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1 Is long-term autogenous shrinkage a creep phenomenon  
2 induced by capillary effects due to self-desiccation?

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13 **Abstract**

14 Long-term shrinkage and creep of concrete can impact the lifetime of  
15 concrete structures. Basic creep of cementitious materials is now known  
16 to be non-asymptotic and evolve logarithmically with time at large times.  
17 However, the long-term kinetics of autogenous shrinkage is not systematically  
18 analyzed. Here we first aim at finding out how autogenous shrinkage evolves  
19 with time at long term. We analyze all experimental data available in the  
20 literature and find that autogenous shrinkage evolves logarithmically with  
21 respect to time at long term, like basic creep. Then, by considering concrete  
22 as a multiscale material, we obtain the bulk creep modulus of the calcium  
23 silicate hydrate gel. In the end, we show that the kinetics of long-term  
24 autogenous shrinkage can be a viscoelastic response to self-desiccation by  
25 comparing the mechanical stress that should be applied to explain this long-

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26 term kinetics of autogenous shrinkage with the capillary force due to self-  
27 desiccation.

28 *Keywords:* Concrete (E), Creep (C), Shrinkage (C), Humidity (A),  
29 Self-desiccation

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## 30 **1. Introduction**

31 Time-dependent behavior (i.e., creep and shrinkage) of cementitious ma-  
32 terials has been studied for more than half a century. In most shrinkage-creep  
33 models [1, 2, 3] and engineering design codes [4, 5, 6], the time-dependent  
34 strain is decomposed into four components: autogenous shrinkage, basic  
35 creep, drying shrinkage and drying creep. In this present work, we focus  
36 only on autogenous shrinkage and basic creep, i.e., on time-dependent defor-  
37 mations of a sealed sample, for which drying is not involved.

38 Both autogenous shrinkage and basic creep are time-dependent strains  
39 that are measured on specimens that do not exchange water with their sur-  
40 roundings. Such condition is achieved either by sealing the sample (e.g., [7]),  
41 or by controlling the relative humidity of the environment to the same rela-  
42 tive humidity as that of the sample (e.g., [8]). For characterization of time-  
43 dependent behavior of cementitious materials under such condition, usually  
44 two specimens are needed: one reference specimen which is not loaded me-  
45 chanically and another specimen which is loaded mechanically. The time-  
46 dependent strain of the reference specimen is called autogenous shrinkage.  
47 Basic creep is obtained by subtracting the time-dependent strain of the refer-

48 ence specimen from the time-dependent strain of the loaded specimen. Basic  
49 creep is the time-dependent strain only due to the mechanical load.

50 For compressive stresses below 40% of the compressive strength, the basic  
51 creep of concrete is non-asymptotic and evolves logarithmically with time at  
52 large times [9, 10, 11, 12]. By analyzing the viscoelastic Poisson's ratio, Aili  
53 et al. [13] showed that even the volumetric basic creep of concrete is non-  
54 asymptotic. Microindentations (e.g., [14] and [15]) and nanoindentations  
55 [16] showed that the basic creep of cement paste and of C-S-H gel evolves  
56 logarithmically with time after a transient period.

57 In contrast, autogenous shrinkage is sometimes assumed to be asymptotic  
58 [4, 6, 3], while some experimental data (e.g. [7, 17, 18, 19]) show that au-  
59 togenous shrinkage evolves logarithmically over time in the long term. For  
60 what concerns its physical origin, many consider that autogenous shrinkage  
61 is caused by the capillary depression due to self-desiccation [20, 21, 22, 23, 24,  
62 25, 26, 27, 28]. However, recently, Ulm and Pellenq [29], Abuhaikal [30] and  
63 Abuhaikal et al. [31] attributed the autogenous shrinkage to eigenstresses  
64 that are developed in the solid skeleton during hydration. Both mechanisms  
65 are likely to play a role, but maybe in a magnitude that depends on the age  
66 of the material: the effect of eigenstresses is indeed expected to be signifi-  
67 cant in particular at early ages, when hydration evolves significantly over  
68 time. For what concerns the modeling of autogenous shrinkage, several au-  
69 thors [21, 23, 24, 27] considered autogenous shrinkage as the elastic strain  
70 under the action of capillary forces induced by self-desiccation, while others

71 [20, 22, 25, 26, 28, 32, 33] suggested that autogenous shrinkage is the vis-  
72 coelastic response of cement-based materials to the capillary forces. However,  
73 modeling autogenous shrinkage as an elastic response to self-desiccation can  
74 capture only asymptotic evolution of autogenous shrinkage at long term as  
75 self-desiccation stops at a certain time. In this work, we aim at shedding  
76 some light on this physical origin, by starting from an exhaustive analysis of  
77 data from the literature.

78 In the first part, we perform an exhaustive analysis of autogenous shrink-  
79 age and basic creep data from the literature. Then, by considering concrete  
80 as a multi-scale material, we use micromechanics to identify a long-term  
81 creep property of the calcium silicate hydrates (C-S-H) gel. In the third  
82 part, we discuss if the kinetics of long-term autogenous shrinkage can be ex-  
83 plained as a creep phenomenon under the action of capillary forces caused  
84 by self-desiccation. To do so, we compare the magnitude of in-pore stresses  
85 necessary to retrieve the measured long-term creep kinetics of autogenous  
86 shrinkage with that of capillary forces.

## 87 **2. Analysis of autogenous shrinkage and basic creep data**

88 This section is devoted to analyze experimental data from the literature  
89 and study the long-term evolution of autogenous shrinkage and basic creep.

### 90 *2.1. Autogenous shrinkage*

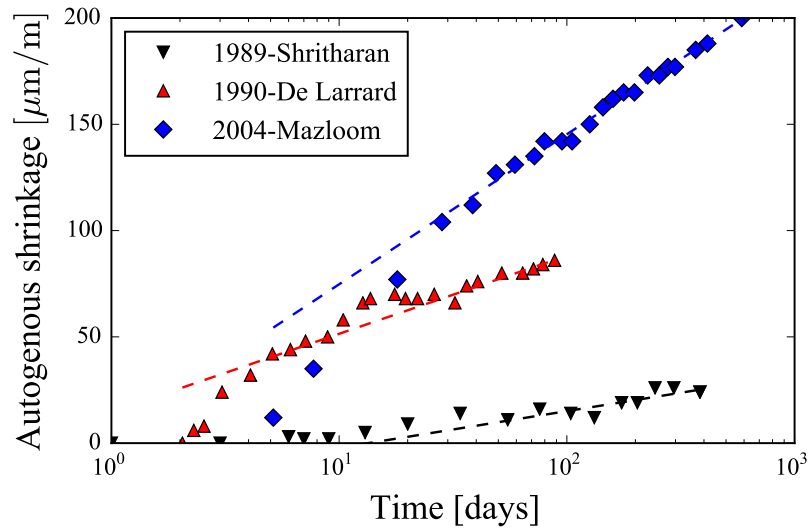
91 We selected autogenous shrinkage data from the comprehensive database  
92 on concrete creep and shrinkage [34] compiled by Prof. Bažant and his col-

93 laborators. The selection procedure is the following:

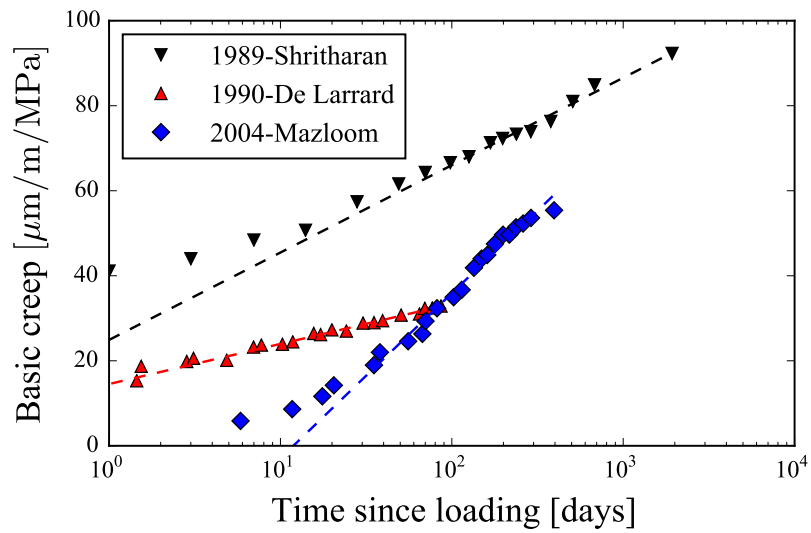
- 94 1. From the database of Northwestern University, we extracted the tests  
95 on concretes and cement pastes made from ordinary Portland cement  
96 (i.e., type CEM I according to Eurocode 2, or type I to type V according  
97 to ASTM standards) without silica fume, fly ash, filler, or slag.
- 98 2. From the extracted tests, we selected only the tests in which the mea-  
99 surement lasted at least until the age of 90 days.
- 100 3. We selected, from the above tests, only the tests performed at a tem-  
101 perature between 20°C and 30°C.

102 In the end, we selected 45 tests in total from the database, among which 29  
103 were performed on concrete and 16 on cement paste.

104 Figure 1a shows the evolution of autogenous shrinkage over time for three  
105 representative tests. At large times, autogenous shrinkage evolves linearly  
106 with the logarithm of time. Such logarithmic evolution was in fact observed  
107 for all concretes and cement pastes prepared at a water-to-cement mass ratio  
108 below 0.5. Among the above 45 tests, mass loss of the sample over the  
109 experiment was measured only in [17] and amounted to about 0.03% at the  
110 age of 240 days. For all other tests, mass was not measured over time, such  
111 that the effectiveness of the sealing during the measurement of autogenous  
112 shrinkage cannot be assessed and we cannot rule out some loss of water  
113 during the test. Hence, part of the measured autogenous shrinkage might  
114 be attributed to drying. However, an uncontrolled loss of water over time



(a)



(b)

Figure 1: (a) Example of autogenous shrinkage data; (b) Example of basic creep data. Data from [35, 7, 17]

115 could not lead to a consistent logarithmic evolution of autogenous shrinkage  
116 for all the tests extracted from the database, and we therefore attribute  
117 this evolution to autogenous shrinkage. As the effectiveness of sealing is an  
118 important feature of autogenous shrinkage or basic creep measurements, a  
119 good practice would be to measure the mass of the sample before and after  
120 testing. Monitoring of the mass loss of sample could be improved, especially  
121 in autogenous shrinkage test, by measuring the mass of sample during the  
122 whole test.

123 For all selected tests we fitted the following empirical relation:

$$\varepsilon_{sh}(t) = \alpha_{sh} \log\left(\frac{t}{\tau_0}\right) + \beta_{sh}, \quad (1)$$

124 to all data points obtained at the age of more than 28 days. In Eq. 1,  $\varepsilon_{sh}$   
125 is the autogenous shrinkage strain,  $\alpha_{sh}$  is the slope of autogenous shrinkage  
126 displayed versus  $\log(t)$ ,  $t$  is the age of concrete and  $\tau_0 = 1$  day is a reference  
127 time. Table 1 summarizes the origin of the data, the mix design properties  
128 of the tested concretes, and the fitted parameter  $\alpha_{sh}$ . For the sake of clarity,  
129 only the data related to the three tests displayed in Fig. 1a are given in  
130 Tab. 2. For the data related to all 45 tests used in this study, see Tabs. A.4  
131 and A.5 in Appendix A.

132 If the kinetics of long-term autogenous shrinkage can be explained as a  
133 creep phenomenon under the action of capillary forces that would induce a  
134 representative stress  $\Sigma_h$ , this representative stress should be related to the



Author	File <sup>1</sup>	w/c <sup>2</sup>	a/c <sup>3</sup>	c <sup>4</sup>	$\alpha_{sh}$ <sup>6</sup>
Shritharan (1989)	e_079_06	0.47	5.09	393	7.51
De Larrard (1990)	A_022_05	0.35	3.96	450	15.96
Mazloom (2004)	A_031_02	0.35	3.70	500	30.64

Table 1: Extract of autogenous shrinkage data. <sup>1</sup>File corresponds to the file number in the database compiled by Prof. Bažant and his collaborators [34]; <sup>2</sup>w/c: water-to-cement ratio; <sup>3</sup>a/c: aggregate-to-cement mass ratio; <sup>4</sup>c: cement per volume of mixture [kg/m<sup>3</sup>]; <sup>5</sup> $\alpha_{sh}$ : Fitted parameter in Eq. 1 [ $\mu\text{m}/\text{m}$ ].

135 fitted parameter  $\alpha_{sh}$  through:

$$\Sigma_h = 3\alpha_{sh}C_c^K, \quad (2)$$

136 where  $C_c^K$  is the bulk creep modulus of the tested concrete or cement paste,  
137 which is defined, in a creep test under the stress  $\sigma_0$ , as the asymptotic value of  
138  $\sigma_0/(t d\varepsilon/dt)$  in the long term [36]. Hence, in order to back-calculate the stress  
139  $\Sigma_h$  that would be necessary to explain the long-term kinetics of autogenous  
140 shrinkage as a creep phenomenon, we first need the bulk creep modulus  $C_c^K$   
141 of each concrete or cement paste. In the next section, we analyze basic creep  
142 data to obtain this parameter. On all the samples used for the 45 tests  
143 of autogenous shrinkage analyzed, only 5 measurements of basic creep were  
144 performed. To back-calculate creep properties of C-S-H, instead of limiting  
145 ourselves to the analysis of those 5 tests only, in the next section we will  
146 analyze a larger number of basic creep tests extracted from the database of  
147 Northwestern University.

148 *2.2. Basic creep*

149 We selected basic creep data also from the comprehensive database on  
150 concrete creep and shrinkage [34] compiled by Prof. Bažant and his collabo-  
151 rators. We selected all basic creep data that satisfy the following criteria:

- 152 1. From the database of Northwestern University, we extracted the tests  
153 on concretes and cement pastes made from ordinary Portland cement  
154 (i.e., type CEM I according to Eurocode 2, or type I to type V according  
155 to ASTM) without silica fume, fly ash, filler, or slag.
- 156 2. From the extracted tests, we selected only the tests for which the mea-  
157 surement after 5 times the age at loading lasted for at least a decade  
158 in logarithmic scale.
- 159 3. We selected, from the above tests, only the tests performed at a tem-  
160 perature between 20°C and 30°C.
- 161 4. We selected, from the above tests, only the tests in which the applied  
162 stress did not exceed 40% of the compressive strength.

163 With these criteria, we selected in total 59 tests from the database. All  
164 those 59 tests were in fact performed on concrete samples.

165 Figure 1b displays the evolution of basic creep over time for three rep-  
166 resentative tests. At large times, the evolution of basic creep is logarithmic  
167 with time for all selected tests. Hence, for all selected tests, on the part of  
168 the data going from the time equal to 5 times the age of loading till the end  
169 of the test, we fitted the following empirical relation:

Author	File <sup>1</sup>	w/c <sup>2</sup>	a/c <sup>3</sup>	c <sup>4</sup>	t <sub>0</sub> <sup>5</sup>	1/C <sub>c</sub> <sup>E6</sup>
Shritharan (1989)	c_079_08	0.47	5.09	390	14	8.93
De Larrard (1990)	D_022_05	0.35	3.96	450	3	4.10
Mazloom (2004)	D_031_02	0.35	3.70	500	7	16.86

Table 2: Extract of basic creep data. <sup>1</sup>File corresponds to the file number in the database compiled by Prof. Bažant and his collaborators [34]; <sup>2</sup>w/c: water-to-cement ratio; <sup>3</sup>a/c: aggregate-to-cement mass ratio; <sup>4</sup>c: cement per volume of mixture [kg/m<sup>3</sup>]; <sup>5</sup>t<sub>0</sub>: loading age [days]; <sup>6</sup>1/C<sub>c</sub><sup>E</sup>: Fitted parameter in Eq. 3 [ $\mu\text{m}/\text{m}/\text{MPa}$ ].

$$\varepsilon_{cr}(t) = \frac{1}{C_c^E} \log\left(\frac{t}{\tau_0}\right) + \beta_{cr}, \quad (3)$$

170 where  $\varepsilon_{cr}$  is the specific basic creep strain,  $1/C_c^E$  is the slope of basic creep  
171 displayed versus  $\log(t)$ ,  $t$  is time since loading and  $\tau_0 = 1$  day is a reference  
172 time. The parameter  $C_c^E$  is called the uniaxial creep modulus of concrete.  
173 Table 2 summarizes the origin of the data, the mix design properties of the  
174 tested concretes, the age at loading, and the fitted parameter  $1/C_c^E$ . For the  
175 sake of clarity, only the data related to the three experiments displayed in  
176 Fig. 1b are given in Tab. 2. For the data related to all 59 tests used in this  
177 study, see Tabs. B.6 and B.7 in Appendix B.

178 In conclusion, we confirmed that basic creep evolves logarithmically with  
179 respect to time at long term. Our analysis of an exhaustive set of data shows  
180 that autogenous shrinkage also evolves logarithmically with respect to time  
181 at long term.

182 **3. Downscaling of creep compliance from the scale of concrete to**  
183 **the scale of the C-S-H gel**

184 The objective of this section is to estimate the long-term creep proper-  
185 ties of the C-S-H gel from the basic creep data on concrete presented in  
186 section 2.2. As the creep of concrete evolves logarithmically with respect  
187 to time in the long term, we can express the bulk creep compliance  $J_c^K$  of  
188 concrete as  $J_c^K = 1/K_c^0 + 1/C_c^K \log(1 + t/\tau_c)$ , where  $C_c^K$  is the bulk creep  
189 modulus that characterizes the-long term kinetics of bulk creep.

190 Aili et al. [13] showed that the viscoelastic Poisson's ratio  $\nu_c$  of concrete  
191 remains quite constant and comprised between 0.15 and 0.2 for mature con-  
192 cretes. Hence, we consider the viscoelastic Poisson's ratio  $\nu_c$  of concrete to  
193 be constant and equal to 0.2. Then, following the same procedure as in Ap-  
194 pendix B of [16], we obtain the relation between the bulk creep modulus  $C_c^K$   
195 and the uniaxial creep modulus  $C_c^E$ :

$$C_c^K = \frac{C_c^E}{3(1 - 2\nu_c)}. \quad (4)$$

196 The objective is now to relate this bulk creep modulus  $C_c^K$  of concrete to  
197 the bulk creep modulus  $C_{gel}^K$  of the C-S-H gel.

198 We first present the multi-scale scheme of concrete that we are going to  
199 use in this article. Then, we derive some theoretical results by adapting elas-  
200 tic homogenization schemes to the viscoelastic case via the correspondence  
201 principle [37]. In the end, we apply the derived equations to relate the bulk

202 creep modulus  $C_c^K$  of concrete to the bulk creep modulus  $C_{gel}^K$  of the C-S-H  
203 gel.

### 204 3.1. Multiscale model for concrete

205 Concrete can be regarded as a multiscale composite material at three  
206 different scales, which are displayed in Fig. 2:

- 207 • At the largest scale of concrete (see Fig. 2a), the aggregates are consid-  
208 ered as spherical inclusions that do not creep and are embedded into a  
209 matrix made of cement paste, which creeps.
- 210 • At a scale below, i.e., at the scale of the cement paste (see Fig. 2b),  
211 portlandite, aluminate phases (i.e., ettringite AFt and mono-sulfo-  
212 aluminate AFm phases) and the unhydrated clinker are considered as  
213 spherical inclusions that do not creep and are embedded into a ma-  
214 trix made of a mixture of C-S-H with capillary pores. This mixture is  
215 considered to creep.
- 216 • At another scale below (see Fig. 2c), the mixture of C-S-H with cap-  
217 illary pores is considered to be a matrix of C-S-H gel (which contains  
218 the gel porosity) which surrounds spherical capillary pores.

219 As explained before, according to the findings of Aili et al. [13], here we  
220 consider the viscoelastic Poisson's ratio of concrete (Fig. 2a) equal to 0.2. As  
221 shown by Aili et al. (Eqs. 14 and 15 in [13]), this assumption implies that for  
222 the multiscale model here considered, we can also consider the viscoelastic

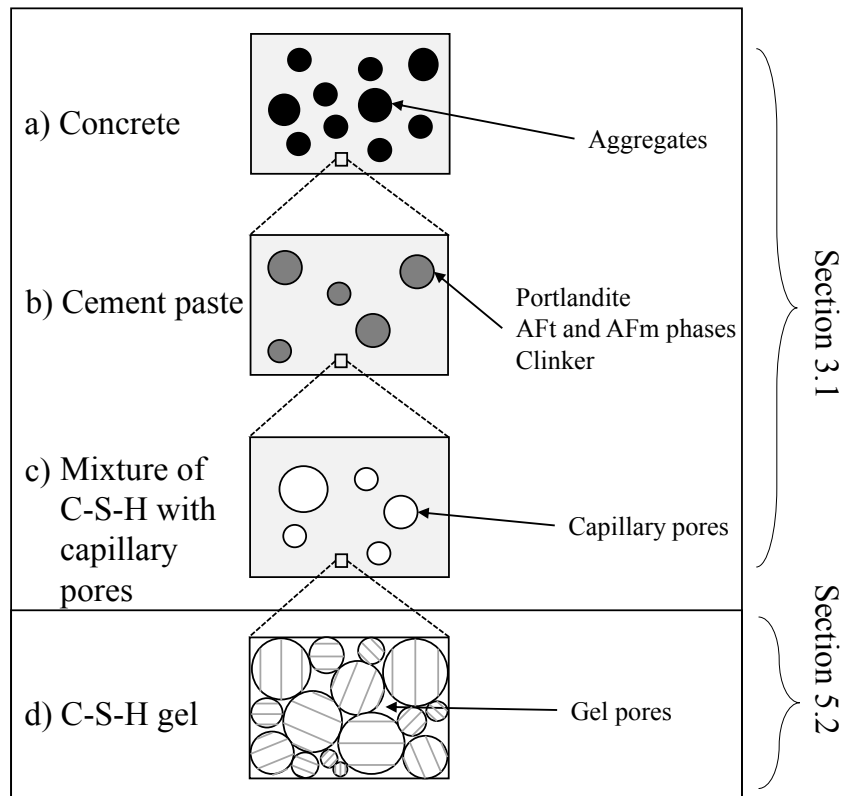


Figure 2: Multiscale structure of concrete: (a) Concrete as a matrix of cement paste embedding aggregates, (b) cement paste as portlandite, AFt and AFm phases and unhydrated clinker embedded into a matrix made of a mixture of C-S-H with capillary pores, (c) mixture of C-S-H with capillary pores as a matrix of C-S-H gel surrounding capillary porosity, and (d) C-S-H gel as a mixture of C-S-H particles and gel pores. The scales (a) (b) (c) are considered in Sec. 3.1 for the downscaling of the creep modulus, while the scale (d) is considered in Sec. 5.2 for estimating the Biot coefficient of the mixture of C-S-H gel with capillary pores.

223 Poisson's ratio of cement paste (Fig. 2b), of the mixture of C-S-H gel and  
 224 capillary pores (Fig. 2c), and of the C-S-H gel as constant and equal to 0.2.

225 *3.2. Theoretical derivation*

226 We consider a composite made of a matrix that embeds spherical inclu-  
 227 sions. Given the microstructure, we employ the Mori-Tanaka scheme [38, 39]  
 228 to calculate the properties of the composite as a function of the properties of  
 229 each phase (i.e., of the matrix and the inclusions). Note that the Mori-Tanaka  
 230 scheme is valid even at high volume fractions of inclusions [40, 41, 42, 43, 44].  
 231 The interface between inclusions and matrix is considered to be perfectly ad-  
 232 hesive. The viscoelastic Poisson's ratio  $\nu_m$  of the matrix is constant and equal  
 233 to 0.2. Applying the correspondence principle [37] to the elastic Mori-Tanaka  
 234 homogenization scheme, making use of the fact that  $\nu_m = 0.2$ , replacing the  
 235 elastic parameters by the  $s$ -multiplied Laplace transform of their correspond-  
 236 ing viscoelastic operator, we obtain the viscoelastic homogenization scheme  
 237 in the Laplace domain:

$$\widehat{K}_{com} = \frac{\widehat{K}_m(1 - f_i) + \widehat{K}_i(1 + f_i)}{\widehat{K}_m(1 + f_i) + \widehat{K}_i(1 - f_i)} \widehat{K}_m, \quad (5)$$

238 where  $f_i$  is the volume fraction of inclusions;  $K_m$ ,  $K_i$  and  $K_{com}$  are the  
 239 viscoelastic bulk relaxation modulus of the matrix, of the inclusion and of  
 240 the composite, respectively;  $\widehat{g}$  is the the Laplace transform of the function  $g$ .

241 These bulk relaxation moduli  $K_j$  are related to bulk creep compliances  
 242  $J_j^K$  through:

$${}_s\widehat{J}_j^K = \frac{1}{{}_s\widehat{K}_j}, \quad (6)$$

243 where  $s$  is the Laplace variable,  $j = m, i, com$  represents matrix, inclusion  
 244 and composite, respectively.

245 We suppose that the bulk creep compliances  $J_j^K$  evolve logarithmically  
 246 with respect to time at large times and can be expressed as  $J_j^K = (1/K_j^0) +$   
 247  $(1/C_j^K) \log(1 + t/\tau_j)$ , where  $j = m, i, com$  and  $C_j^K$  is the bulk creep modu-  
 248 lus. By using the final value theorem [45] and the Laplace transform of a  
 249 derivative, we obtain the following relation between the bulk creep modulus  
 250  $C_j^K$  and the Laplace transform  $\widehat{J}_j^K$  of the bulk creep compliance:

$$\begin{aligned} \frac{1}{C_j^K} &= \lim_{t \rightarrow \infty} t \dot{J}_j^K = \lim_{s \rightarrow 0} s \left( \widehat{t \dot{J}_j^K} \right) = \lim_{s \rightarrow 0} \left( -s \frac{d}{ds} \widehat{(J_j^K)} \right) \\ &= \lim_{s \rightarrow 0} \left( -s \frac{d}{ds} \left( s \widehat{(J_j^K)} - J_j^K|_{t=0} \right) \right) = \lim_{s \rightarrow 0} \left( -s \frac{d}{ds} \left( s \widehat{(J_j^K)} \right) \right), \end{aligned} \quad (7)$$

251 where  $\dot{g}$  is the derivative of the function  $g$  with respect to time. Equation 7  
 252 means that  $d(s \widehat{J}_j^K)/ds$  can be approximated by  $-1/s C_j^K$  for small  $s$ , from  
 253 which follows that  $\widehat{J}_j^K$  can be approximated by  $-\log(s)/C_j^K s$  for small  $s$ :

$$\widehat{J}_j^K \approx -\log(s)/C_j^K s, \text{ for } s \rightarrow 0. \quad (8)$$

254 Using the final value theorem [45], letting  $s \rightarrow 0$  in Eq. 5 and combining  
 255 the result with Eqs. 6 and 8, we obtain:

$$C_{com}^K = \frac{C_m^K(1 - f_i) + C_i^K(1 + f_i)}{C_m^K(1 + f_i) + C_i^K(1 - f_i)} C_m^K, \quad (9)$$



256 which makes it possible to relate the bulk creep modulus  $C_{com}^K$  of the com-  
 257 posite to that of its constituents.

258 We consider the following two cases:

- 259 • Case 1: Composite made of a matrix embedding non-creeping inclu-  
 260 sions. The matrix is considered to creep logarithmically with respect to  
 261 time in the long term. The long-term volumetric creep kinetics of the  
 262 matrix is characterized by its creep modulus  $C_m^K$ . Letting  $C_m^K/C_i^K \rightarrow 0$ ,  
 263 Eq. 9 yields:

$$C_{com}^K = \frac{1 + f_i}{1 - f_i} C_m^K; \quad (10)$$

- 264 • Case 2: Porous composite made of a matrix embedding spherical pores.  
 265 Letting  $C_i^K/C_m^K \rightarrow 0$ , Eq. 9 yields:

$$C_{com}^K = \frac{1 - f_i}{1 + f_i} C_m^K; \quad (11)$$

### 266 3.3. From concrete to C-S-H gel

267 In this section, we derive a relation between the bulk creep modulus  $C_c^K$  of  
 268 concrete and  $C_{gel}^K$  of the C-S-H gel, by performing three steps of downscaling  
 269 following the multi-scale scheme displayed in Fig. 2.

- 270 • To relate the bulk creep modulus  $C_c^K$  of concrete to the bulk creep  
 271 modulus  $C_p^K$  of cement paste (Fig. 2a), we apply Eq. 10.

- 272 • To relate the bulk creep modulus  $C_p^K$  of cement paste to the bulk creep  
 273 modulus  $C_{mix}^K$  of the mixture of C-S-H gel with capillary pores (Fig. 2b),  
 274 we apply again Eq. 10.
- 275 • To relate the bulk creep modulus  $C_{mix}^K$  of the mixture of C-S-H gel  
 276 with capillary pores to the bulk creep modulus  $C_{gel}^K$  of the C-S-H gel  
 277 (Fig. 2c), we apply Eq. 11.

278 With these three steps of downscaling, we obtain:

$$C_c^K = \left( \frac{1 + f_a}{1 - f_a} \right) \left( \frac{1 + f_b}{1 - f_b} \right) \left( \frac{1 - \phi_c}{1 + \phi_c} \right) C_{gel}^K, \quad (12)$$

279 where  $f_a$  is the volume fraction of aggregates (counted with respect to the  
 280 volume of concrete);  $f_b$  is the volume fraction of portlandite, AFt and AFm  
 281 phases and unhydrated clinker (counted with respect to the volume of cement  
 282 paste);  $\phi_c$  is the volume fraction of the capillary porosity (counted with  
 283 respect to the volume of the mixture of C-S-H gel with capillary pores).

284 Combining Eqs. 4 and 12, we obtain:

$$C_{gel}^K = \left( \frac{1 - f_a}{1 + f_a} \right) \left( \frac{1 - f_b}{1 + f_b} \right) \left( \frac{1 + \phi_c}{1 - \phi_c} \right) \frac{1}{3(1 - 2\nu_c)} C_c^E. \quad (13)$$

285 This equation makes it possible to compute the bulk creep modulus  $C_{gel}^K$  of  
 286 the C-S-H gel from the uniaxial creep modulus  $C_c^E$  obtained from the analysis  
 287 of basic creep data, as long as the microstructural parameters  $f_a$ ,  $f_b$ , and  $\phi_c$   
 288 are known.

289 To determine the microstructural parameters  $f_a$ ,  $f_b$ , and  $\phi_c$ , we use the  
290 following:

- 291 • The volume fraction  $f_a$  of aggregates in concrete is computed from the  
292 mix design properties of concrete:  $f_a = 1 - c/\rho_c - c \times w/c/\rho_w$ , where  $c$   
293 and  $w/c$  are the mass of clinker per volume of mixture and the water-  
294 to-cement mass ratio, respectively, and where  $\rho_c = 3.15 \text{ g/cm}^3$  and  
295  $\rho_w = 1 \text{ g/cm}^3$  are the density of cement and of water, respectively.
- 296 • The volume fraction  $f_b$  of portlandite, AFt and AFm phases and un-  
297 hydrated clinker (counted with respect to the volume of the cement  
298 paste) is the sum of the volume fractions of portlandite, of AFt and  
299 AFm phases, and of unhydrated clinker. Each of them is computed  
300 by using Powers' model [46, 47], which considers that the volume of  
301 cement paste is composed of bulk hydrates (i.e., solid hydrates plus  
302 gel pores), unhydrated clinker, and capillary pores. As Powers et al.  
303 [46] stated, complete hydration almost never occurs in practice. There-  
304 fore, instead of the final hydration degree given by Powers' model, here  
305 we consider the long-term hydration degree  $\xi^\infty$  of the sample to be  
306 equal to  $\xi^\infty = 1 - \exp(-3.3w/c)$  [48]. The volume fraction of bulk  
307 hydrates per unit volume of cement paste is  $2.12(1 - p)\xi^\infty$ , where  
308  $p = (w/c)/((w/c) + (\rho_w/\rho_c))$ . The volume of portlandite per unit  
309 volume of bulk hydrates is estimated to be equal to 25%, which is a  
310 typical value for CEM I cement pastes [49]. Hence, the volume fraction

311 of portlandite (counted with respect to the volume of cement paste) is  
 312  $0.53(1-p)\xi^\infty$ . The volume fraction of AFt and AFm phases per unit  
 313 volume of bulk hydrates is estimated to be equal to 15% [49], from  
 314 which the volume fraction of AFt and AFm phases (counted with re-  
 315 spect to the volume of cement paste) is  $0.32(1-p)\xi^\infty$ . The volume frac-  
 316 tion of unhydrated clinker (still counted with respect to the volume of  
 317 cement paste) is estimated also with Powers' model [46, 47] to be equal  
 318 to  $(1-p)(1-\xi^\infty)$ . Therefore, the volume fraction  $f_b$  of portlandite,  
 319 AFt and AFm phases and unhydrated clinker (counted with respect to  
 320 the volume of the cement paste) is  $f_b = 0.85(1-p)\xi^\infty + (1-p)(1-\xi^\infty)$ .

- 321 • The volume fraction  $\phi_c$  of capillary porosity with respect to the volume  
 322 of the mixture of C-S-H gel with capillary pores is also computed by  
 323 using Powers' model [46, 47]. The volume fraction of capillary pores  
 324 counted with respect to the volume of cement paste is estimated as  
 325  $p - 1.12(1-p)\xi^\infty$ . The volume fraction of the mixture of C-S-H gel with  
 326 capillary pores (counted with respect to the volume of cement paste) is  
 327 equal to  $1 - f_b$ . Hence, the capillary porosity  $\phi_c$  (i.e., volume fraction of  
 328 the capillary pores counted with respect to the volume of the mixture of  
 329 C-S-H gel with capillary pores) is equal to  $(p - 1.12(1-p)\xi^\infty)/(1 - f_b)$ .

330 Inserting the uniaxial creep modulus  $C_c^E$  from Tab. 2 and the above-  
 331 calculated microstructural parameters  $f_a$ ,  $f_b$  and  $\phi_c$  into Eq. 12, we obtain  
 332 the bulk creep modulus  $C_{gel}^K$  of the C-S-H gel. The results are displayed

333 in Fig. 3. The bulk creep modulus  $C_{gel}^K$  of C-S-H gel does not exhibit any  
334 specific trend with water-to-cement ratio. Its mean value is 13 GPa and its  
335 standard deviation is 6.7 GPa. The quite large standard deviation may partly  
336 be due to the fact that creep moduli are fitted on measurements that last for  
337 extended periods of time and hence are difficult to perform, and also to the  
338 fact that creep moduli characterize a rate and are therefore quite sensitive  
339 to experimental noise.

340 In Fig. 3, we also display creep moduli of C-S-H gel obtained by back-  
341 calculation of microindentation creep data on cement pastes obtained by  
342 Zhang [50] and by Frech-Baronet et al. [15] at various relative humidities.  
343 Zhang [50] found that the contact creep modulus of the C-S-H gel is con-  
344 stant for relative humidities greater than 75%. Consequently, to be consis-  
345 tent with the internal relative humidities observed in autogenous conditions,  
346 out of the various microindentation creep experiments performed by Zhang  
347 and by Frech-Baronet et al., we only analyzed those performed at a relative  
348 humidity larger than 75%, which resulted in the analysis of 3 of Zhang's  
349 tests, and of 1 of Frech-Baronet's tests. Following the same procedure as  
350 described above, those results were downscaled to yield the bulk creep mod-  
351 ulus of the C-S-H gel, which is displayed in Fig. 3. While creep moduli  
352 back-calculated from Frech-Baronet's results lie slightly above the range of  
353 creep moduli back-calculated from uniaxial creep experiments on concrete,  
354 creep moduli back-calculated from Zhang's results lie in that range. This  
355 agreement is consistent with the fact that indentation of cement paste has

356 been shown to be a reliable tool to estimate the long-term creep kinetics of  
 357 concrete [14], through a comparison of microindentation creep experiments  
 358 with uniaxial basic creep experiments on concrete. In [14], Zhang et al. tested  
 359 cement pastes that had the same mix design as those found in concrete sam-  
 360 ples manufactured and tested by Le Roy [10]. The excellent agreement that  
 361 Zhang et al. obtained in [14] by comparing their data with Le Roy's data is  
 362 consistent with the fact that, in Fig. 3, creep moduli of C-S-H back-calculated  
 363 from Zhang's microindentation creep experiments almost overlap with those  
 364 back-calculated from Le Roy's uniaxial creep experiments on concrete.

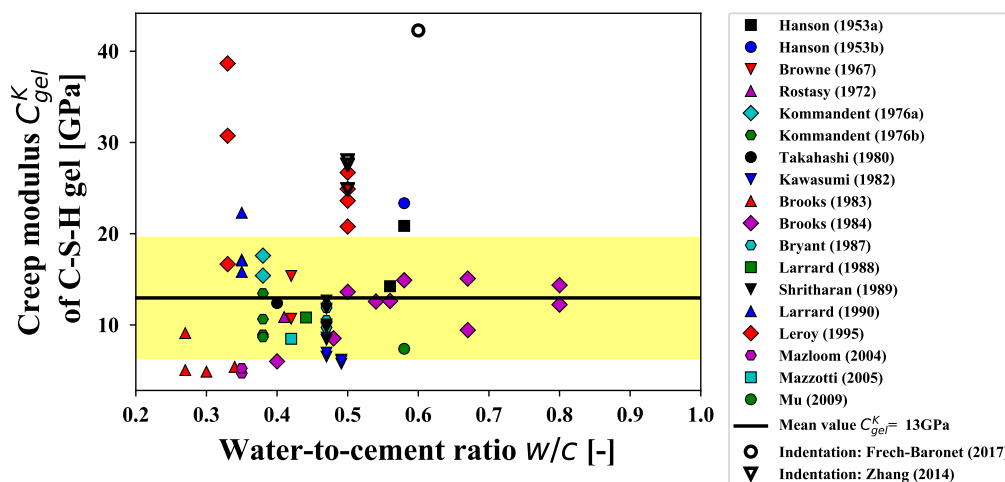


Figure 3: Bulk creep modulus of C-S-H gel as a function water-to-cement ratio, computed from basic creep data on concrete available in [51, 52, 53, 54, 55, 56, 19, 18, 57, 58, 35, 7, 10, 17, 59, 60] and from microindentation creep data on cement paste available in [14, 15]. The mean value of 13 GPa and standard deviation of 6.7 GPa displayed in the figure are calculated by including the basic creep data on concrete only.

365 **4. In-pore stress necessary to explain long-term kinetics of auto-**  
366 **genous shrinkage**

367 In this work we aim at testing the following hypothesis: may capillary  
368 forces due to self-desiccation be the driving force of the long-term kinetics  
369 of autogenous shrinkage? To do so, we compare the in-pore stress necessary  
370 to explain the long-term kinetics of autogenous shrinkage with the capillary  
371 stress induced by self-desiccation of concrete under autogenous conditions.  
372 In this section, we compute the in-pore mechanical stress  $\sigma_h$  that should act  
373 on the mixture of C-S-H gel with capillary pores to explain the long-term  
374 kinetics characterized by the parameter  $\alpha_{sh}$ . The next section is devoted to  
375 compute the capillary stress due to self-desiccation.

376 We compute first the mechanical stress  $\Sigma_h$  that should act on concrete to  
377 explain the long-term kinetics of autogenous shrinkage, which was captured  
378 through the fitted parameter  $\alpha_{sh}$  (see Tab. 1), using Eq. 2. Then, we down-  
379 scale the stress  $\Sigma_h$  to the scale of the C-S-H gel to calculate the stress  $\sigma$  that  
380 should act on the C-S-H gel to explain the long-term kinetics of autogenous  
381 shrinkage of the concrete. To do so, we perform two steps of downscaling.  
382 The two steps are the same as the first two steps of the downscaling scheme  
383 described in section 3.1.

384 At each step, we are dealing with a composite made of a matrix that  
385 creeps with no asymptote and of spherical inclusions that do not creep. The  
386 matrix is subjected to a stress  $\underline{\underline{\sigma}}$ . We aim at computing an equivalent macro-  
387 scopic stress  $\underline{\underline{\Sigma}}$  that should act on the composite to obtain an identical strain

388 response [39, 61, 41].

389 In the elastic case, the macroscopic stress reads [39, 61, 41]:

$$\underline{\underline{\Sigma}} = (1 - f_i)\underline{\underline{\sigma}} : \langle \underline{\underline{A}} \rangle_m, \quad (14)$$

390 where  $f_i$  is the volume fraction of inclusions;  $\underline{\underline{A}}$  is the 4<sup>th</sup>-order strain local-  
 391 ization tensor;  $\langle g \rangle_m$  is the mean value of the parameter  $g$  on the matrix  
 392 domain. For an isotropic stress  $\underline{\underline{\sigma}} = \sigma \underline{\underline{1}}$  where  $\underline{\underline{1}}$  is the identity tensor (hence,  
 393  $\underline{\underline{\Sigma}} = \Sigma \underline{\underline{1}}$ ), Eq. 14 can be simplified to a scalar form by taking the spherical  
 394 part  $A_i^{sph}$  of the localization tensor  $\underline{\underline{A}}$  of the inclusion in the Mori-Tanaka's  
 395 scheme [39]:

$$\Sigma = (1 - f_i)\sigma \left( \frac{1 - f_i A_i^{sph}}{1 - f_i} \right) = \frac{\left(1 + \frac{\alpha_m}{K_m}(K_i - K_m)\right) (1 - f_i)}{1 + \frac{\alpha_m}{K_m}(K_i - K_m)(1 - f_i)}\sigma, \quad (15)$$

396 where  $\alpha_m = 3K_m/(3K_m + 4G_m)$ . For  $\nu_m = 0.2$ , we obtain  $\alpha_m = 1/2$ .

397 In the viscoelastic case with the viscoelastic Poisson's ratio of the matrix  
 398  $\nu_m(t) = 0.2$ , using the elastic-viscoelastic correspondence principle, we re-  
 399 place all elastic parameters in Eq. 15 by the  $s$ -multiplied Laplace transform  
 400 of their corresponding viscoelastic operator. Then, considering that, at long  
 401 term, the inclusion is much stiffer than the matrix, i.e.,  $K_i^\infty \gg K_m^\infty$ , we use  
 402 the final value theorem and obtain:

$$\Sigma^\infty = \sigma^\infty. \quad (16)$$



403 In two steps of downscaling from the scale of concrete to the scale of the  
 404 mixture of C-S-H gel with capillary pores, we use Eq. 16 twice. The in-pore  
 405 stress  $\Sigma_h$  that should act on concrete to explain the long-term kinetics of  
 406 autogenous shrinkage corresponds to an identical stress  $\sigma_h = \Sigma_h$  that should  
 407 act on the mixture of C-S-H gel with capillary pores. Hence, combining  
 408 Eqs. 2 and 12, we can relate this stress  $\sigma_h$  to the fitted parameter  $\alpha_{sh}$  via  
 409 the bulk creep modulus  $C_{gel}^K$  of the C-S-H gel:

$$\sigma_h = 3\alpha_{sh}C_{gel}^K \left( \frac{1+f_a}{1-f_a} \right) \left( \frac{1+f_b}{1-f_b} \right) \left( \frac{1-\phi_c}{1+\phi_c} \right). \quad (17)$$

410 This equation provides the mechanical stress  $\sigma_h$  that must act in the cap-  
 411 illary pore system at the scale of the mixture of C-S-H gel with capillary pores  
 412 to explain the long-term logarithmic kinetics of autogenous shrinkage of the  
 413 concrete or cement paste specimen, characterized by the parameter  $\alpha_{sh}$ . For  
 414 all autogenous shrinkage experiments considered in section 2.1, we compute  
 415 the mechanical stress  $\sigma_h$  from the measured parameter  $\alpha_{sh}$  with Eq. 17 and  
 416 display it as a function of water-to-cement ratio in Fig. 6. This stress  $\sigma_h$  is  
 417 going to be compared with the capillary forces due to self-desiccation in the  
 418 next section.

## 419 5. Capillary stress due to self-desiccation

420 In this section, we first analyze experimental data of evolution of relative  
 421 humidity under autogenous conditions to characterize the self-desiccation.  
 422 Then, making use of Power's hydration model [46] and of the theory of

423 poromechanics [62], we estimate the capillary stress due to self-desiccation.  
424 By comparing this capillary stress with the mechanical stress  $\sigma_h$  calculated  
425 in the previous section, we check the hypothesis that capillary forces due to  
426 self-desiccation are the driving force of the long-term kinetics of autogenous  
427 shrinkage.

### 428 *5.1. Self-desiccation of cementitious materials*

429 Hydration of cement is a water-consuming process. In sealed conditions,  
430 i.e., in absence of any external water supply, consumption of water desatu-  
431 rates the cement paste as the porosity decreases less slowly than the quantity  
432 of water. As a result, the relative humidity inside the cement paste decreases  
433 [63]. Flatt et al. [64, 65] showed that hydration stops below a certain rela-  
434 tive humidity. On the other hand, Jensen [63] showed that self-desiccation  
435 is limited by thermodynamics. Therefore, we expect that, under autogenous  
436 conditions, the relative humidity will reach an equilibrium value when hydra-  
437 tion stops. The objective of this section is to relate this relative humidity at  
438 equilibrium to the water-to-cement ratio of the concrete or the cement paste.

439 Many authors [66, 67, 68, 69, 70, 71, 72, 73, 74, 75] measured relative  
440 humidity inside concrete or cement paste under autogenous conditions as a  
441 function of age. For each of these tests, author, year, water-to-cement ratio  
442 and duration of test are summarized in Tab. 3.

443 As the relative humidity is expected to reach an asymptotic value, we  
444 propose the following simple empirical relation for the evolution of relative

Author	w/c <sup>1</sup> [-]	$\tau_T$ <sup>2</sup> [days]	$h_r^\infty$ <sup>3</sup> [-]	$\tau_{h_r}$ <sup>4</sup> [days]
Baroghel-Bouny (1991)	0.35	800	0.87	237
Baroghel-Bouny (1991)	0.49	365	0.94	52
Jensen (1996)	0.30	1	0.89	0.12
Jensen (1996)	0.35	14	0.93	0.71
Persson (1997)	0.25	450	0.76	40
Persson (1997)	0.33	450	0.82	62
Persson (1997)	0.47	450	0.88	135
Persson (1997)	0.58	450	0.94	98
Kim (1999)	0.28	11	0.87	2.12
Kim (1999)	0.40	11	0.91	2.24
Kim (1999)	0.68	12	0.97	15.24
Yssorche (1999)	0.33	365	0.84	15.24
Yssorche (1999)	0.44	365	0.90	0.95
Yssorche (1999)	0.59	365	0.99	0.57
Yssorche (1999)	0.75	337	0.99	0.06
Jiang (2005)	0.20	300	0.81	8.56
Jiang (2005)	0.30	300	0.87	19.29
Jiang (2005)	0.40	300	0.90	27.76
Jiang (2005)	0.50	300	0.93	41.37
Zhutovsky (2013)	0.21	7	0.81	0.44
Zhutovsky (2013)	0.25	7	0.84	0.61
Zhutovsky (2013)	0.33	7	0.86	0.62
Wyrzykowski (2016)	0.21	7	0.78	4
Wyrzykowski (2016)	0.24	7	0.79	5
Wyrzykowski (2016)	0.30	7	0.83	5
Wyrzykowski (2016)	0.35	7	0.88	4
Aili (2017)	0.52	127	0.90	10

Table 3: Summary of experimental data of evolution of relative humidity with respect to time under autogenous conditions, and of the fitted parameters. Data from [66, 67, 68, 69, 70, 71, 72, 73, 74, 75]. <sup>1</sup>w/c: water-to-cement ratio; <sup>2</sup> $\tau_T$ : duration of the test; <sup>3</sup> $h_r^\infty$ : long-term relative humidity under autogenous conditions, obtained by fitting of Eq. 18; <sup>4</sup> $\tau_{h_r}$ : characteristic time of decrease of relative humidity under autogenous conditions, obtained by fitting of Eq. 18.

445 humidity over time under autogenous conditions:

$$h_r(t) = h_r^\infty + (1 - h_r^\infty) \exp\left(-\frac{t}{\tau_{h_r}}\right), \quad (18)$$

446 where  $h_r^\infty$  and  $\tau_{h_r}$  are fitted parameters which depend on the water-to-cement  
 447 ratio and correspond to the long-term relative humidity and to a character-  
 448 istic time, respectively. For the sake of simplicity, in Fig. 4 we present only  
 449 the experimental measurements performed in [66] and the corresponding fit  
 450 with Eq. 18. However, we analyzed a set of 27 experiments, see Fig. C.8  
 451 in Appendix C.

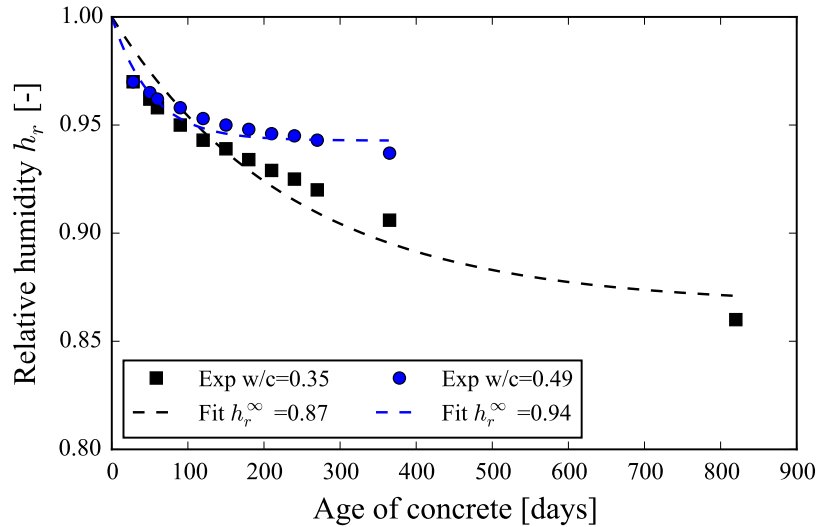


Figure 4: Evolution of relative humidity under autogenous conditions, data retrieved from [66].

452 To assess the importance of the choice of the fitting function in the  
 453 estimation of long-term relative humidity, instead of Eq. 18, we also fit-

454 ted the evolutions of relative humidity over time with a rational function  
 455  $h_r(t) = ((h_r^\infty)^2 t + \tau_{h_r}) / (h_r^\infty t + \tau_{h_r})$  and with a hyperbolic function  $h_r(t) =$   
 456  $1 - (1 - h_r^\infty) \tanh(t/\tau_{h_r})$ , where  $h_r^\infty$  and  $\tau_{h_r}$  are the fitting parameters. For  
 457 the 27 tests considered, with respect to the fitting performed with Eq. 18,  
 458 the fitted long-term relative humidity differed by a maximum of 0.039 when  
 459 using the rational function, and by a maximum of 0.055 when using the hy-  
 460 perbolic function. Consequently, the fitted long-term relative humidity  $h_r^\infty$   
 461 does not seem to depend much on the shape of the function used to fit the  
 462 evolutions of relative humidity over time.

463 Since we are interested in the long-term kinetics of autogenous shrinkage,  
 464 we listed the long-term relative humidities  $h_r^\infty$  in Tab. 3 and plotted them  
 465 against water-to-cement ratio in Fig. 5. From Fig. 5, we can see that the  
 466 long-term relative humidity  $h_r^\infty$  for a concrete with water-to-cement ratio  
 467  $w/c$  will be comprised between values given by the following equations:

$$\text{Upper bound for } h_r^\infty : h_{r,u}^\infty = \begin{cases} 1 - (0.4 - w/c), & \text{if } w/c < 0.4, \\ 1, & \text{otherwise.} \end{cases} \quad (19)$$

$$\text{Lower bound for } h_r^\infty : h_{r,l}^\infty = \begin{cases} 1 - 0.45(0.77 - w/c), & \text{if } w/c < 0.75, \\ 1, & \text{otherwise.} \end{cases} \quad (20)$$

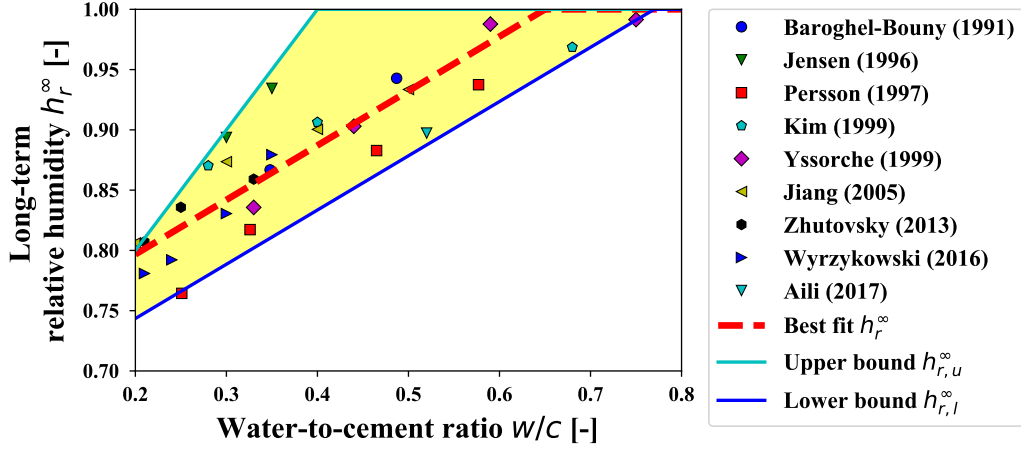


Figure 5: Long-term relative humidity under autogenous conditions as a function water-to-cement ratio, computed from experimental data in [66, 67, 68, 69, 70, 71, 72, 73, 74, 75].

468 *5.2. Estimation of capillary force*

469 Partially saturated poromechanics, under the assumption of pore iso-  
 470 deformation, states that the capillary stress due to capillary pressure is equal  
 471 to  $bS_l P_c$ , where  $P_c$  is the capillary pressure,  $S_l$  is the saturation degree in  
 472 liquid water and  $b$  is the Biot coefficient (see Eq. 9.77 in [62]). In the  
 473 following, we compute those three parameters.

474 The long-term relative humidity  $h_r^\infty$  in autogenous conditions, displayed  
 475 in Fig. 5, can be expressed as [76]:

$$h_r^\infty = h_{r,K}^\infty h_{r,S}^\infty, \quad (21)$$

476 where  $h_{r,K}^\infty$  captures the variations of relative humidity solely due to surface  
 477 tension effects and to the curvature of the fluid/vapour menisci in the pore

478 space, and where  $h_{r,S}^\infty$  captures the variations of relative humidity due to the  
 479 presence of ions in the pore solution. The term  $h_{r,S}^\infty$  can be estimated with  
 480 Raoult's law [62] to be equal to the long-term molar fraction of water in the  
 481 pore solution. The term  $h_{r,K}^\infty$  can be related to capillary pressure through a  
 482 combination of Kelvin's equation with Laplace equation:

$$P_c = -\frac{RT}{V_w} \log(h_{r,K}^\infty), \quad (22)$$

483 where  $R$ ,  $T$  and  $V_w$  are the ideal gas constant, the absolute temperature and  
 484 the molar volume of water, respectively.

485 Combining Eqs. 21 and 22 makes it possible to compute the capillary  
 486 pressure  $P_c$  from the long-term relative humidity  $h_r^\infty$  and the molar fraction  
 487 of water in the pore solution. However, the composition of the pore solution  
 488 has not been much measured for cement pastes under autogenous condition  
 489 in the long term, despite numerous studies [67, 77, 21, 78, 25, 79, 80] devoted  
 490 to the effect of ions in the pore solution of cement pastes. Chen et al. [79]  
 491 measured the concentration of ions in a cement paste with water-to-cement  
 492 mass ratio of 0.4 at the age of 7 days, for which, based on their measured ion  
 493 concentration of 1.6 mol/L, we calculate a molar fraction of water around  
 494 0.97. Hu [80] measured the concentration of ions for cement pastes with  
 495 water-to-cement mass ratios of 0.3, 0.35, 0.39 and 0.46, up to the age of  
 496 28 days, and showed that  $h_{r,S}$  is around 97%. In absence of any long-term  
 497 measurement, we hence assumed that, for all cement pastes considered in

498 this study, the long-term parameter  $h_{r,S}^\infty$  was equal to 0.97. A combined use  
 499 of Eq. 21 and Eq. 22 then makes it possible to estimate the capillary pressure  
 500  $P_c$  from the long-term relative humidity. Considering the upper bound  $h_{r,u}^\infty$  of  
 501 long-term relative humidity leads a lower bound  $P_{c,l}$  of the capillary pressure,  
 502 while considering the lower bound  $h_{r,l}^\infty$  of long-term relative humidity leads  
 503 an upper bound  $P_{c,u}$  of the capillary pressure.

504 The saturation degree  $S_l$  (i.e., the volume fraction of the capillary and  
 505 gel pores spaces that is occupied with liquid water with respect to the total  
 506 volume of capillary and gel pores) is computed, in the same manner as in  
 507 [21], from Power's model as follows: For a given volume  $V$  of cement paste,  
 508 the volume  $V_p$  of total pore space is equal to the total volume minus the  
 509 volume  $0.53(1-p)\xi^\infty V$  of portlandite,  $0.32(1-p)\xi^\infty V$  of AFt and AFm  
 510 phases,  $(1-p)(1-\xi^\infty)V$  of clinker and  $V_{\text{CSH}} = 1.52(1-p)(1-\alpha^\infty)V$  of  
 511 C-S-H solid (i.e., C-S-H without its gel porosity). The volume of chemical  
 512 shrinkage is equal to  $V_{cs} = 0.20(1-p)\xi^\infty V$ . The saturation degree  $S_l$  is then  
 513 obtained as:

$$S_l = 1 - \frac{V_{cs}}{V_p} = \frac{p - 0.72(1-p)\alpha^\infty}{p - 0.52(1-p)\alpha^\infty}. \quad (23)$$

514 The Biot coefficient is computed by two steps of upscaling:

- 515 • In the first step, at the scale of the C-S-H gel, we compute the porosity  
 516 of the C-S-H gel as the mean value of the porosity of high-density C-  
 517 S-H and low-density C-S-H: considering that 40% of C-S-H gel is high-



518 density C-S-H with porosity 0.24, and the other 60% is low-density  
 519 C-S-H with porosity 0.37 [81], the mean porosity of C-S-H gel is esti-  
 520 mated to be equal to  $\phi_{gel} = 0.32$ . Considering that the C-S-H gel is  
 521 composed from spherical C-S-H particles and gel pores, we apply the  
 522 self-consistent homogenization scheme. From micro-poroelasticity, the  
 523 tensor of Biot coefficients for a porous material reads [82]:

$$\underline{\underline{b}}^{hom} = \phi_0 \underline{\underline{1}} : \langle \underline{\underline{A}} \rangle_p = \underline{\underline{1}} : \left( \underline{\underline{I}} - f_s \langle \underline{\underline{A}} \rangle_s \right), \quad (24)$$

524 where subscripts  $p$  and  $s$  indicate the pore space and the solid skeleton,  
 525 respectively, while  $\phi_0$  is the initial porosity. For the C-S-H gel, for  
 526 which the Poisson's ratio is assumed equal to 0.2 (see section 3.1), we  
 527 introduce the strain localization tensor of the self-consistent scheme in  
 528 Eq. 24, which leads the Biot coefficient  $b_{gel}$  of the C-S-H gel:

$$b_{gel} = 2\phi_{gel}. \quad (25)$$

529 In the viscoelastic case, using the elastic-viscoelastic correspondence  
 530 principle, we replace the elastic parameters in Eq. 25 with the  $s$ -  
 531 multiplied Laplace transform of their corresponding viscoelastic opera-  
 532 tor. At long term, we consider that the microstructure of the material  
 533 does not evolve anymore, from which follows that the gel porosity  $\phi_{gel}$   
 534 is constant over time. As a result, Eq. 25 holds true for the viscoelastic

535 case, under the hypothesis that the viscoelastic Poisson's ratio of the  
 536 C-S-H gel is constant and equal to 0.2.

537 • In the second step, at the scale of the mixture of C-S-H gel with cap-  
 538 illary pores (Fig. 2c), we consider the Biot coefficient of a composite  
 539 made from a porous matrix (i.e., C-S-H gel) and capillary pores. As  
 540 was done by Pichler et al. [41] or by Ghabezloo [82], we assume that  
 541 the pores at the various scales are connected and that the pore pressure  
 542 is identical in all pores. For such porous materials with pores at var-  
 543 ious scales, micro-poroelasticity provides the following tensor of Biot  
 544 coefficients [82]:

$$\underline{\underline{b}}^{hom} = \underline{\underline{1}} - f_m \langle \underline{\underline{A}} \rangle_m : \left( \underline{\underline{1}} - \underline{\underline{b}}_m \right), \quad (26)$$

545 where the subscript  $m$  indicates the solid skeleton, which here acts as  
 546 a matrix. For the mixture of the C-S-H gel with capillary pores, in the  
 547 elastic case, assuming again the Poisson's ratio of the C-S-H gel equal  
 548 to 0.2 (see section 3.1), we apply the Mori-Tanaka's homogenization  
 549 scheme by introducing the Mori-Tanaka strain localization tensor into  
 550 Eq. 26, from which we obtain the Biot coefficient  $b$  of the mixture of  
 551 C-S-H gel with capillary pores:

$$b = 1 - \frac{1 - \phi_c}{1 + \phi_c} (1 - b_{gel}). \quad (27)$$

552 In the viscoelastic case, using the elastic-viscoelastic correspondence  
553 principle, we replace the elastic parameters in Eq. 27 with the  $s$ -  
554 multiplied Laplace transform of their corresponding viscoelastic op-  
555 erator. At long term, we consider again that the microstructure of  
556 the material does not evolve anymore, from which follows that Eq. 27  
557 holds true for the viscoelastic case (again, under the hypothesis that  
558 the viscoelastic Poisson's ratio of the C-S-H gel is constant and equal  
559 to 0.2).

560 Knowing the upper bound  $P_{c,u}$  and lower bound  $P_{c,l}$  of capillary pressure,  
561 the saturation degree  $S_l$ , and the Biot coefficient  $b$  of the sample, we estimate  
562 the upper bound  $bS_lP_{c,u}$  and lower bound  $bS_lP_{c,l}$  of the macroscopic mechan-  
563 ical volumetric compressive stress due to the capillary forces acting on the  
564 mixture of capillary pores with C-S-H gel. The upper and lower bounds of  
565 the stress  $bS_lP_c$  are displayed in Fig. 6, from which we can observe that the  
566 stress  $bS_lP_c$  increases with a decreasing water-to-cement ratio and reaches  
567 about 15 to 20 MPa for a water-to-cement ratio equal to 0.2.

568 It is worth mentioning that the two-step procedure here used to upscale  
569 the Biot coefficient assumes that both capillary pores and gel pores are sub-  
570 jected to the same capillary stress  $bS_lP_c$ , which is not verified in practice.  
571 Indeed, water starts to evaporate from gel pores only when the relative hu-  
572 midity drops below 40% [81] while, under autogenous condition, the relative  
573 humidity remains always above 75%, as shown in Fig. 5. Therefore, in long-  
574 term autogenous conditions, the gel pores remain fully saturated. In contrast,

575 capillary pores are partially saturated. As a result, the average pressure in  
 576 gel pores differs from the average pressure in capillary pores. This difference  
 577 of average pressures can be taken into account into the upscaling scheme, for  
 578 instance with a double porosity model making use of two Biot coefficients  
 579 [83, 61, 84, 85]. Coussy and Brisard [86] also developed a micromechanical  
 580 model that can take into account the fact that the pore saturation degree  
 581 varies with pore size. However, for the sake of simplicity and considering  
 582 the relatively low precision of the estimated long-term relative humidities,  
 583 we chose to assume that both gel pores and capillary pores were subjected  
 584 to the same average pressure.

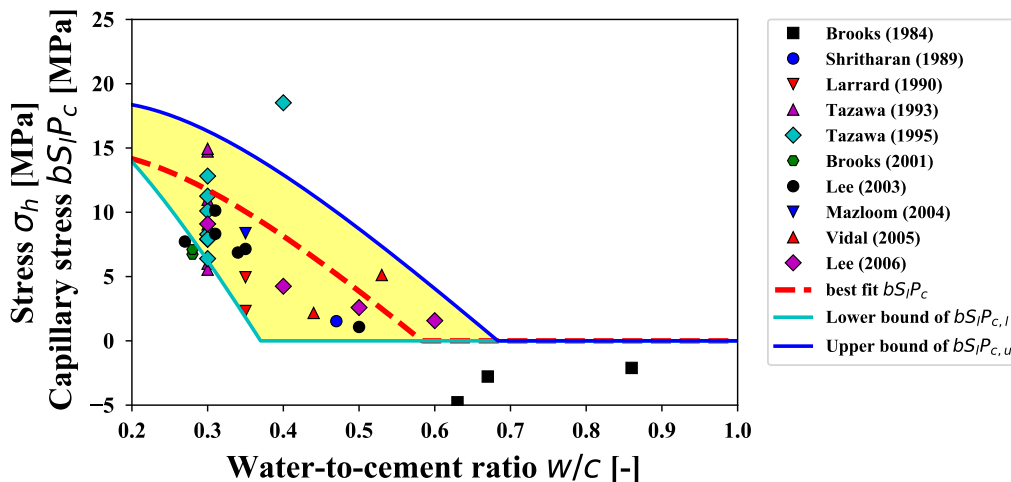


Figure 6: Mechanical stress  $\sigma_h$  that should act on the mixture of C-S-H gel displayed together with capillary pores to explain the long-term kinetics of autogenous shrinkage of data in [19, 35, 7, 87, 88, 89, 90, 91, 92, 17, 93, 94], displayed together with estimated bounds of the capillary stress  $bS_lP_c$ .

585 Figure 6 compares estimated bounds of the stress  $bS_lP_c$  due to the cap-

586 illary forces and acting on the mixture of C-S-H gel and capillary pores  
587 with the mechanical stress  $\sigma_h$  that should act to explain the long-term ki-  
588 netics of autogenous shrinkage characterized by the parameter  $\alpha_{sh}$  fitted on  
589 the measurements. The capillary stress obtained from the best fit on the  
590 long-term relative humidity slightly overestimates the experimentally back-  
591 calculated mechanical stress  $\sigma_h$ , even though those two quantities exhibit  
592 quite similar trends with water-to-cement ratio. However, almost all points  
593 of the mechanical stress  $\sigma_h$  back-calculated from experiments lie between the  
594 model-predicted upper bound and lower bound of the stress  $bS_lP_c$  induced  
595 by capillary forces. Therefore, we conclude that the long-term kinetics of  
596 autogenous shrinkage is compatible with the hypothesis that the evolution  
597 of autogenous shrinkage in the long term is due to creep under the action of  
598 capillary forces due to self-desiccation.

## 599 **6. Discussion on the choice of hydration model**

600 Powers' hydration model [46] was used to quantify the volume fractions  
601 of the different phases in the multiscale microstructure of cement paste dis-  
602 played in Fig. 2. Based on water vapor sorption isotherms, this model pro-  
603 vides the amount of capillary water and physically adsorbed water. Powers'  
604 model considers the porosity of hydrates to be rather constant, which is why  
605 we considered a constant gel porosity  $\phi_{gel}$  while estimating the Biot coeffi-  
606 cient of the C-S-H gel (see section 5.2).

607 Recently, nuclear magnetic resonance (NMR) measurements of Muller et

608 al. [95, 96] showed that the density of the C-S-H gel varies: the gel porosity  
609 depends on water-to-cement ratio and hydration degree. Königsberger et  
610 al. [97] proposed an alternative hydration model that takes into account  
611 this densification effect explicitly. In this section, we check how different the  
612 capillary stresses  $bS_l P_c$  estimated with this alternative hydration model are  
613 from the ones computed with Powers’s model in section 5.2 and displayed in  
614 Fig. 6.

615 In a first step, based on the hydration model of Königsberger et al.[97], we  
616 compute the alternative volume fractions  $f_b$  of portlandite, AFt and AFm  
617 phases and unhydrated clinker and  $\phi_c$  of capillary pores, which intervene  
618 in Eq. 13. Using these alternative volume fractions  $f_b$  and  $\phi_c$ , we perform  
619 again the analysis of basic creep data, following the same procedure as in  
620 section 3.3, from which we obtain a mean bulk creep modulus of C-S-H gel  
621 of 10 GPa with a standard deviation of 6.3 GPa. Then, using again those  
622 alternative volume fractions  $f_b$  and  $\phi_c$ , we perform the analysis of autogenous  
623 shrinkage data following the same procedure as in section 4 and calculate  
624 the mechanical stress  $\sigma_h$  that should act on the mixture of C-S-H gel with  
625 capillary pores to explain the long-term kinetics of autogenous shrinkage as  
626 a creep phenomenon. This stress  $\sigma_h$  is displayed in Fig. 7. For the sake of  
627 consistency, we used the same limits for the axes of this figure as for those  
628 of Fig. 6: note that, for Tazawa’s sample at a water-to-cement mass ratio of  
629 0.4, the calculation yields  $\sigma_h = 30.2$  MPa, which is outside of the figure.

630 In a second step, we compute the Biot coefficient and saturation degree

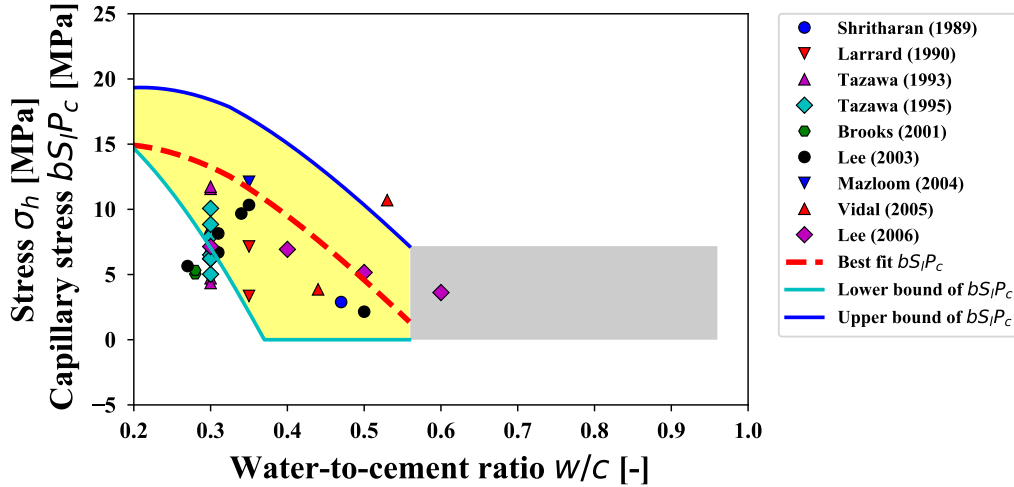


Figure 7: Mechanical stress  $\sigma_h$  that should act on the mixture of C-S-H gel with capillary pores to explain the long-term kinetics of autogenous shrinkage as a creep phenomenon (data from [19, 35, 7, 87, 88, 89, 90, 91, 92, 17, 93, 94]), displayed together with estimated bounds of the capillary stress  $bS_l P_c$ . All volume fractions in the multiscale scheme of concrete are computed based on the hydration model of Königsberger et al [97].

631 based on the alternative volume fractions  $\phi_c$  of capillary pores and  $\phi_{gel}$  of  
632 gel pores obtained from the hydration model of Königsberger et al. The  
633 gel porosity  $\phi_{gel}$  is then a function of the water-to-cement mass ratio. In  
634 Königsberger's hydration model, the gel porosity  $\phi_{gel}$  is higher than 0.5 for  
635 cement pastes with water-to-cement mass ratio higher than 0.55 if the hy-  
636 dration degree is taken to be equal to  $\xi^\infty = 1 - \exp(-3.3w/c)$  [48]. The  
637 applicability of the self-consistent scheme with spherical particles that we  
638 used for homogenizing the C-S-H gel is limited to cases where the volume  
639 fraction of pores is lower than 0.5. Hence, we limited our analysis to water-  
640 to-cement mass ratios lower than 0.55 for the estimation of the capillary

641 stress  $bS_lP_c$ . The alternative values of Biot coefficient and saturation pro-  
642 vide alternative boundaries of the capillary stress  $bS_lP_c$ , which are displayed  
643 in Fig. 7 against the mechanical stress  $\sigma_h$ . We observe that Fig. 7 does not  
644 differ much from Fig. 6. Therefore, using Königsberger’s model than Pow-  
645 ers’ model to calculate volume fraction still allows us to conclude that the  
646 long-term kinetics of autogenous shrinkage is compatible with the hypothesis  
647 that the evolution of autogenous shrinkage in the long term is due to creep  
648 under the action of capillary forces due to self-desiccation.

## 649 7. Conclusions

650 We performed an exhaustive study of experimental data from the litera-  
651 ture on basic creep and autogenous shrinkage. We downscaled these results  
652 with the help of elastic homogenization schemes extended to linear viscoelas-  
653 ticity and discussed the origin of long-term autogenous shrinkage using the  
654 theory of poromechanics. Several conclusions can be drawn:

- 655 • For materials that are kept under autogenous conditions, the creep  
656 modulus of C-S-H gel exhibits no specific trend with water-to-cement  
657 ratio, with a mean value of  $13 \pm 6.7$  GPa. This creep modulus is lower  
658 than the value obtained from microindentation testing [14, 15], which  
659 is 32 GPa.
- 660 • For concretes made with a water-to-cement mass ratio below 0.5, the  
661 autogenous shrinkage is not asymptotic and evolves logarithmically



662 with respect to time in the long term. In contrast, for concretes with a  
663 water-to-cement mass ratio larger than 0.5, under the hypothesis that  
664 autogenous shrinkage is due to creep under the action of capillary forces  
665 due to self-desiccation, autogenous shrinkage is negligible.

- 666 • An upper bound and a lower bound are proposed for the long-term rel-  
667 ative humidity under autogenous conditions by analyzing experimental  
668 measurements of internal relative humidity over time from the litera-  
669 ture.
- 670 • The long-term stress  $bS_tP_c$  induced by the capillary forces due to self-  
671 desiccation (see Fig. 6) increases with a decreasing water-to-cement  
672 ratio and reaches about 15 to 20 MPa for a water-to-cement ratio equal  
673 to 0.2.
- 674 • The long-term kinetics of the logarithmically-evolving autogenous shrink-  
675 age is compatible with the hypothesis that, in the long term, the in-  
676 crease of autogenous shrinkage is due to creep under the action of cap-  
677 illary forces due to self-desiccation.

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682 Biot coefficient.

683 **Appendix A. Autogenous shrinkage database**

684 This section is devoted to present autogenous shrinkage data that are  
685 displayed in Fig. 6. For each data are given author and year of the work, file  
686 number that corresponds database [34] collected in Northwestern University,  
687 mix design properties and long-term log-slope of autogenous shrinkage, see  
688 Tab. A.4 and A.5.

689 **Appendix B. Basic creep database**

690 This section is devoted to present basic creep data that are displayed in  
691 Fig. 3. For each data are given author and year of the work, file number that  
692 corresponds database [34] collected in Northwestern University, mix design  
693 properties, age of loading and long-term log-slope of basic creep, see Tabs. B.6  
694 and B.7.

695 **Appendix C. Experimental data of evolution of relative humidity**  
696 **with respect to time under autogenous conditions**

697 This section is devoted to present the experimental data of evolution of  
698 relative humidity under autogenous conditions. For each data, the fitted  
699 long-term relative humidity  $h_r^\infty$  is displayed in legend of figure, see Fig. C.8

Author	File <sup>1</sup>	w/c <sup>2</sup>	a/c <sup>3</sup>	c <sup>4</sup>	$\alpha_{sh}$ <sup>6</sup>
Brooks (1984)	e_074_20	0.67	4.75	366	-26.3
Brooks (1984)	e_074_29	0.76	4.75	383	-98.94
Brooks (1984)	e_074_30	0.62	4.75	344	-83.22
Brooks (1984)	e_074_33	0.86	4.75	457	-46.73
Brooks (1984)	e_074_35	0.63	4.75	387	-42.43
Shritharan (1989)	e_079_6	0.47	5.09	393	7.51
Larrard (1990)	A_022_2	0.35	3.96	450	82.13
Larrard (1990)	A_022_3	0.35	3.96	450	7.49
Larrard (1990)	A_022_5	0.35	3.96	450	15.96
Tazawa (1993)	A_062_6	0.3	0	533	129.92
Tazawa (1993)	A_062_7	0.3	0	533	221.16
Tazawa (1993)	A_062_8	0.3	0	533	224.33
Tazawa (1993)	A_062_9	0.3	0	533	90.08
Tazawa (1993)	A_062_12	0.3	0	533	83.14
Tazawa (1993)	A_062_13	0.3	0	533	136.05
Tazawa (1993)	A_062_14	0.3	0	533	132.06
Tazawa (1993)	A_062_15	0.3	0	533	164.82
Tazawa (1995)	A_063_22	0.3	0	NAN	1.71
Tazawa (1995)	A_063_27	0.4	0	NAN	1.69
Tazawa (1995)	A_063_39	0.3	0	NAN	4.35
Tazawa (1995)	A_063_42	0.3	0	NAN	2.42
Tazawa (1995)	A_063_44	0.3	0	NAN	0.03
Tazawa (1995)	A_063_49	0.3	0	NAN	9.01
Tazawa (1995)	A_063_50	0.3	0	NAN	8.49
Tazawa (1995)	A_063_51	0.3	0	NAN	8.85

Table A.4: Details of autogenous shrinkage data (first part). <sup>1</sup>File corresponds to the file number in the database compiled by Prof. Bažant and his collaborators [34]; <sup>2</sup>w/c: water-to-cement ratio; <sup>3</sup>a/c: aggregate-to-cement mass ratio; <sup>4</sup>c: cement per volume of mixture [kg/m<sup>3</sup>]; <sup>5</sup> $\alpha_{sh}$ : Fitted parameter in Eq. 1 [ $\mu\text{m}/\text{m}$ ].

Author	File <sup>1</sup>	w/c <sup>2</sup>	a/c <sup>3</sup>	c <sup>4</sup>	$\alpha_{sh}$ <sup>6</sup>
Weiss (1998)	A_068_1	0.3	3.04	485	63.01
Weiss (1998)	A_068_16	0.3	3.04	485	59.57
Weiss (1998)	A_068_19	0.3	3.04	485	61.54
Brooks (2001)	A_007_8	0.28	4	450	14.39
Brooks (2001)	A_007_12	0.28	4	450	15.18
Lee (2003)	A_023_1	0.5	4.66	370	5.59
Lee (2003)	A_023_2	0.35	3.85	450	23.07
Lee (2003)	A_023_3	0.31	3.4	500	24.23
Lee (2003)	A_023_4	0.27	3.05	550	19.58
Lee (2003)	A_023_8	0.34	3.73	440	20.44
Lee (2003)	A_023_9	0.31	3.4	500	29.5
Zhang (2003)	A_072_1	0.26	3.7	496	38.97
Zhang (2003)	A_072_2	0.3	3.6	497	40.17
Mazloom (2004)	A_031_2	0.35	3.7	500	30.64
Vidal (2005)	A_065_3	0.44	3.7	450	10.95
Vidal (2005)	A_065_5	0.53	5.25	350	27.99
Lee (2006)	A_024_1	0.3	2.73	583	29.96
Lee (2006)	A_024_2	0.4	3.92	438	17.17
Lee (2006)	A_024_3	0.5	5.09	350	12.57
Lee (2006)	A_024_4	0.6	6.43	292	9

Table A.5: Details of autogenous shrinkage data (second part). <sup>1</sup>File corresponds to the file number in the database compiled by Prof. Bažant and his collaborators [34]; <sup>2</sup>w/c: water-to-cement ratio; <sup>3</sup>a/c: aggregate-to-cement mass ratio; <sup>4</sup>c: cement per volume of mixture [kg/m<sup>3</sup>]; <sup>5</sup> $\alpha_{sh}$ : Fitted parameter in Eq. 1 [ $\mu\text{m}/\text{m}$ ].

Author	File <sup>1</sup>	w/c <sup>2</sup>	a/c <sup>3</sup>	c <sup>4</sup>	t <sub>0</sub> <sup>5</sup>	1/C <sub>c</sub> <sup>E6</sup>
Hanson (1953a)	C_002_1	0.58	5.62	346	28	6.76
Hanson (1953a)	C_002_3	0.56	6.14	320	7	8.39
Hanson (1953b)	C_101_1	0.58	9.6	362	28	6.37
Browne (1967)	C_025_15	0.42	4.4	418	28	5.94
Browne (1967)	C_025_16	0.42	4.4	418	60	8.54
Rostasy (1972)	C_043_3	0.41	5.59	332	28	6.13
Kommendant (1976a)	C_104_1	0.38	4.34	419	28	4.88
Kommendant (1976a)	C_104_2	0.38	4.34	419	90	4.28
Kommendant (1976b)	C_054_1	0.38	4.34	419	28	8.42
Kommendant (1976b)	C_054_2	0.38	4.34	419	90	8.67
Kommendant (1976b)	C_054_14	0.38	4.03	449	28	6.07
Kommendant (1976b)	C_054_15	0.38	4.03	449	90	7.67
Takahashi (1980)	J_015_3	0.4	4.45	400	30	6.35
Kawasumi (1982)	J_018_1	0.47	6.01	304	7	9.16
Kawasumi (1982)	J_018_2	0.47	6.01	304	28	12.08
Kawasumi (1982)	J_018_3	0.47	6.01	304	91	11.32
Kawasumi (1982)	J_018_9	0.49	6.79	286	7	12.96
Kawasumi (1982)	J_018_10	0.49	6.79	286	28	13.95
Kawasumi (1982)	J_018_11	0.49	6.79	286	91	13.21
Brooks (1983)	C_072_2	0.27	3.3	535	28	5.83
Brooks (1983)	C_072_3	0.34	2.6	608	28	17.62
Brooks (1983)	C_072_4	0.27	2.6	628	28	12.75
Brooks (1983)	C_072_5	0.3	2.08	725	28	19.21
Brooks (1984)	C_074_19	0.8	4.75	405	14	24.09
Brooks (1984)	C_074_20	0.67	4.75	366	14	13.61
Brooks (1984)	C_074_21	0.58	4.75	337	14	9.17
Brooks (1984)	C_074_22	0.54	4.75	326	14	9.01
Brooks (1984)	C_074_23	0.5	4.75	311	14	6.73
Brooks (1984)	C_074_24	0.8	4.75	389	14	26.88
Brooks (1984)	C_074_25	0.67	4.75	351	14	20.68
Brooks (1984)	C_074_26	0.56	4.75	317	14	9.36

Table B.6: Details of basic creep data (first part). <sup>1</sup>File corresponds to the file number in the database compiled by Prof. Bažant and his collaborators [34]; <sup>2</sup>w/c: water-to-cement ratio; <sup>3</sup>a/c: aggregate-to-cement mass ratio; <sup>4</sup>c: cement per volume of mixture [kg/m<sup>3</sup>]; <sup>5</sup>t<sub>0</sub>: loading age [days]; <sup>6</sup>1/C<sub>c</sub><sup>E</sup>: Fitted parameter in Eq. 3 [μm/m/MPa].

Author	File <sup>1</sup>	w/c <sup>2</sup>	a/c <sup>3</sup>	c <sup>4</sup>	t <sub>0</sub> <sup>5</sup>	1/C <sub>c</sub> <sup>E6</sup>
Brooks (1984)	C_074.27	0.48	4.75	292	14	9.21
Brooks (1984)	C_074.28	0.4	4.75	267	14	8.29
Bryant (1987)	D_075.1	0.47	1.37	390	8	8.63
Bryant (1987)	D_075.2	0.47	1.37	390	14	8.86
Bryant (1987)	D_075.3	0.47	1.37	390	21	10.83
Bryant (1987)	D_075.4	0.47	1.37	390	28	10.28
Bryant (1987)	D_075.5	0.47	1.37	390	84	10.01
Larrard (1988)	C_122.4	0.44	3.75	410	28	9.08
Shritharan (1989)	C_079.7	0.47	5.09	390	8	8.3
Shritharan (1989)	C_079.8	0.47	5.09	390	14	8.93
Shritharan (1989)	C_079.9	0.47	5.09	390	21	12.57
Shritharan (1989)	C_079.10	0.47	5.09	390	28	10.47
Shritharan (1989)	C_079.11	0.47	5.09	390	84	10.78
Larrard (1990)	D_022.2	0.35	3.96	450	5	4.42
Larrard (1990)	D_022.3	0.35	3.96	450	3	3.14
Larrard (1990)	D_022.4	0.35	3.96	450	7	4.08
Larrard (1990)	D_022.5	0.35	3.96	450	3	4.1
Leroy (1995)	C_123.1	0.5	5.46	342	0.83	4.11
Leroy (1995)	C_123.3	0.5	5.46	342	3	3.83
Leroy (1995)	C_123.4	0.5	5.46	342	7	4.33
Leroy (1995)	C_123.5	0.5	5.46	342	28	4.92
Leroy (1995)	C_123.34	0.33	4.35	426	3	1.52
Leroy (1995)	C_123.35	0.33	4.35	426	7	1.91
Leroy (1995)	C_123.36	0.33	4.35	426	28	3.52
Mazloom (2004)	D_031.2	0.35	3.7	500	7	16.86
Mazloom (2004)	D_031.10	0.35	3.7	500	28	15.1
Mazzotti (2005)	D_033.3	0.42	4.32	418	7	10.75
Mu (2009)	D_036.11	0.58	7.15	275	3	14.61

Table B.7: Details of basic creep data (second part). <sup>1</sup>File corresponds to the file number in the database compiled by Prof. Bažant and his collaborators [34]; <sup>2</sup>w/c: water-to-cement ratio; <sup>3</sup>a/c: aggregate-to-cement mass ratio; <sup>4</sup>c: cement per volume of mixture [kg/m<sup>3</sup>]; <sup>5</sup>t<sub>0</sub>: loading age [days]; <sup>6</sup>1/C<sub>c</sub><sup>E</sup>: Fitted parameter in Eq 3 [ $\mu\text{m}/\text{m}/\text{MPa}$ ].

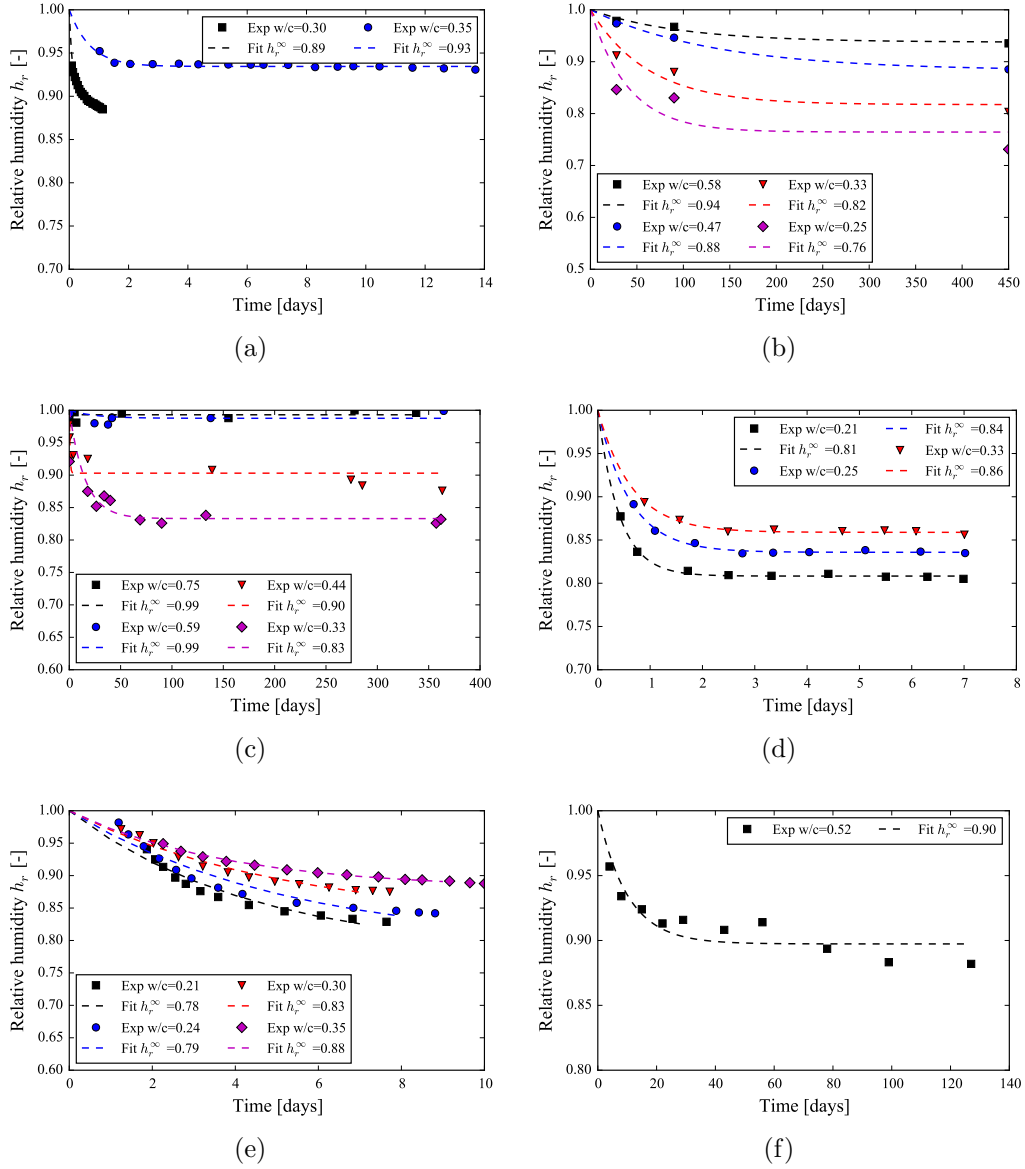


Figure C.8: Evolution of relative humidity with respect to time under auto-genous conditions. Data from (a) [67, 68], (b) [69], (c) [71], (d) [73], (e) [74], (f) [75].

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