
\Delta S_{ij} = \Delta \sigma_{ij} - \delta_{ij} \Delta (\phi p) \tag{6.9}
\]
\[
\Delta (\phi p) = \phi \Delta p + p \Delta \phi \tag{6.10}
\]

where \( i, j = 1, 2, 3 \) are Cartesian tensorial subscripts; \( \delta_{ij} = \) Kronecker delta (unit tensor), and \( \sigma_V = \sigma_{kk}/3 \) (note that \( S_V \) and \( \sigma_V \) are positive for tension, unlike \( p \)). Although Eq. (6.8) has the form of Terzaghi’s effective stress, its physical meaning is different. The effective stress, based on the boundary porosity, is appropriate only for the failure criterion, while Eq. (6.8) applies at all stages of loading.

### 6.5. Effect of Various Loading Conditions on ASR Induced Expansion

Several experimental and numerical studies show that the type of applied stress state, particularly the ratio of various components of applied stress, has a large effect on the ASR-induced expansion in various directions \([8, 10, 137, 174, 195]\). However, the effect on the volume expansion of concrete is minimal. For instance, large compression in one direction simply transfers the expansion to another direction under smaller or zero compression.
The ability of the present ASR model to predict ASR-induced expansion and deterioration at different stress states can be checked by various published experimental data. First we analyze the accelerated laboratory tests of Multon and Toutlemonde, in which the deformation and expansion due to ASR during the test period of 450 days are believed to be comparable to those during 5–50 years in actual structures. In these tests, which involved concrete cylinders of 130 mm in diameter and 240 mm in height, with water-cement ratio 0.5 and cement content 410 kg/m$^3$, the availability of alkali was ensured by dissolving potassium hydroxide in the mixing water to increase the Na$_2$O$_{eq}$ content to 1.25% of the mass of cement. The model was first calibrated the case free expansion. Next we try to predict, with the same material parameters, the Multon and Toutlemonde’s tests with various loadings and confinements. Figs. 6.5 and 6.6 show that the computer predictions match closely the measured axial and radial strains. In particular they confirm that the present model can predict the so-called expansion transfer, i.e., the loading-induced transfer of ASR expansion to another direction.
Figure 6.5. Axial strain-reaction time for: (a) unconfined under 10 MPa axial load, (b) unconfined under 20 MPa axial load, (c) 3 mm confinement under no axial load, (d) 5 mm confinement under no axial load, (e) 3 mm confinement under 10 MPa axial load, (f) 5 mm confinement under 10 MPa axial load, (g) 3 mm confinement under 20 MPa axial load, (h) 5 mm confinement under 20 MPa axial load

In the cylinders confined by tubular steel envelopes, the concrete was considered to slide against the steel. The friction coefficient was not reported. In the simulations, it was assumed to be 0.15.
Figure 6.6. Radial strain–reaction time for: (a) unconfined under 10 MPa axial load, (b) unconfined under 20 MPa axial load, (c) 3 mm confinement under no axial load, (d) 5 mm confinement under no axial load, (e) 3 mm confinement under 10 MPa axial load, (f) 5 mm confinement under 10 MPa axial load, (g) 3 mm confinement under 20 MPa axial load, (h) 5 mm confinement under 20 MPa axial load.

The damage and cracking pattern for various stress states are rather different, as seen in Fig. 6.7. For the unconfined load-free case, the cracks favor no direction. Adding an
6.6. Degradation of Mechanical Properties Caused by ASR

As shown by many studies, the ASR induced expansion produces microcracks and cracks which weaken the concrete \cite{253, 64, 65, 97, 103, 188, 206, 241, 252, 254}. Here we investigate the effect of ASR on: 1) the compressive strength; 2) the tensile strength; and 3) Young’s modulus. We compare the model predictions to the experimental data of Ben Haha \cite{64, 65} from his accelerated tests of concrete prisms of dimensions...
Figure 6.8. Calibrating ASR induced expansion for concrete specimen: (a) simulated specimen and (b) experimental vs. simulated ASR induced expansion.

$70^2 \times 280$ mm, submerged in water. Since the autogenous shrinkage and the swelling were not measured, we must neglect it, although they might have had non-negligible effects, particularly at short times. First we calibrate the model to fit Ben Haha’s measured ASR induced expansion. Having the model calibrated to predict the measured expansion, we can examine its power in predicting the effect of ASR on the mechanical properties. To this end, we analyze the effects of axial load, tensile or compressive, at different stages of the ASR.

6.7. Effect of Alkali Content on ASR Induced Expansion

The alkali content can have big effect on the ASR reaction [138, 240]. The availability of alkali ions and hydroxyl ions is what controls ASR kinetics. It is necessary to assess ASR for concrete with a lower alkali content which is insufficient for a complete reaction. In that case the amount of produce ASR gel may decrease with a factor $f_{alk} < 1$. To this
Figure 6.9. Mechanical properties change due to ASR reaction: (a) compressive strength of affected concrete vs. unaffected one, (b) Tensile strength of affected concrete vs. unaffected one, (c) Young’s modulus of affected concrete vs. unaffected one.

At the end, we introduce the following empirical equation which led to good data fits:

\[ f_{\text{alk}} = \left( \frac{C_{\text{alk}} - C_{\text{alk}}^0}{C_{\text{alk}}^* - C_{\text{alk}}^0} \right)^{1/3} \]  

where \( C_{\text{alk}} \) = alkali content (ratio of the mass of alkali to cement mass), \( C_{\text{alk}}^0 \) = alkali content at which ASR stops and \( C_{\text{alk}}^* \) is the alkali content at which alkali ions are sufficient for complete reaction. In regard to the effect of alkali content, we consider two sets of experiments. The first is the same as that already considered for examining the ASR effect on mechanical properties. The second is Ben Haha’s (2006, 2007) set of tests of saturated mortar prisms of size \( 40 \times 40 \times 160 \), with aggregate size \( \leq 3 \) mm (Fig. 6.10a).

The present model was first calibrated by fitting Ben Haha’s test results for \( C_{\text{alk}} = 1.2\% \) and temperature = 20°C. After this calibration, we predict the effect of alkali content on the ASR expansion.
6.8. Effect of Temperature on ASR Induced Expansion

Like all chemical reactions, ASR kinetics changes significantly with temperature \[159, 166, 204, 173, 232, 252, 213\], and this affects the ASR induced expansion and damage.
The temperature effect is considered to follow the Arrhenius equation for permeability of water through ASR gel. For the experimental comparisons and calibrations, Ben Haha’s tests \[64, 65\] were used again to predict, with the same parameters as calibrated for \(T_0\), the effect of temperature on the role of alkali; see Fig. 6.12a,b for concrete and mortar prisms. The fits are seen to be quite close.

Finally, consider the combined effect of alkali content and temperature when both are different from the calibration experiments. Fig. 6.13a,b compares the experimental and simulated results for concrete and mortar prisms at \(T = 40^\circ\text{C}\), with different alkali contents. Fig. 6.13c demonstrates the same for \(T = 60^\circ\text{C}\).

### 6.9. Effect of Drying on ASR

Availability of water is crucial for ASR. No water—no ASR. The more water, the greater the ASR damage. The role of water is fundamental for three reasons\[59\]:

![Figure 6.12. ASR expansion for concrete and mortar specimens with different temperatures: (a) experimental vs. simulated results for concrete specimens with different temperatures and (b) experimental vs. simulated results for mortar specimens with different temperatures](image)
Figure 6.13. ASR expansion for specimens with different temperature and alkali content respect to calibrated one: (a) simulated results for concrete specimens at T = 40°C. with different alkali contents, (b) simulated results for mortar specimens at T = 40°C. with different alkali contents and (c) simulated results for morta specimens at T = 60°C. with different alkali contents.

1. The pore water acts as the necessary transport medium for the mass transport of hydroxyl and alkali ions required by the reaction.

2. The expansion of the gel is essentially governed by the imbibition of water.

3. For the reaction to continue, water must be supplied by macro-diffusion through the pores of concrete.

A decrease of water supply not only reduces the ASR swelling, but also reduces drastically the diffusivity of cement paste or mortar around the reactive aggregate and thus decelerates the swelling, even by one or two orders of magnitude. To model it, we adopt Bažant and Najjar’s [28] empirical equation (embedded in fib Model Code 2010) for the dependence of local diffusivity $C_i$ of moisture in the mortar or cement paste around the aggregate:

$$C_i = C_{sat} \left( \alpha_0 + \frac{1 - \alpha_0}{1 + \left( \frac{1-h^*}{1-h} \right)^n} \right)$$

(6.12)
Figure 6.14. ASR expansion at different environmental humidity ratios: (a) RH=96%, (b) RH=82%, (c) RH=76% and (d) RH=59%

where $C_{sat}^i = \text{value of } C_i \text{ at } h = 100\%$; $\alpha_0, n$ and $h^*$ are empirical constants. To verify the ASR model for drying concrete, the experiments Poyet et al. [217] conducted at various environmental relative humidity ratios are used. The specimens were mortar cylinders, 16 cm in length and 2 cm in diameter, and the tests were done at $T = 60^\circ C$. Fig. 6.14a shows the evolution of expansion in time for sealed condition and and Fig. 6.14b,c and d compares the experimental and simulated ASR induced expansions at various environmental humidity ratios.
CHAPTER 7

The Enigma of Large-Scale Permeability of Gas Shale: Preexisting or Frac-Induced?

Hydraulic fracturing of oil and gas-bearing rocks, also known as fracking, is an established technology that has been developed gradually since 1947, with no government support until the success has been proven. Although the recent advances in fracking have been nothing less than astonishing, the knowledge of the actual fracturing process is mostly empirical. Therefore, it is important to understand why and how fracking works and are current beliefs are correct or not. One important subject to study is the very significant discrepancy (four orders difference) between the permeability of intact shale and apparent permeability of reservoir shale. In literature, this high permeability discrepancy is attributed to widely open natural fractures. In this chapter, we study the validity of this assumption and try to explain the main reasons for this discrepancy. In this study, it is shown that the creep must close all the cracks tightly (except for residual openings of the order of 10 nm) even if the cracks are propped open by surface asperities. The inevitability of secondary creep (or steady-state flow) is explained theoretically by activation of new creep sites at stress concentrations caused by prior creep deformation.

7.1. Introduction

Extraction of gas (or oil) from deep shale strata is made possible by hydraulic fracturing (aka fracking). The steel casing of a horizontal section of the wellbore, at the depth
Figure 7.1. Parallel hydraulic cracks as predicted in current practice (a single crack per perforation cluster, with no branching.), pictured before localization [58].

of about 3 km, is perforated by clusters of shaped explosive charges. The perforations provide inlets for pressurized frac water. According to computer simulations, this causes one vertical hydraulic fracture (or frac, crack) to grow vertically from each perforation cluster. Comparisons of gas production rates for various frac spacings along the horizontal wellbore (Fig. 7.1) [244] indicated that the optimal spacing of these clusters (and fracs) is about 10 m (or up to 20 m).

Fracture mechanics studies show that the hydraulic cracks cannot branch. However, the 10 m spacing of the hydraulic fractures is so large that, according to the permeability values observed on slices of drilled shale cores tested in the laboratory, the gas extraction from shale would take about 100,000 years, as shown in [58]. Yet, in reality, it takes less than about ten years.

To explain this perplexing discrepancy, it has generally been assumed that, despite the large tectonic stress in the shale stratum, there is a huge difference between the
large-scale (or regional) permeability of shale and the small-scale (or local) permeability measured in the laboratory. To match the gas production rates observed at the wellhead, the large-scale permeability of shale must be assumed to be (even at a typical 3 km depth) about 10,000-times higher than the (properly measured) laboratory value (sometimes the permeability increase is said to be ‘only’ 1000-fold or 100-fold, but this must be due to permeability testing on surface outcrops or in unrealistic conditions, e.g., without the correct triaxial confining pressures). The 10,000-fold discrepancy has been vaguely explained by preexisting natural cracks and joints, whose typical spacing is roughly 0.1 m, and can be up to 1 m.

Here it is argued that this explanation is dubious. The opening width of these natural cracks at the typical depth of 3 km must be zero or below 100 nm, which cannot contribute appreciably to the large-scale permeability. Rather, the explanation lies in the frac process itself, which is opening new as well as preexisting cracks of approximately 0.1 m spacing. This process has been theoretically justified and predicted by the recently advanced three-phase medium theory [92].

7.2. Problematic Current Consensus

The question is whether or not large primary cracks can branch laterally, to produce hydraulic cracks of much smaller spacing. According to fracture mechanics, cracks can branch at the tip only if they propagate at a speed greater than about 40% of the Rayleigh wave speed [272, 227]. But the frac process is static, as it takes one to several hours. So crack tip branching is impossible. Indeed, all the published calculations, as well as softwares, based on linear or cohesive fracture mechanics or the discrete element model
(DEM) predict propagation of parallel hydraulic cracks, a single crack per perforation cluster, and no branching (Fig. 7.1).

What is needed is an initiation and propagation of lateral vertical secondary cracks emanating from the walls of the primary vertical cracks, and of subsequent tertiary cracks emanating from the walls of the secondary cracks. However, according to linear elastic fracture mechanics (LEFM), no cracks can initiate from a smooth surface of the primary crack wall. Rocks, though, are quasibrittle materials, characterized by a large fracture process zone. So, one must use the cohesive (or quasibrittle) fracture mechanics, in which cracks are initiated when the material tensile strength gets exceeded. Lateral cracks in vertical planes normal to the primary vertical cracks will thus initiate if the normal stress parallel to the wall of the primary crack exceeds the tensile strength.

However, fluid pressure in a crack produces no tensile stresses along the crack walls \[12\] e.g., Consequently, commercial softwares (marketed, e.g., by Schlumberger or NSI technologies \[235, 257\]) predict no lateral branching of the vertical primary cracks (often simulated as narrow crack bands if the software uses the discrete element model, DEM). Thus, in accord with the existing commercial software, the consensus has been that no secondary cracks can form, and that the only cracks created by hydraulic fracturing are those emanating from the perforation clusters on the horizontal wellbore casing, which are 10 m apart or more.

This consensus has, however, led to a dilemma: Diffusion analysis based on the values of gas permeability of shale measured in the laboratory (typically on the order of $10^{-21}$ m$^2$) \[231\] would predict the halftime of the history of gas production to be about 100,000 years, while the halftime of the gas production observed at the wellhead is just a few years
Based on this evidence, it has been hypothesized that the large-scale permeability of shale mass ought to have a value about 10,000-times larger than the properly measured laboratory value (sometimes the increase is thought to be ‘only’ 1000-fold or 100-fold, but this is likely due to oversimplified permeability testing).

As an explanation, the 10,000 fold increase of permeability has generally been attributed to the preexisting system of natural cracks and joints. For a long time, no other explanation has been known, even though it has never been explained how, at the depth of several kilometers, the natural cracks and joints in shale, tens to a few hundred million years old, could have kept their opening widths wide enough to cause such a huge permeability boost.

7.3. Choice of Two Hypotheses

So, to explain the gas production rate at wellhead, we must now make a choice between two hypotheses:

(1) **Hypothesis I.** There is no hydraulic crack branching (as predicted by current commercial softwares), but the preexisting natural cracks and joints have, prior to fracking, a sufficient opening width to boost the large-scale permeability of shale mass by about 4 orders of magnitude.

(2) **Hypothesis II.** The preexisting natural cracks or joints are either perfectly closed or too narrow to boost the large-scale permeability, but the frac process opens the preexisting closed cracks and possibly also creates new hydraulic cracks in intact shale. This produces a dense system of branched cracks whose spacing
and opening width suffice to boost the large-scale permeability by about 4 orders of magnitude.

Here it will be argued that the latter is far more likely. It will make a big difference for the understanding and control of the frac process, though little difference for the long-term forecasts of gas production.

**7.4. Gas Permeability of Shale and Errors in Its Testing**

This key property varies significantly from one shale to another. For one and the same shale, the measured permeability can vary even by orders of magnitude, depending on the method of measurement. Since attaining a steady state of gas flow takes a very long time, transient methods of gas permeability measurement are preferred. The pressure-decay profile method and the pulse-decay method both use a slice of a drilled core, and the third, the pressure-decay method, uses crushed rock. Regarding the first two methods, the following points should be noted:

1. The shale is anisotropic. The gas escapes into the hydraulic cracks predominantly along the bedding planes, which are the planes of prevailing orientation of the nano-platelets of clay minerals. The permeability in transverse direction, which is an order of magnitude lower, is, therefore, virtually irrelevant.

2. The permeability must be measured on drilled cores extracted from the shale stratum, typically at 3 km depth (the permeability of the surface outcrops can be orders of magnitude higher).

3. The microcracks in gas reservoirs must be closed (as argued later) because the shale has been, for tens of millions of years, under high overburden and tectonic pressures (and at temperatures around 80°C). The test specimens should be
be kept at that same temperature. They must also be subjected to the same confining pressure (about 40 and 80 MPa, at the depth of about 3 km) [275], because pressure decrease is known to increase the gas permeability significantly [172, 116, 90, 102, 230] (by an order of magnitude for the data in Fig. 7.2a).

(4) Creep due to confining stress decreases the gas permeability [96] because it tends to close any flow nano-channels that may exist. Therefore, the confining stress should be applied for a long enough time (perhaps for 1 month) [102, 96](see, e.g., the data in Fig. 7.2b).

(5) The permeability must be measured at an unaltered moisture state, i.e., at no drying, not only because the nanopore water content greatly hinders the gas flow but also because a pore water loss causes an increase of the effective compressive stress in the solid phase, and thus a decrease of pore widths [132, 122].

(6) The analysis of gas flow must take into account the slip flow, or Klinkenberg effect [169, 165, 131, 189], in the nanopores that are not much wider than the gas adsorption layers on pore surfaces (this leads to a modified gas transport equation).

To make the tests easier, these conditions have often been disobeyed. Then the permeability of shale stratum may be overestimated by several orders of magnitude. Such simplified tests could be useful only if the ratio of conversion to the actual permeability value in the deep shale stratum were established a priori.

The measurements on crushed shale do not satisfy most of these conditions. Nevertheless, the permeability overestimation may be milder than with the other methods.
Figure 7.2. Permeability of shale; a) the effect of volumetric (or hydrostatic) confining stress (test data reported in [90]); and b) the effect of creep (test data reported in [96]).

The reason is that the shale gets fragmented along surfaces that pass through the largest pores, thereby eliminating these pores.
7.5. Opening Width of Preexisting Natural Joints or Cracks Required by Hypothesis I

A glaring question: To boost large-scale permeability 10,000-times, what should be the opening width of the preexisting cracks or joints? (in the case of a crack filled by calcite deposit, one must consider a crack within the calcite or a crack between the calcite and the shale wall).

Figure 7.3. Idealized array of parallel planar preexisting natural cracks, assumed to calculate gas transport to adjacent primary hydraulic cracks spaced at $L = 10 \text{ m}$

To answer it, imagine an array of preexisting planar cracks of width $h$ and regular spacing $s$, parallel to the bedding planes. These cracks connect two adjacent vertical
primary hydraulic cracks at distance \( L = 10 \text{ m} \) (Fig. 7.3). As confirmed by the present calculations, the required crack width is expected to be big enough to make the nanoscale slip flow (or Klinkenberg effect) negligible. Then, assuming a viscous (or Poiseuille) flow of gas, we can calculate the effective permeability of the crack system, per unit cross section area of shale, as follows:

\[
\kappa_h = k_t \frac{h^2 h}{12 s}
\]  

(7.1)

Here \( k_t \) (< 1, assumed to be 0.5) is the tortuosity factor. It accounts for the fact the crack surface is rough and that the gas flow is obstructed by asperities or fragments bridging the crack faces.

Consider now that the permeability of intact shale, measured properly in the laboratory, is \( k_1 = 10^{-6} \text{ mD} \) (where mD = millidarcy = \( 10^{-15} \text{ m}^2 \)). Then, assuming \( k_t = 0.5 \), the ratio \( \kappa_h/\kappa_1 = 10,000 \) is obtained for the crack width:

\[
h \approx 2.89 \mu\text{m} \text{ if } s = 0.1 \text{ m}, h \approx 6.21 \mu\text{m} \text{ if } s = 1 \text{ m}
\]  

(7.2)

These opening widths are 24 or 240 times bigger, respectively, than the average width of the nanopores filled by kerogen with gas. How come that natural cracks of opening 2.89 \( \mu\text{m} \) or bigger are not evident in the lab, under the microscope? Doubtless it is because, during the extraction of the deep drilled core and its transfer to the surface, the cracks opened more. One of the reasons surely is the loss of confining pressure.
7.6. Can Gas Permeability be Inferred from Tests of Water Permeability in Deep Strata?

The ‘large-scale’ water permeability measured in deep shale strata has been shown to be 2 to 4 orders of magnitude higher than the ‘local’ water permeability measured on drilled cores in the laboratory [76, 77, 152]. The discrepancy is attributed to natural faults [261, 276], and is often claimed to explain the observed 10,000 fold boost in gas permeability. However, several differences between water and gas permeability negate this claim.

1) By contrast to small slices of intact shale in the lab, the gas in deep shale mass moves as part of a two-phase transport of gas and water. Studies of such transport in shale, as well as coal [186, 237, 141, 209, 133], show strong effects of viscosity, wettability and surface slippage factor. Especially, they reveal an enormous gas permeability increase with a decreasing saturation degree (which is a behavior opposite to nano-scale water permeability). In the lab, usually the shale specimen is partially or fully dried. At depth, by contrast, the shale pores (excluding those occupied by compressed gas and organic matter, typically representing 5%–20% of shale volume) are almost completely saturated by water. Its pressure equals the hydrostatic pressure for the particular depth, typically about 30 MPa (note that, as generally agreed, the pore water pressure in rock equals the hydrostatic pressure, up to the depth of at least 10 km).

2) The drying exposure in the lab can cause the gas permeability to increase by two orders of magnitude.
(3) Transport of gas through water-filled pores is extremely slow, with effective permeability only about $10^{-25} \text{m}^2$ [143, 144, 11]. While for soluble gases the transport through water occurs by molecular diffusion, the shale gas, mostly methane, tends to form nano-bubbles because it is insoluble in water. This makes the gas permeability even lower.

(4) The gas transport is further inhibited by water blocking narrow throats on the flow passages through shale mass. The throats probably have the same width as the gas-filled pores in intact shale, which is 0.5 to 20 nm. Because the shale minerals are hydrophillic, part or all of the width of such throats must be filled and blocked by adsorbed water layers. How thick? Four layers on each of the opposite throat surfaces (each having one $\text{H}_2\text{O}$ molecule thickness, 0.263 nm) is a minimal estimate (because in cement hydrates, which are more hydrophillic, the adsorbed water forms 5 layers at saturation) [21] e.g.). Thus all the throats < 2.1 nm wide are completely filled by adsorbed water, and even in throats < 20 nm wide the adsorbed water plays a role. Now the point is that for gas nano-bubbles, it must be next to impossible to move through the throats filled by adsorbed water layers, by displacing adsorbed water molecules which are far less mobile than free water molecules (as their lingering times attain about $10^5$ periods of atomic thermal vibration).

(5) The confining pressure, too, suppresses the gas phase permeability, and does so by several orders of magnitude [133] (although the absolute permeability suppression is several times less, due to the high compressibility of gas). To compare
the permeabilities $\kappa_{rl}$ and $\kappa_{rg}$ of the liquid and gas phases, it is found [91] that, approximately, $\kappa_{rl} \propto S^n$ and $\kappa_{rg} \propto (1 - S)^n$ where $S =$ saturation degree.

(6) One more effect complicating the permeability measurements is the swelling of shale (and pores) caused by water imbibition [236]. This phenomenon may increase water permeability.

To conclude, the answer to the question posed in the heading must be no.

7.7. Age of Preexisting Natural Fractures

To decide whether the natural cracks or joints, after being formed in some tectonic upheavals, could have remained open up to now, we must consider their age. The age of various deep shale strata varies between 100 and 300 million years. The natural fractures seen in the shale surely did not form during the last century. They must have been opened (or reopened) by various seismic or tectonic upheavals occurring at a roughly uniform frequency over the entire age of the shale formation.

Therefore, the average age of the natural cracks and joints must be at least 50 million years. Over such a time span, the creep and flow of shale cannot be neglected.

The past tectonic upheavals doubtless produced mostly shear cracks. Due to their roughness, the slip on the cracks is accompanied by dilatancy. The dilatancy leads to interrupted openings of the shear cracks or slip planes, propped by asperities in contact with the opposite faces. The question is whether these opening can be sustained, and for how long.
7.8. Creep and Flow of Shale on the Geologic Time Scale

Although some materials, such as perfect crystals or polycrystalline metals at low enough temperatures and not too high stress, do not creep, most others do, sedimentary, metamorphic and polycrystalline rocks included.

Creep of all materials under constant stress may generally be subdivided into three phases (Fig. 7.4a):

(1) The primary, or transient, creep, in which the creep rate is decaying;
(2) the secondary, or steady-state, creep, also called the flow, in which the strain rate remains constant; and
(3) the tertiary creep, in which the creep accelerates and leads to failure. The tertiary creep is not a strict materials property but is intertwined with a structural failure process dependent on geometry, and is irrelevant to our problem (since the shale stratum is not about to collapse).

Considering the geologic time span, we can generally describe the total creep (transient plus flow) of the lithosphere rocks under constant stress $\sigma$ by the equation:

$$\epsilon(t) = \epsilon_{\text{trans}}(t) + \epsilon_{\text{flow}}(t) = \frac{\sigma}{E} \left[ 1 + \beta \left( \frac{t}{\tau_1} \right)^n + \gamma \left( \frac{t}{\tau_2} \right)^m \right] + \xi \left( \frac{\sigma}{E} \right)^r \frac{t}{\tau_M}$$

in which the last term expands the equation used in [197]; $\epsilon = \text{total strain}, \eta = \text{flow viscosity};$ and $E, r, \beta, \gamma, \xi, n, m, \tau_1, \tau_2$ and $\tau_M =$ material constants to be determined from laboratory tests and geologic observations (they all depend on temperature); $E$ is the instantaneous elastic modulus (for the sake of simplicity, the shale is considered as isotropic, with $E$ characterizing the stiffness in the vertical direction).
Figure 7.4. (a) Three phases of creep evolution under constant stress; (b) creep evolution over 200 million years when only the primary creep terms are considered; (c) the same when the secondary creep (or flow) is included
The first two terms, of exponents $n$ and $m$, represent the primary (or transient) creep, and the last term the secondary, steady-state, creep (or flow). The linear dependence of $\epsilon(t)$ on $\sigma$ in primary creep is a simplification, but probably an acceptable one (especially for the initial creep), and Boltzmann’s superposition principle is approximately applicable, as widely accepted in geology for the rocks in the lithosphere [72, 224].

The term with exponent $n$, and $\tau_1 \approx 1$ day, gives the short-term, rapidly decaying, part of transient creep (in which the number of initially activated creep sites is getting exhausted fast). According to the published creep measurements on Haynesville and Eagle Ford shales, most of which lasted only a few hours [246, 245] and others only several weeks [226], the exponent, $n$, of the initial transient creep of shale appears to lie between 0.05 and 0.20. For $n = 0.05$, which is the value observed in [246, 245, 226], the initial creep is very close to the logarithmic creep, which was previously considered for the lithosphere rocks in general [197]). For Haynesvile shale, recent experiments [226] of a few days in duration, furnished $n = 0.178$ (the other parameters being $E = 16.7$ GPa, $\beta = 2.48 \cdot 10^{-6}$ and $\tau_1 = 1$ day). Since $n = 0.05$ gives the fastest creep rate decay, this value is used here, to get a conservative, minimalist, estimate of crack closure.

Geologic evidence [197, 72], based mainly on post-glaciation rebound of the Earth mantle, suggests that exponent $m$ of the second primary creep term is much higher, between 1/3 and 1/2, the former corresponding to what is known as the Andrade creep law [197, 72]. Since the effective viscosity decreases with depth, the value of 1/3 seems more realistic for the upper lithosphere, and is used in the present calculations for the minimalist assessment of crack closure.
As sketched in Fig. 7.4, the transient (or primary) creep eventually transits to the secondary, steady-state, creep, or viscous flow, which is given by the last term in Eq. (7.3) having the time exponent of 1. According to geologic literature [87, 168, 198], the stress exponent, \( r \), in steady-state creep is between 2 and 3.

Based on geologic studies [224, e.g.], the transition from primary to secondary creep is doubtless gradual and is centered at a time characterized approximately by the Maxwell time, \( \tau_M = \frac{\eta}{E} \), which represents the time at which the flow strain becomes equal to the elastic strain, \( \sigma/E \). On the other hand, some laboratory studies suggest that the transition to steady-state creep may be centered at a certain fixed strain (approximately 0.01) rather than at a fixed time, \( \tau_M \), regardless of the applied constant stress [72]. Accordingly, the higher the sustained stress, the shorter would be the transient creep stage. This issue, however, is unimportant for the present conclusions.

The viscosity of flow over tens of millions of years can be estimated only indirectly. Geologists estimate the average viscosity \( \eta \) of the Earth mantle (about 40 to 280 km thick) to be of the order of \( 10^{21} \) Pa s, with the extreme estimate as high as \( 10^{23} \) Pa s [224, p.198-199]. For the elastic modulus of shale, \( E \approx 17.6GPa \), this would give Maxwell time \( \tau_M = \frac{\eta}{E} \approx 2,000 - 200,000 \) years. Since the viscosity averaging along the lithosphere includes igneous and other rocks with a much smaller creep than shale, we will consider the Maxwell time as

\[
(7.4) \quad \tau_M = \frac{\eta}{E} \approx 1,000 \text{ years} - 100,000 \text{ years}
\]

The higher limit is a conservative choice for estimating the maximum possible time to crack closure.
The foregoing broad range of \( \tau_M \) is considered to compensate for various uncertainties. For example, it is hard to determine how much of the viscous resistance of the mantle is contributed by the lithosphere (i.e., the Earth crust plus asthenosphere, or the upper mantle). For processes of shorter durations, the lithosphere is in geology considered as elastic (or viscoelastic), and subject to brittle fracture, but for stresses of long enough durations it surely exhibits flow.

Along the lithosphere (consisting of the crust and upper mantle), the segments of (not too hot) crystalline (igneous) rocks would probably exhibit little creep or flow, even over a 100 million years span. Since the series coupling of various segments along the mantle implies summation of their inverse viscosities \( [72] \), the viscosity of the shale segments is doubtless much lower than the average. This means that the estimate of \( \tau_M \) in Eq. (7.4) is probably very much on the high side.

Further note that that the temperature increase with increasing depth in the lithosphere reduces the viscosity of solid rock by about an order of magnitude. Hence, the tectonic force must be concentrated approximately in the upper layer of the lithosphere, of about 10 km in thickness. Since the viscosities are additive in parallel coupling, this again implies that the viscosity of the layer should be much higher than its average over the whole thickness of the lithosphere.

In the logarithmic plot of Fig. 7.4c, the initial primary (or viscoelastic) strain growth (i.e., the primary creep) is shown as the initial straight line of slope \( n = 0.05 \). This slope transits to slope \( m = 0.3 \) of the second part of primary creep. Then, centered at \( \tau_M \), the primary creep gradually transits to viscous flow, or secondary creep, which finally approaches a straight line of slope 1. To cover all possibilities, we consider in calculations
three orders of magnitude of Maxwell times:

\[ \tau_M = 10^3, 10^4, 10^5 \text{ years} \]

(7.5)

As for \( \tau_2 \), a reasonable value should be close neither to \( \tau_M \) nor to \( \tau_1 \). It should be somewhere in the middle between \( \tau_M \) and \( \tau_1 \) in the log \( t \) scale. So we will use

\[ \tau_2 = \sqrt{\tau_M} = 10^{3/2}, 10^2, 10^{5/2} \text{ years} \]

with 1000 years as the mean guess (in Fig. 7.4, neither the different values of \( \tau_2 \) nor the two terms of primary creep are distinguished, to keep the trends uncluttered; distinguishing them would make negligible difference).

7.9. Proposed Mechanism of Shale Creep and Its Transition from Primary to Secondary Creep

The transition of primary creep to steady-state flow of shale and of other lithosphere rocks at temperatures far below melting has never been experimentally observed, and never will. So the transition must be explained theoretically. Proposed here is an nanoscale mechanism justifying this transition.

For polycrystalline metals, specific simple examples of the atomistic creep mechanism are the Nabaro-Herring theory \[101\] of bulk vacancy diffusion within the crystal grains, the Coble theory of vacancy diffusion along grain boundaries, and the Harper-Dorn theory of vacancy controlled dislocation climb \[94\]. In shale (like in hydrated cement), the atomistic mechanism of creep is more complicated. It probably consists mainly of progressive nano-scale slips between adjacent nano-platelets of clay minerals.
Figure 7.5. Active creep sites (left) and newly created creep sites (right) of bond breakage at stress concentrations in the nanoscale of shale subjected to shear under confining pressure; (a) during the primary (or transient) creep; (b) during the secondary creep (or steady-state flow)

In general, the increments of creep deformation are caused by breakages and restorations of interatomic bonds at sites of high stress concentration, which may be called the
creep sites (they are marked by x-points in Fig. 7.5, where (a) and (b) give the view normal to bedding planes, (c) the view across the bedding planes, with the short lines showing the clay nano-platelets. After each slip, bond restorations must occur, or else the unloading stiffness would decrease (which would be the case of material damage rather than viscoelasticity). The breakages relax the stress concentration in the nano-structure. By transferring the applied load to nearby locations, the slips due to bond breakages must be assumed to create new creep sites of high stress concentration (Fig. 7.5a,b on the right).

In primary creep, the creep rate decays as the number, \( \nu_1 \), of initially activated creep sites per unit volume of material and per unit time is gradually getting exhausted (Fig. 7.5). If no new creep sites with high stress concentrations were created, no creep sites would soon be left and the creep would eventually stop. This is unlikely, though.

During a creep deformation increment, the stress concentrations under constant load are getting transferred to new locations, which creates new creep sites (Fig. 7.5a,b on the right). So,

\[
(7.7) \quad \nu_1 > \nu_2 > 0
\]

where \( \nu_2 \) is the number of new creep sites created by the deformation under constant load per unit volume of material and per unit time. The difference \( \nu_1 - \nu_2 \) gradually decreases, which explains the increase of time exponent from \( n \) to \( m \) in Eq. (7.3). The primary (transient) creep eventually terminates and the secondary (steady-state) creep, or flow,
begins once both numbers become equal. So,

\[(7.8) \quad \text{for secondary creep: } \nu_1 = \nu_2 > 0\]

Is the transition to secondary creep inevitable? If it did not exist, then the deformation increments would have to be creating no new creep sites, i.e., \(\nu_2 = 0\). But this is improbable. If creep deformation increments under constant load create new high stress concentrations, new creep sites must form (Fig. 7.5). Thus the secondary creep appears to be inevitable. The only question, yet the most difficult one, is when this transition occurs.

7.10. Closure of Open Cracks Propped by Asperities over Geologic Times

The shale is typically intersected by systems of approximately parallel and equidistant natural cracks or joints. In surface outcrops, these cracks or joints look to be open. So they do on the deep drilled cores brought to the surface, as the confining pressure is reduced to zero. But they were doubtless tightly closed before being cored from the deep stratum.

The spacing of natural cracks or joints varies, among different sites, from about 0.1 m to 1 m or more [126]. Let us now examine whether a preexisting natural crack or joint could have a wide enough opening and keep it for a long enough time.

**Estimate of closing of elliptical channel.** To this end, consider an infinite two-dimensional space in plane strain, containing an elongated elliptical hole of length \(2a\) and maximum width \(h = 2b\). The hole is imagined to represent the cross section of a preexisting flow channel (Fig. 7.6). For simplicity of calculations, consider the shale as
isotropic, with elastic modulus $E = 16.7$ GPa and Poisson ratio $\nu = 0.3$. Imposed at infinity are the horizontal principal tectonic stresses, $\sigma_h = 30$ MPa and $\sigma_H = 40$ MPa. The major axis of the ellipse is normal to the smaller tectonic stress, $\sigma_h$.

The initial elliptical hole (Fig. 7.6), considered to have been created tens of millions of years ago, could not have had proportions that would make it collapse immediately.

We assume that $a > b$ where $a, b$ are the major and minor axes of the ellipse (in the $x$ and $y$ directions, Fig. 7.6). Stress $\sigma_{yy}$ at the sharper apex must exhaust neither the
compression strength, \( f_c \), nor the tensile strength, \( f_t \), and the same applies to the flatter apex. According to Inglis’ solution \cite{153, 260}, the conditions for the stresses at the apices are:

\[
\begin{align*}
\sigma_{xx} & > f_t = \sigma_H \left(1 + 2 \frac{a}{b}\right) - \sigma_h > -f_c \quad (7.9) \\
\sigma_{yy} & > f_t = -\sigma_H + \sigma_h \left(1 + 2 \frac{a}{b}\right) > -f_c \quad (7.10)
\end{align*}
\]

where both \( f_t \) and \( f_c \) are positive, while stresses \( \sigma_{xx} \) and \( \sigma_{yy} \) are positive for tension and negative for compression.

We expect the decisive inequality to be \( \sigma_{yy} > f_c \) at the sharper apex (which is checked later). Thus we have for the aspect ratio \( a/b \) the limitation:

\[
\frac{a}{b} < \frac{f_c + \sigma_H - \sigma_h}{2\sigma_h} \quad (7.11)
\]

Consider the shale to have the short-time uniaxial compression and tensile strengths \( f_c' = 140 \text{ MPa} \) and \( f_t' = 10 \text{ MPa} \). For sustained loading, assume, by analogy with concrete, that the strengths for sustained long-time loading are reduced to 80% in compression and to 70% in tension, and that the biaxiality of stress (due to plane strain state) increases the strength in compression by 10% and in tension by nothing. Thus the effective strength values to substitute into Eq. (7.11) are \( f_c = 0.88 f_c' = 123.2 \text{ MPa} \) and \( f_t = 0.7 f_t' = 7.0 \text{ MPa} \). For the tectonic loading we consider \( \sigma_H = 40 \text{ MPa} \) and \( \sigma_h = 30 \text{ MPa} \) (compression being positive for \( \sigma_h \) and \( \sigma_H \)). Substitution of these values into Eq. (7.11) yields the following
maximum possible aspect ratio of the initial elliptical hole (or fluid flow conduit):

\[
\frac{a}{b} = 2.22
\]  

(7.12)

Checking with this ratio \(a/b\) the remaining three inequalities in (7.9) and (7.10), one finds them satisfied.

Note that the strength limits restrict only the initial shape of the elliptical hole but not its size.

Although the initial small deflections due to elasticity and initial primary creep could be obtained analytically, we are interested in the complete closing of the hole, for which finite strains must be considered. The finite element software Abaqus, having the facility to simulate the contact of opposite faces of the collapsing hole, has been used for this purpose.

Since, in view of all the other crude approximations, high accuracy is not needed, it suffices to treat the creep and flow deformations as the elastic deformation for effective modulus \(E_{ef}(t) = 1/J(t)\) and a constant Poisson ratio, where \(J(t) = \epsilon(t)/\sigma = \) compliance function of shale. For the sake of simplicity, we consider the creep to be linearly viscoelastic, given by Eq. (7.3) for \(\sigma = \sigma_h\), i.e.,

\[
J(t) = \frac{1}{E} \left[ 1 + \beta \left( \frac{t}{\tau_1} \right)^n + \gamma \left( \frac{t}{\tau_2} \right)^m \right] + \xi \frac{\sigma_h^{r-1}}{E^r} \frac{t}{\tau_M}
\]  

(7.13)

where \(\beta = 2.48 \cdot 10^{-6}, \tau_1 = 1\) day, \(\gamma = 1.0 \cdot 10^{-5}, \xi = 0.5\) and \(r = 2.5\).

The computed subsequent profiles of the closing of the elliptical hole (or conduit) are plotted in Fig. 7.6. Since the profile evolutions for different \(\tau_M\) values are similar, only one figure suffices. The times to reach each profile are in the figure indicated for \(\tau_M =\)
1,000 days and 100,000 days. The times, $t_c$, for the hole to close completely are obtained as:

$$
\text{for } \tau_M = 10^3 \text{ years: } t_c = 2.10 \times 10^5 \text{ years}
$$

(7.14) \hspace{1cm} \text{for } \tau_M = 10^5 \text{ years: } t_c = 9.36 \times 10^6 \text{ years (upper bounds)}

These time estimates represent upper bounds. In reality, the times to close the hole would be much shorter because we neglected the increased flow due to stress concentrations near the ellipse surface, especially near the sharper apex (where $\sigma_{yy} = 123 \text{ MPa} = 4.11\sigma_h$). These concentrations would make the nonlinear flow term much greater, and thus the times to closing much shorter (according to the assumed creep law). However, an accurate calculation of creep closing times is not important because even the upper bounds in Eq. (7.14) represent small fractions of the average age of natural cracks in the shale stratum ($> 50$ million years).

**Estimate of closing of propped flat crack.** Another case that can be easily calculated is a flat open crack of a large extent propped by surface asperities formed during its creation by some seismic event in the distant past. To allow simple calculation, consider the two-dimensional plane-strain problem of an infinite initial crack of width $h_c$ in infinite plane, propped at intervals $L_q$ by pillar walls of width $L_p$; see Fig. [7.7] (1). The shale properties are the same as before.

The pillar walls are subjected to the average compression stress $\sigma_p$, much higher than $\sigma_h$. Therefore a compliance function with a greater flow term is considered for the pillar
Figure 7.7. (a) Idealization of planar crack in shale kept open by asperities imagined as rectangular pillar walls; (b) Subsequent states of creep closure for the expected and greatly increased values of Maxwell time $\tau_M$ of shale walls,

$$J(t) = \frac{1}{E} \left[ 1 + \beta \left( \frac{t}{\tau_1} \right)^n + \gamma \left( \frac{t}{\tau_2} \right)^m \right] + \frac{\xi}{E'} \frac{t}{\tau_M}, \quad \sigma_p = \sigma_h \frac{L_g}{L_p}$$
while \( J(t) \) according to Eq. (7.13) is considered for all the rest, i.e., the increased flow due to the stress concentrations near the corners is neglected, which causes the closing times estimate to be an upper bound. The maximum possible ratio \( L_g/L_p \) is such that the pillar wall is at the limit of crushing, i.e., the stress in the pillar walls would not exceed the compression strength \( f_c \). This condition yields \( L_g/L_p = 4 \).

The Abaqus finite element program with finite strain and a contact algorithm was used to calculate the progressive closing of the cracks; see Fig. 7.7, which also gives the times to reach subsequent closure states for the expected and extreme estimates of \( \tau_M \). The times needed for a virtually complete closure are:

\[
\begin{align*}
\text{for } \tau_M = 10^3 \text{ years: } t_c &= 5,500 \text{ years} \\
(7.16) \quad \text{for } \tau_M = 10^5 \text{ years: } t_c &= 35,600 \text{ years (upper bounds)}
\end{align*}
\]

Again it is found that the pillar walls, simulating crack surface asperities, cannot prevent crack closure over a time period that would matter.

In summary although the existence of a steady-state flow of shale over a 100 million year time span is, and will remain, unproven by direct observations, it must logically be expected—based on analysis of various geological observations, on analogies with many other materials, and on micromechanical considerations. It would be farfetched to assume the preexisting natural cracks and joints in shale to have, at 3 km depth, a width sufficient to enhance the large-scale gas permeability appreciably.
7.11. Minimum Pore or Crack Width at Nano-Scale

It should be pointed out, though, that there exists a certain maximum nanoscale opening width of natural cracks that can be sustained even over tens of millions of years. This is demonstrated by the fact that the pores and channels filled by kerogen and gas have a finite width, distributed between 0.5 nm and about 20 nm, with 7 nm as the average.

Why do they not close?–Because the closure is resisted by development of high pressure in the entrapped kerogen and gas, equal to the tectonic stress (to realize the magnitudes, note that, according to the ideal gas equation, the pressure in a spherical gas-filled pore of diameter 100 nm increases from the atmospheric pressure of 0.1 MPa to 40 MPa if the diameter is reduced to 13.6 nm).

The gas-filled nanopores or nanochannels, representing 5%–20% of volume in various types of gas shale, are the source of the local permeability of shale. This permeability suffices to explain the observed gas output when the hydraulic cracks are considered to be roughly 0.1 m apart [58]. But such nano-channels can make virtually no contribution to the gas output when the hydraulic cracks are assumed to be 10 m apart.

7.12. Sealing of Preexisting Natural Cracks by Calcite

Independently of the foregoing creep analysis, one other phenomenon, which alone suffices to render Hypothesis I dubious, is the tight filling of open cracks by calcite [126, 125, 120]. When the drilled cores are brought to the surface, the natural cracks or joints might seem not be filled by calcite tightly. But, under the confining pressures in deep shale strata, they probably are, and thus cannot increase the large-scale gas permeability.
In geologic history, the filling by calcite must have been completed before the cracks, freshly created by some tectonic event, could have been closed by creep. It follows that the time needed for the tight filling of natural cracks by calcite is negligible compared to the average age of natural cracks.

Note: Before closing, we may digress to granite. Aside from instability of parallel crack systems, the lack of sufficient porosity and pore connectivity now appears to be another reason why a branched hydraulic crack system could not be produced in hot granite, as a means to harness geothermal energy for generating electricity [48, 49, 50]. But it should work if the hot rock is sufficiently porous.

7.13. Conclusions

(1) Hypothesis I, which posits that preexisting natural cracks and joints boost the large-scale permeability of shale by about 4 orders of magnitude compared to the permeability measured on deep drilled cores, is not the only way to explain the observed gas production history from wells in which the spacing of perforation clusters is about 10 m.

(2) Recently, an alternative explanation, Hypothesis II, emerged. It is based on the recent development of a three-phase medium theory for hydraulic fracturing. This theory, which takes into account the body forces due to pressure gradients of pore water diffusing (or leaking) from the primary hydraulic cracks into the shale, predicts the development of a branched system of vertical hydraulic cracks with the spacing of about 0.1 m. With such a crack spacing, the observed gas production rate can be explained without postulating any huge large-scale permeability enhancement.
(3) Hypothesis I is predicated on assuming the preexisting natural cracks or joints to be empty and have an opening width of at least 2.8 μm. A much bigger width would be necessary if these cracks or joints were perfectly saturated by water.

(4) Opening widths > 2.8 nm are highly improbable because, over the span of 100 million years or more, the shale most likely exhibits secondary creep or flow. This flow would have had to close the cracks tightly (except the cracks less than about 100 nm wide which, however, cannot have an appreciable effect on the rate of gas production from primary hydraulic cracks spaced about 10 m apart).

(5) Because of primary and secondary creep, a flow channel in shale, with a cross section approximated as an ellipse, must get closed in much less than within mere 210,000 years for the expected Maxwell time and within 9,390,000 years for the extreme estimate of Maxwell time. For a flat crack propped by rectangular pillar walls the corresponding closure times are 5,500 years 256,000 years, respectively (due to simplifications of analysis, all these times are mathematically upper bounds, in view of the assumptions made).

(6) The existence of secondary creep in shale or other rocks over geologic time span appears necessary if one realizes that every increment of creep deformation must not only deactivate some creep sites but also activate new ones. Denying it would be tantamount to a denial of activation of new creep sites.

(7) It appears that the time of transition to secondary creep, or steady-state flow, can be roughly estimated as the Maxwell time. Its value may be inferred from the viscosity indicated by geologic models for the history of the upper layer of Earth lithosphere.
(8) If a fresh natural crack created by a tectonic upheaval is getting filled by calcite, the time to tight filling must be much shorter than the time to close the cracks by creep.

(9) Preexisting or frac-induced?—Most likely frac-induced.

**Closing comment:** Resolving the present questions is necessary to enable a rational control of the frac process.
CHAPTER 8

Branching of Hydraulic Cracks and Their Effect on Permeability of Reservoir

In the last chapter, it was shown that due to creep and calcification, natural fractures cannot remain open. Therefore, these closed natural fractures cannot explain high apparent permeability that is observed in unconventional oil and gas reservoirs. Therefore, the question now is, what caused this large discrepancy between apparent viscosity of reservoir and intact shale?. In this chapter, it is shown, although natural fractures and weak layers in shale are closed, they are significantly weaker than adjacent intact shale. This weakness means they have higher Biot coefficient, lower strength, and higher permeability. It is shown that these properties of weak layers can initiate the branching of hydraulic fractures and thus produce complex cracking of reservoir. Having this complex cracking can explain the high permeability of oil reservoirs.

8.1. Introduction

Significant advances have been made in fracture mechanics of propagation of a single hydraulic crack in elastic rock under tectonic stress [1, 63, 6, 7, 86, 110, 5, 262, 275, 108, 107]. They include characterization of the stress singularity at the tip of a water-filled advancing crack, flow of water of controlled viscosity along the crack, with or without proppant grains, and water leak-off into the shale. Interactions of parallel cracks, their stability, closing, and stress-shadow effect, have also been clarified [47, 58, 85, 84].
Discrete element models used in most commercial softwares, in which the hydraulic crack was simulated by a band of inter-element separations \cite{235, 257}, led to similar results. These studies, however, predicted no branching of the hydraulic cracks, originally spaced at cca 10 m. This presented a dilemma since branching is the only way to reduce the crack spacing to about 0.1 m, which is necessary to explain the gas production rate. Consequently, it has been universally hypothesized that the preexisting natural cracks, spaced at cca 0.1 m, would somehow increase the overall permeability of the shale mass. A 10,000-fold increase of permeability would be necessary to match the gas production rate. But recent analysis \cite{93, 35} showed that the natural, tectonically produced, cracks, which are on the average about 100 million years old, must have been closed by secondary creep (or viscous flow) of shale under tectonic stress within 10,000 to 1 million years (if not filled earlier by calcite deposit). This invalidated the hypothesis.

It might be objected that water in the cracks could have prevented crack closing. But the open spaces in shear cracks, created (due to shear dilatancy) by a tectonic event, could not have been filled by water immediately. If the water had to seep in from the ground surface, it would take about 10 million years, if from a nearby water-filled rock formation, certainly over a million years. This must have left plenty of time for the creep closing to proceed uninhibited.

A recent paper \cite{92} presented a new model which, by contrast with all the previous studies, took into account: 1) the seepage forces (i.e., the body forces due gradients of pore pressure in Darcy diffusion of water into porous shale), and 2) the variation of effective Biot coefficient for the water pressure on the crack plane, caused by gradually vanishing bridges between the opposite faces of a widening bridged crack (another difference from
Figure 8.1. Schematic branching due to natural fractures; a) water is injected at high pressure through damaged zones and weak layers, b) crack branching initiates due to the presence of damaged zones and natural fractures, and c) dense cracking happens in all directions due to the presence of damaged zones, weak layers at closed natural fractures (downward view normal to bedding plane)

the previous studies was to abandon the assumption of incompressibility of water in the cracks, since water is about 20-times more compressible than shale). This model [92] did predict extensive lateral crack branching.

Later analysis, however, showed that the branching indicated by the computer program in [92] was, in fact, triggered by the unintended coding of a sudden change of Biot coefficient for transverse water pressure on the crack. This change abruptly increased the water pressure on the solid phase and triggered dynamic response. Such a sudden trigger
is probably unrealistic, which represents a vital correction to the preceding study [92] (this correction nevertheless reveals a useful fact, namely that producing shocks in fluid pressure could greatly enhance crack branching).

If the change in Biot coefficient is made gradual, the model from the previous study [92] would predict no crack branching, although the branching must occur to explain the observed gas production rate. This study will show that if the previous model [92] is enhanced by introducing, into the shale mass, significant heterogeneity due to damaged weak layers along preexisting natural cracks, then an extensive and dense crack branching is predicted.

It may be noted that the fracking companies are aware of the necessity of branched cracks running along preexisting natural fractures. Fig. 8.1 shows a picture similar to what is found on the websites of some companies. However, this awareness seems to be merely intuitive and empirical. The existing commercial softwares, as well as fracture mechanics studies, predict no branching. So an intersecting system of open natural fractures is assumed to either exist a priori or to develop according to some empirical criteria with no basis in mechanics, supported by some recent experiments indicating the possibility of branching [267, 73, 205, 274]. A physics-based model for branching, which is our goal, seems lacking.

sectionFluid flow in porous solid, without or with cracking damage

Two types of flow play a role in hydraulic fracturing: 1) The flow along the hydraulically created cracks, typically a few millimeters wide, and 2) the flow through nano-scale pores and micro- or nano-cracks in shale with preexisting damage. The latter is negligible after continuous hydraulic cracks form, but here it is found to be crucially important for
crack initiation and branching. The volume flow, \( q \), of water through the pores and nano- or micro-cracks of isotropic material may be approximately calculated from the Darcy law: 

\[
q = -\frac{K}{\mu} \nabla \psi
\]

where \( K \) = permeability, \( \mu \) = dynamic viscosity and \( \psi \) = phase potential calculated as, \( \psi = p - \gamma g z \) Here \( p \) = pore pressure, \( \gamma g \) = pressure gradient due to gravity, and \( z \) = depth from a datum. However, the permeability \( K_v \) in the direction normal to the bedding planes \((x, y)\), i.e., in vertical direction \( z \) is much lower than permeability \( K_h \) along these planes (horizontal). Therefore, the three-dimensional Darcy law is, in general, anisotropic. In Cartesian coordinates \((x, y, z)\), the resulting volume flux vector \( \mathbf{q} = (q_x, q_y, q_z) \) may be written as

\[
(8.1) \quad \mathbf{q} = -\mu^{-1} \mathbf{K} \cdot \nabla \psi
\]

where \( \nabla \) is the vector of gradient operator; and \( \mathbf{K} \) is the \( 3 \times 3 \) permeability matrix, which is diagonal if (and only if) the cartesian axes \( x, y, z \) are chosen to be parallel and normal to the bedding planes.

Although the natural (or preexisting) cracks in shale strata at 3 km depth must have been closed by hundred million years of creep, the damage bands along these cracks, which always accompany propagation of fracture process zone, certainly remain (in fact, based on the known surface energy of shale, it can be shown that even empty pores and cracks cca < 15 nm in size, at depth 3 km, cannot close, and this is confirmed by the known size of pores containing shale gas). Permeability \( K_{xx} \) along these bands is surely much higher than it is in the intact shale (but pores <15 nm contribute nothing globally).

To prevent the formation of horizontal cracks, the pumping pressure is assumed not to exceed the overburden pressure, which is about 75 MPa. The hydraulic fracturing
is considered to produce a system of mutually orthogonal vertical cracks, normal to the
directions of the minimum and maximum principal tectonic stresses. The flow of the sec-
ond type, along the hydraulically created cracks, may be assumed to follow the Reynolds
equations of the classical lubrication theory, which are based on the Poiseuille law for
viscous flow. Thus the horizontal and vertical flow vector components in \( x, y, z \) directions
along with the cracks may be calculated as

\[
Q_x = -\frac{h_y^2}{12\mu} \nabla_x p, \quad Q_y = -\frac{h_x^2}{12\mu} \nabla_y p, \quad Q_z = -\frac{h_x^2 + h_y^2}{12\mu} \nabla_z p
\]

where \( \nabla_x = \partial/\partial_x, \ldots \); \( h_x, h_y \) = opening widths of vertical cracks normal to axes \( x \) and \( y \)
that are positioned into the bedding plane.

An effective way to simulate the hydraulic cracks numerically is the crack band model
\[46, 89, 51\], in which cracking deformation is considered measured over the band (or
element) width. The widths of cracks normal to \( x \) and \( y \) are:

\[
h_x = l_x \epsilon''_{xx}, \quad h_y = l_y \epsilon''_{yy}
\]

(Fig 8.2) where \( \epsilon''_{xx}, \epsilon''_{yy} \) = damage parts of normal strains due to smeared cracking
normal to \( x \) and \( y \) directions; \( l_x, l_y \) = crack band widths, assumed equal to the minimum
possible spacing of adjacent parallel hydraulic cracks (\( l_x, l_y \) must be treated as a material
property, related to fracture energy \( G_f \) of shale; here \( l_x, l_y \) are not changed but if they
were the postpeak would have to be adjusted to preserve \( G_f \)). Furthermore,

\[
\epsilon''_{ij} = \epsilon_{ij} - \epsilon_{ij}^{el}, \quad \epsilon_{ij}^{el} = C_{ijkl} \sigma_{kl}
\]
where $C_{ijkl}$ = transversely isotropic elastic compliance tensor of shale (for unloading); $\sigma_{ij}, \epsilon_{ij}$ are the stress and strain tensors in the rock, calculated from a constitutive model for smeared cracking damage (with a localization limiter \(^{[46]}\)), for which the spherocylindrical microplane constitutive model \(^{[178]}\) has been used. The coordinates are Cartesian, $x_i, i = 1, 2, 3$ ($x_1 \equiv x, x_2 \equiv y, x_3 \equiv z$). Note that, the same as in \(^{[92]}\), water is considered as compressible. It is, in fact, about 20-times more compressible than concrete, and the water pressure during fracking can be high (up to 70 MPa).
8.2. Equilibrium in Two-Phase Solid and Biot Coefficient

The shale may be modeled as a two-phase medium with water-saturated pores, for which the classical Biot-type relations for the equilibrium of the phases apply. For undamaged shale, they read:

\[ S_{ij} = \sigma_{ij} - \delta_{ij} b_0 p \]

where \( p \) = pore pressure, \( b_0 \) = Biot coefficient of undamaged shale; \( S_{ij} \) = total stress tensor; \( \sigma_{ij} \) = stress tensor in the solid phase, and \( \delta_{ij} \) = Kronecker delta. As a special case, \( S_V = \sigma_V - b_0 p \) where \( S_V = S_{kk}/3 \) = volumetric total stress, and \( \sigma_{ij} = \sigma_{kk}/3 \) = volumetric stress in the solid phase.

While typically \( \varphi = 0.1 \), the Biot coefficient of shales can vary between 0.2 and 0.7. Test results \([147, 239, 99, 271, 238, 55]\) show that it increases with the cracking damage and depends on the load direction. This requires generalizing the Biot coefficient as a tensor, \( b_{ij} \) \([207]\). The following, tensorially consistent, empirical relation, which appears to match test data, is proposed:

\[ b_{ij} = \min \left\{ b_0 + \beta \epsilon''_{ij} (\epsilon''_{kk}/3)^{-2/3}, \ 1 \right\} \ (\varphi \leq b_0 \leq 1) \]

Here \( b_0 \) refers to undamaged material, \( \beta \) = empirical parameter, \( \epsilon''_{ij} \) is the inelastic damage strain tensor, and \( \varphi \) = natural porosity of shale. For the Biot coefficient in the direction of unit vector \( \nu_i \), this equation gives \( b_\nu = \nu_i\nu_j b_{ij} = b_0 + \beta \epsilon''_{\nu} (\epsilon''_V)^{-2/3} \) (but \( \leq 1 \)), where \( \epsilon''_V = \epsilon''_{kk}/3 \) = inelastic relative volume expansion, and \( \epsilon''_{\nu} = \nu_i\nu_j \epsilon''_{ij} \) = inelastic normal strain component in direction of vector \( \nu_i \).
For the special case of micro- or nano-cracking normal to $x_1$ direction only, one has $\epsilon''_V = \epsilon''_{11}/3$ and $b_\nu = b_0 + \beta(9\epsilon''_{11})^{1/3}$ (but $\leq 1$). This equation can be interpreted graphically as seen in Fig. 8.2, which shows section A-A of a band of preexisting, mostly aligned, microcracks and the compressive stresses applied by the pore fluid onto the microcrack faces, resisted by tensile stresses in the ligaments of the solid between the microcrack tips.

The viscous drag of water flowing through a soil imposes a seepage force on the soil in the direction of flow. The seepage forces are body forces defined as

\[(8.7) \quad f_s = b \nabla p \]

They are applied on the porous solid and must be balanced by stresses in the solid. Seepage in an upward direction reduces the effective stress within the soil. When the water pressure at a point in the soil is equal to the total vertical stress at that point, the effective stress is zero and the soil has no frictional resistance to deformation \[259, 146\]. They have long been considered in geotechnical engineering to assess the risk of sand liquefaction in cofferdams \[17, 18\] or under dams. However (except for \[92\]) they have been ignored in previous studies of hydraulic fracturing, although they do play a crucial role in crack branching. A poromechanical finite element (FE) code for a two-phase solid automatically takes the seepage forces into account in the form of nodal forces.

8.3. Two-Phase Finite Element (FE) Simulations for a single Damage Band

To clarify the role of nano- or micro-cracking, consider first a horizontal two-dimensional (2D) square block of shale of dimensions 1.1 m $\times$ 1.1 m, supported at the sides by springs
approximately equivalent to an infinite medium, as shown in Fig. 8.3a. Water is injected at the center of south side at the constant rate of 2 m$^3$/s. The anisotropic spherocylindrical microplane model, with the default parameters of shale given in [178], is used as the constitutive model; and $l_x = l_y = 2.1$ m. The initial Biot coefficient is $b_0 = 0.4$. The tectonic stresses are $T_x = -30$ MPa, $T_y = -30$ MPa.

Consider that there is a single preexisting band of nano- or micro-cracks predominantly aligned with axis $y$, represented by the two red elements in Fig. 8.3a (which is what remains after a crack was closed by up to a million years of secondary creep, or viscous flow). These cracks cause the vertical permeability in these two elements to increase cca 1000-times compared to undamaged shale, while the Biot coefficient increases up to 1 and the initial strength decreases to 10% of intact shale.

Figs. 8.3b,c show how damage and pressure propagate after water injection. For this case, the crack band with high water pressure is seen to propagate straight forward, without branching. Now look at stress variation. Fig. 8.3d shows the stress evolution within in the solid part of the first element above the initial damaged elements. Obviously, the damage during post-peak softening is captured in a stable manner. Finally, consider how the Biot coefficient and permeability vary in one cracked element (the first above the initial damaged elements). Fig. 8.3e contrasts the evolution of Biot coefficient in the transverse direction with its constancy in the forward direction, which agrees with experimental observations.
8.4. Do the Seepage Forces Suffice to Induce Crack Branching?

It is well known in classical fracture mechanics that pressurizing a crack cannot produce tension along the crack faces, and thus cannot initiate lateral crack branching (branching is possible only at the tip of a crack propagating at nearly the Raleigh wave speed). In a preceding study [92], it was surmised, under various simplifications, that the seepage forces (Eq. 8.7) would suffice for produce tension along the crack face and thus initiate lateral crack branching. Let us examine it more rigorously. Consider again a horizontal 2D square domain 2.5 m × 2.5 m, containing one line crack (Fig. 8.4a). By virtue of symmetry, only a half-domain is simulated (Fig. 8.4b). The water pressure in the line crack is gradually ramped up to reach the maximum of 50 MPa. Water diffusion
Figure 8.4. a) Pressurized line crack in a 2D domain of a two-phase porous solid (shale) supported by springs at boundaries, subjected to tectonic stresses $T_x$ and $T_y$; b) FE mesh for one half of the domain; c) Extension of the band of high strain; d) Evolution of stress $\sigma_{xx}$ in solid part in the element at the center of initial crack face.

From the pressurized crack into the shale is simulated via Darcy law. First we neglect the increase of Biot coefficient due to damage ($\beta = 0$). Fig. 8.4 shows that the damage, as well as the crack, propagates only in the direct extension of the initial line crack, i.e, there is no branching. Fig. 8.4 shows the evolution of stress in the solid part, $\sigma_{xx}$, along the crack face. The results show that the Biot coefficient can have a major effect and cannot be ignored.
Lateral crack branching would happen if the stress in the solid phase became positive (tensile) and attained the tensile strength of shale. The results show that this cannot happen, regardless of the tectonic stress value (even if vanishing). Nevertheless, the seepage forces reduce the magnitude of compressive stress along the crack face significantly.

We must thus conclude (as an update of [93]) that the seepage forces alone do not suffice to explain and model lateral branching of hydraulic cracks. So, what other phenomena could explain the lateral branching? Not surprisingly, the explanation is the natural (preexisting) fractures even though they must have been completely closed due to millions of years of secondary creep, or flow. We demonstrate it next.

8.5. Hydraulic Crack Branching in Two-Phase Porous Solid with Closed Natural Fractures

In Fig. 8.5a, we now consider the same 2D domain of two-phase porous solid as before, except that now there are two natural weak layers (or preexisting damage bands) in both $x$ and $y$ directions. The crack is uniformly pressurized and water diffuses out. The transverse Biot coefficient within the weak layers that represent the closed natural fractures is $b_{nat} = 1$ because the weak layer (or natural fracture) may consist of separate original crack faces in contact (uncemented by limestone deposit), while in the intact shale the $b_{ij}$-values increase according to Eq. (8.6) from the initial value $b_0 = 0.4$ (Fig. 8.5d) or 0.2 (Fig. 8.5d).

Fig. 8.5b reveals that the hydraulic crack tends to propagate simultaneously along the initial crack and along the weak layer. This confirms that branching can occur if transverse weak layers exist. Further, consider the normal stress parallel to the crack in
one element of the weak layer. If this stress attains the tensile strength, a lateral crack branch can initiate and shale branching can happen. Fig. 8.5b shows the spreading of high and lower transverse strains along both weak layers for the case of Biot coefficient $b = 0.4$, with permeability $K_{\text{weak}}$ along the weak band 5-times bigger than $K_0$ for intact shale.

The computed effect of ratio $K_{\text{weak}}/K_0$ on the $\sigma_{xx}$ evolution in the first element of the weak layer above initial crack is plotted in Fig. 8.5c,d for the initial Biot coefficients, $b_0 = 0.4$ and 0.2. As water diffuses into the shale, the stress in the weak layer increases from negative to tensile values until it finally reaches the tensile strength of the weak layer. Evidently, a greater difference in Biot coefficient between the weak layer and the shale facilitates, and speeds up the crack branching.

Finally, to clarify the effect of the transverse tensile strength of the weak layer, three relative strength $S_{\text{rel}}$ values are considered in Fig. 8.5e (here $S_{\text{rel}}$ is the damaged-to-intact strength ratio of shale). As seen, a smaller $S_{\text{rel}}$ leads to smaller stress, but generally, the effect of $S_{\text{rel}}$ is almost negligible. Hence, whether or not the natural cracks are cemented by limestone is almost irrelevant.

It is instructive to see the evolution of the seepage force vectors acting on the mesh nodes, as portrayed in Fig. 8.5. Fig. 8.5f shows schematically the seepage forces acting on an ellipse around the crack. Fig. 8.5g-i illustrates the evolution of seepage forces. Their orientations make it intuitively clear that they must produce in the porous solid a biaxial tension.

From all these observations, it transpires that a major stimulus for crack branching is the difference in the Biot coefficient and in the permeability between the weak layers and
Figure 8.5. a) Schematic line crack with weak layer and spring boundaries; b) Strain and damage evolution; c) stress in solid part of one element in weak layer considering a shale with $b_0 = 0.4$ and different permeabilities for the weak layer; d) stress in solid part of one element in weak layer considering $b_0 = 0.2$ and different permeabilities for the weak layer; e) stress in solid part of one element in weak layer considering $b_0 = 0.4$ and different relative strength values for weak layer; f) schematic of seepage forces; and g-i) evolution of seepage forces
the intact shale, as well as the shale mass heterogeneity due to the alternation of weak layers and intact porous solid.

It is worth mentioning that the expansion of solid due to the effect of Biot coefficient has been thought to prevent any tension parallel to the crack face, and thus cause the closing of any lateral crack. The preceding results show that this skeptical view does not extend to a heterogeneous shale mass containing weak layers alternating with intact shale.

To demonstrate the present theory on a larger scale, consider a bigger horizontal section of shale, a square domain $5 \text{ m} \times 5 \text{ m}$, containing a uniform orthogonal system of closed natural fractures with aligned preexisting weak layers (Fig. 8.7). To be more realistic, unequal tectonic stresses are considered in $x$ and $y$ directions; $T_x = 30\text{MPa}$ and $T_y = 40\text{MPa}$.

Water is injected at three points at the bottom of the figure. Figs. 8.7c–8.7h show the evolution of water pressure. Water flow and damage strain are seen to follow the path of weak layers. Extensive branching occurs. Obviously, this branching can create closely spaced hydraulic cracks and thus increase the overall permeability of shale stratum by orders of magnitude, compared to non-branching cracks in intact shale.

It has also been checked that omitting the natural fractures leads to no branching. This is evident from the pressure propagation pattern in Fig. 8.8. This figure also documents the localization instability of parallel crack system (aka the stress shadow effect), which causes that the crack emanating from the middle injection point cannot grow long (the long simultaneous growth of both remaining cracks is made possible by the proximity of the boundaries).
Figure 8.6. FE simulation of hydraulic crack branching in a small domain of shale with several orthogonal weak layers; (a) Ring of elastic elements providing elastic support of the boundaries; b) FE mesh, preexisting natural weak layers, and fracking water inlet; c-f) evolution of pressure in a shale with weak layers.
8.6. Conclusions

(1) The natural fractures have a major effect on hydraulic fracturing and are crucial for its success (although they are currently neglected by the commercial fracking softwares).

(2) Even though the natural fractures must have been closed by millions of years of creep, or sealed by mineral deposits, a weak layer of nano- and micro-cracks along these fractures must be expected to facilitate water diffusion.

(3) Poromechanics with Biot coefficient depending on the damage of the solid phase must be used in the analysis of fracking.

(4) Increase of the Biot coefficient in the transverse direction, caused by oriented cracking damage inflicted by fracking, is essential to achieve crack branching.

(5) The typical spacing between natural fractures is roughly 0.1 m. This value matches the spacing of hydraulic cracks that is necessary to explain the typical gas production rate at the wellhead.

(6) The widespread opinion that preexisting natural fractures somehow explain why the overall permeability of shale mass, inferred from the gas production rate, appears to be about 10,000 times higher than what is measured on shale cores in the laboratory, has been basically correct. But these fractures are completely closed, do not convey any gas and their role is indirect.

(7) a) No porosity ⇒ no branching. b) No seepage forces ⇒ no branching. c) No weak layers ⇒ no branching. d) Constant Biot coefficient ⇒ no branching. (Note: consequently, branching in granite is impossible)
Natural fractures

Figure 8.7. 2D FE simulation of fracking process in a horizontal domain with a larger system of natural fractures or weak layers (the red zone shows the propagation of high water pressure)
Figure 8.8. Water pressure propagation for the case with no weak layers, all else the same.
References


APPENDIX A

Publications

A.1. Peer Reviewed Publications


• Rasoolinejad, M., Rahimi-Aghdam, S., & Bažant, Z. P. (Accepted). Prediction of Autogenous Shrinkage in Concrete from Material Composition or Strength Calibrated by a Large Database, as Update to Model B4. *Materials and Structures*.

### A.2. Conference Papers


Vita

My name is Saeed Rahimiaghdam. I am currently a Ph.D. student in the Civil and Environmental Engineering Department at Northwestern University (NU), where I also earned my M.S. degree. I got my B.S. in the fields of Mechanical Engineering and Civil Engineering (Double Major) with highest honors from Sharif University of Technology in my home country, Iran. I perform research on computational and applied mechanics, with emphasis on modeling the behavior of concrete and shale structures. My work on modeling the long-term behavior of concrete structures is one of the most accurate and reliable approaches to simulate the deformation of concrete from time of casting till centuries after that. In addition, my recent work on modeling hydraulic fracturing process and branching of hydraulic cracks has received widespread attention in the technical community and in the media. In my career I have received several fellowships and awards including Walter P. Murphy Fellowship from Northwestern University, Terminal Year Fellowship from Northwestern Transportation Center and Outstanding Paper 2018 Award from Materials and Structures Journal.