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A staggered multiphysics framework for salt crystallization-induced damage in porous building materials

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Abstract

In this paper, a staggered multiphysics framework is proposed for the numerical simulation of salt crystallization-induced damage in porous building materials, such as masonry. This staggered framework is based upon a multiphase model to account for salt transport and crystallization within the porous material and a plastic-damage model to account for the nonlinear mechanical behavior of the material. The staggered structure is composed of a two-way data exchange between the multiphase and the mechanical models. Firstly, crystallization pressure information is passed to the mechanical model to analyze the mechanical response of the material. Secondly, the mechanical outcomes (e.g. damage distribution) are used to update some multiphase model properties (e.g. tortuosity) allowing simulations also beyond the onset of damage. Few simple geometry-based relationships are discussed to update multiphase model properties along with damage. Numerical examples are used to show the capability of the proposed staggered framework for simulating complex interactions among salt transport, salt crystallization, and damage within the porous material, highlighting the possibilities of this modeling approach to conduct simulations also beyond the onset of damage.

Keywords: Multiphase model, Damage, Pore pressure, Spalling, Salt crystallization, Staggered solution, porous materials, masonry.

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1. Introduction

Environmental actions, which are likely to get worse in the future according to climate change predictions [1], can give rise to degradation processes and aging in historic building materials, such as masonry [2]. One of the main threats to degradation of porous building materials is salt crystallization [3, 4]. Indeed, salt crystallization-induced damage in porous building materials can affect the structural performance of existing buildings [5] and the effectiveness of strengthening systems applied on them [6], [7].

In the last decades, a large amount of experimental and theoretical research was dedicated to study the transport of dissolved salt in porous materials [8, 9, 10, 11], salt phase change [12, 13, 14], and pore pressure which results from salt crystallization [12, 15, 16, 17].

Differently from laboratory tests which can typically deal with small-scale specimen and relatively short-terms weathering conditions (whose representativeness of in situ environment is still object of debate [18]), numerical strategies can potentially simulate long-term real-life conditions also on large- and full-scale structures. To this aim, several numerical models have been developed in the last years. In [19], a chemo-hydro-thermo-mechanical model for salt transport and crystallization in porous building materials has been presented considering a linear elastic response of the material matrix. An extension of this model to damage has been discussed in [20], where the spalling in masonry due to salt crystallization has been investigated up to the onset of damage. Derluyn et al. [21] developed a coupled model for heat and mass transport, salt crystallization, deformation and damage able to identify crack nucleation by comparing the effective stress and the tensile strength. Additionally, part of the Authors developed in [22], [23], [24] a coupled multiphase model for the hygro-thermal analysis of masonry structures and prediction of stress induced by salt crystallization. Particularly, this model allowed the simulation of an aging test conducted on a masonry wall exposed to long-term weathering conditions. Such model has been successively extended in [25], where a multi-scale approach for the analysis of mechanical effects induced by salt crystallization in porous media has been developed through the real 3D micro geometry of the porous material and using a linear elastic material mechanical behavior. Lately, Choo &

29 Sun [26] developed a coupled multiphase and phase-field model to investigate cracking and
30 damage from crystallization in pores. This coupled strategy enables simulation of complex
31 fractures in the porous material without explicitly tracking their geometry, allowing the
32 simulation up to the onset of cracking and damage from in-pore crystallization of minerals.
33 A common drawback of this state-of-the-art resides in the limitation of the numerical sim-
34 ulations up to the onset of damage, as many assumptions in the constitutive models break
35 down upon damaging of the material.

36 In this paper, a staggered multiphysics framework is proposed for the numerical simula-
37 tion of salt crystallization-induced damage in porous building materials, such as masonry.
38 This staggered framework is based upon a multiphase model to account for salt transport
39 and crystallization within a porous material [22] and a plastic-damage model to account for
40 the nonlinear mechanical behavior of the material [27].

41 A two-way staggered framework allows for the data exchange between the multiphase and
42 the mechanical models: pore pressure information is passed to the mechanical model to ana-
43 lyze the mechanical response of the material; subsequently, the damage distribution obtained
44 from the mechanical response is used to update some multiphase model properties allowing
45 simulations also beyond the onset of damage. Few simple geometry-based relationships are
46 discussed to update multiphase model properties along with damage.

47 The paper is organized as follows. Section 2 highlights the main aspect of the staggered
48 multiphysics framework herein presented. The multiphase model for salt transport, diffusion
49 and crystallization is briefly recalled in Section 3. Section 4 presents the main aspect of the
50 mechanical model. Section 5 illustrates the strategy for the updating of the multiphase
51 model properties within the staggered framework. Section 6 shows the structure of the
52 staggered framework. Numerical results are presented and discussed in Section 7. Section 8
53 discusses the conclusions of this research work.

54 **2. Multiphysics staggered framework**

55 In this paper, the multiphase model (MPM) presented in [22] for the salt and crystalliza-
 56 tion is coupled with a mechanical model (MM) through a staggered multiphysics framework
 57 to account for the damage induced by salt crystallization. The structure of the coupled
 58 problem is sketched in Figure 1: at a generic step, the distributions of the crystallization
 59 pressure and of the saturation degree of crystallized salt, predicted by the multiphase model,
 60 are used as input data for the nonlinear mechanical model. This, gives in turn the distribu-
 61 tion of the damage in the solid matrix of the porous material as output. Finally, the damage
 variable is used to update some of the multiphase model parameters. In this regards, in the

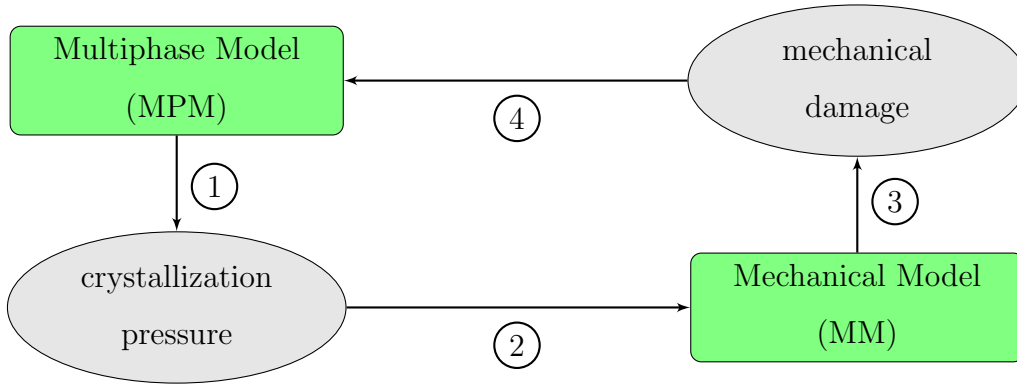


Figure 1: Sketch of the coupled multiphysics model.

62 following the attention is focused only on the tortuosity, for simplicity. However, it is worth
 63 to note that the list of the parameters of the multiphase model to be updated can be easily
 64 enriched without substantially altering the proposed framework.

65 The tortuosity τ is defined as the ratio of the effective length L_e to the direct length L
 66 of a path that connects two points A and B of a porous material. It is considered to vary
 67 during an evolutive analysis not only due to the change in porosity, ϕ , resulting from the
 68 crystallized salt, but also due to the evolution of mechanical damage, as sketched in Figure
 69 2. In particular, as illustrated in Figure 2b, precipitated salt crystals change the porosity
 70 ($\phi_1 < \phi_0$) and the pore geometry resulting in an increase of effective length ($L_{e1} > L_{e0}$) and,
 71 hence, in an increase of the tortuosity ($\tau_1 > \tau_0$). Moreover, as illustrated in Figure 2c, if the
 72

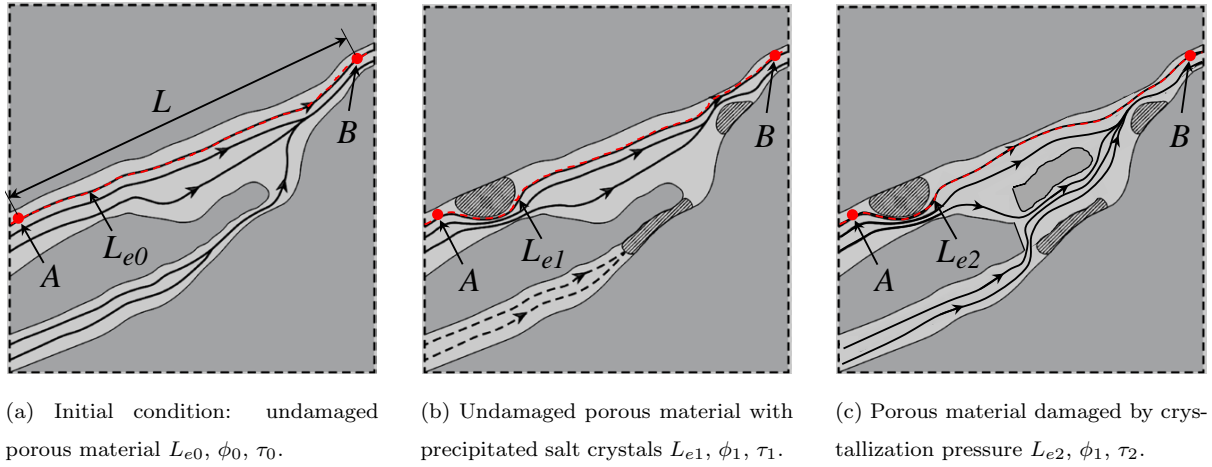


Figure 2: Salt crystallization-induced evolution of porosity and tortuosity of the porous material.

73 damage of the porous material is activated by the crystallization pressure, a further change
 74 may occur in the effective length and, therefore, in the tortuosity.

75 Obviously, in addition to the coupling between the two models, inside each model some
 76 of the parameters are updated during the analysis. Inside the MPM, some parameters are
 77 function of internal variables of the model itself (e.g. the porosity is updated according to
 78 the saturation degree of crystallized salt). Analogously, some parameters of the mechanical
 79 model are updated during the analysis as a function of internal variables of the mechanical
 80 model itself (e.g. the elastic properties are updated according to the level of damage).

81 From a phenomenological point of view, the above-mentioned phenomena (i.e. crystal-
 82 lization process and damaging process) possess different time scale: the first has a slow
 83 evolution characterized by long time units (days, months, years); the latter has a fast evo-
 84 lution, characterized by short time units (seconds, minutes, hours).

85 This is certainly one of the main motivations of using a staggered solution scheme which
 86 allows to exchange the data periodically, at defined time intervals Δt . In this regards,
 87 here we propose a special staggered framework to solve the coupled problem and track
 88 the salt crystallization-induced damage within the porous material, whose organization is
 89 sketched in Figure 3. In particular, the proposed staggered solution has been implemented
 90 in two versions to foster comparisons: one-way coupling (Figure 3a), and two-way coupling

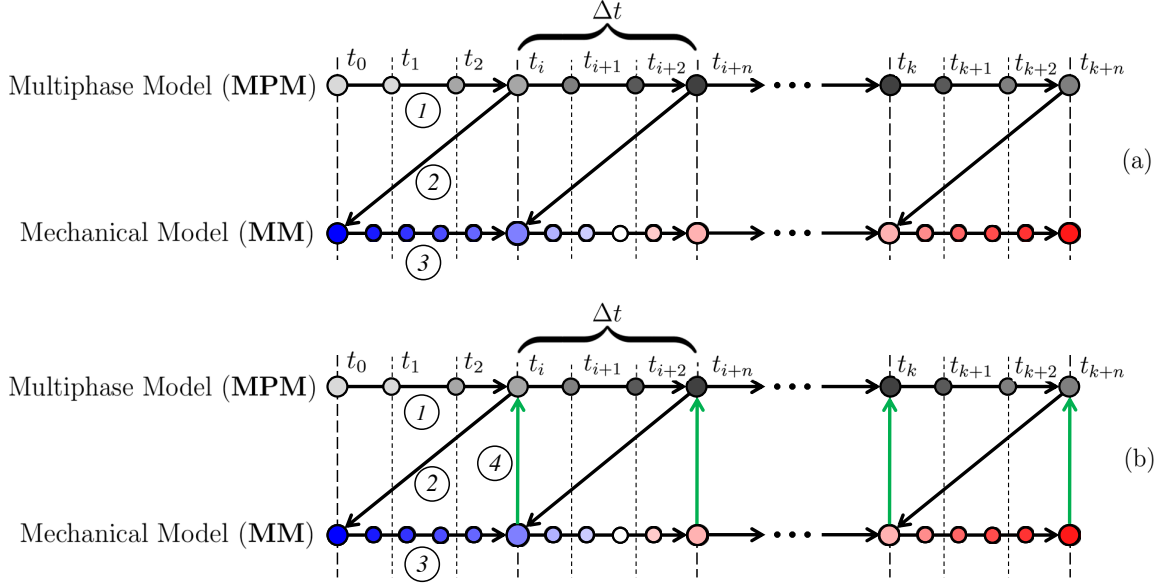


Figure 3: Staggered scheme. One-way coupling (a); two-way coupling (b).

91 (Figure 3b). In the first case, the multiphase model is solved at time t (step 1); then, pore
 92 pressure information is passed to the mechanical model (step 2); the mechanical response
 93 of the material is solved (step 3). In the second case, a further step is performed and the
 94 mechanical damage distribution is used to update the tortuosity τ (step 4).

95 Another motivation to adopt a staggered resolution scheme is related to its computa-
 96 tional convenience and convergence. Indeed, the separate solution of each problem appears
 97 particularly convenient in terms of computational cost and more robust with respect to the
 98 case of a monolithic scheme of such a nonlinear problem [28].

99 3. Multiphase model for salt transport and crystallization

100 In this section, the multiphase model for salt transport and crystallization developed in
 101 [22] is briefly recalled. The porous material is considered as a multiphase medium, which
 102 consists of the skeleton and voids, that may be partly filled with moist air, salt dissolved in
 103 solution and/or salt crystals, see Figure 4. We apply here a macroscopic formulation that
 104 makes use of a representative elementary volume (REV) of a porous medium, large enough
 105 to include all the phases present. Moreover, the region is assumed to be of sufficient size

106 that the average values that characterize a phase are independent of that size [29].
 107 Mass exchange between different phases might arise (i.e. water evaporation, salt crystalliza-
 108 tion/dissolution), as well as the interaction between the fluid phases and the skeleton.

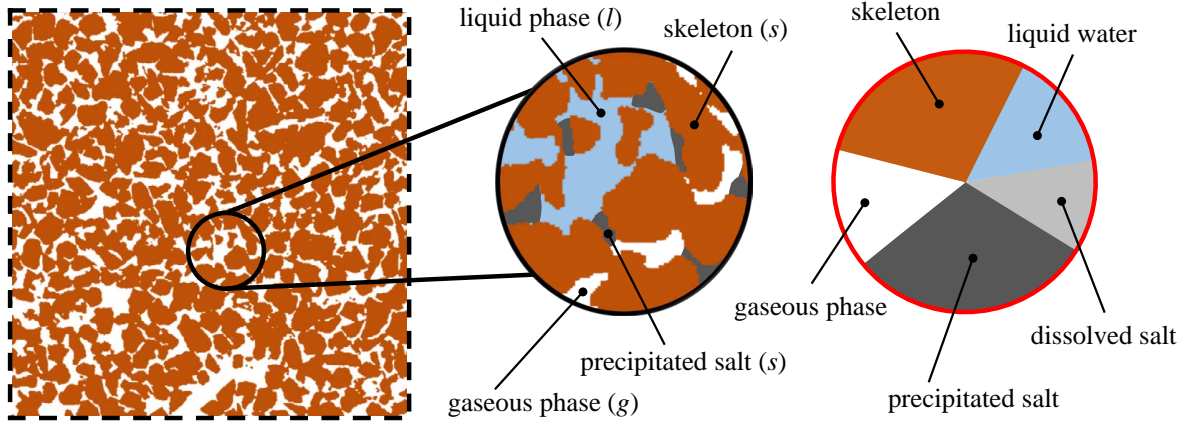


Figure 4: Porous medium: Representative Elementary Volume (REV) of a four-phase mixture composed of the solid matrix (s), liquid solution (l), gas (g), and precipitated salt (s).

109 The content of each component is described by the concentration c_{α}^{π} , defined as the mass
 110 of α in π -phase per unit volume of porous material, or by the corresponding saturation
 111 degree S_{α}^{π} , defined as the pore volume occupied by α in π -phase. The concentration c_{α}^{π} and
 112 the corresponding saturation degree S_{α}^{π} are related as $c_{\alpha}^{\pi} = (\phi_0 \cdot S_{\alpha}^{\pi}) \cdot \rho_{\alpha}^{\pi}$, where ρ_{α}^{π} is the
 113 mass density of α in π -phase.

114 Assuming isothermal conditions, the independent variables assumed to describe the phe-
 115 nomenon are the pore relative humidity h (the vapor pressure divided by the vapor pressure
 116 at saturation) and the mass fraction of dissolved salt ω :

$$\omega = \frac{c_s^l}{c_w^l + c_s^l} \quad (1)$$

117 being c_s^l the concentration of salt in liquid phase and c_w^l the liquid water concentration.
 118 These independent variables are supplemented by the internal variable c_s^s (concentration of
 119 crystallized salt).

120 *3.1. Balance equation*

121 With reference to the REV illustrated in Figure 4, the mass balance equations of moisture
122 and salt can be written as:

$$\frac{\partial c_w}{\partial t} + \nabla \cdot \mathbf{j}_w = -\mu_w^{ls} \quad (2)$$

$$\frac{\partial c_s^l}{\partial t} + \nabla \cdot \mathbf{j}_s^l + \frac{\partial c_s^s}{\partial t} = 0 \quad (3)$$

124 where c_w is the concentration of moisture (defined as the sum of liquid water concentration
125 c_w^l and the vapor water concentration c_w^g), \mathbf{j}_w the moisture flux (defined as the sum of water
126 vapor and liquid water flux: $\mathbf{j}_w = \mathbf{j}_w^g + \mathbf{j}_w^l$), and μ_w^{ls} the rate of liquid water trapped in
127 hydrated salt crystals. The fluxes of liquid water and dissolved salt can be expressed in
128 terms of ω as:

$$\mathbf{j}_w^l = (1 - \omega)\mathbf{j}_{ws}^l - \mathbf{j}_{s,diff}^l \quad (4)$$

$$\mathbf{j}_s^l = \omega\mathbf{j}_{ws}^l + \mathbf{j}_{s,diff}^l$$

129 where \mathbf{j}_{ws}^l is the flux of the liquid phase and $\mathbf{j}_{s,diff}^l$ is the diffusive flux of dissolved salt.

130 *3.2. Constitutive equations*

131 The constitutive equations can be written as:

$$\mathbf{j}_w^g = -\frac{D_v}{R_v T} \nabla p_v \quad (5)$$

$$\mathbf{j}_{ws}^l = -g_\omega(\omega) D_l (S_w^l)^{n_l} \nabla p_c \quad (6)$$

$$\mathbf{j}_{s,diff}^l = -\rho_{ws}^l \frac{D_s}{\tau} f_s(S_w^l) \nabla \omega \quad (7)$$

132 where p_v the vapor pressure, p_c the capillary pressure, ρ_{ws}^l the mass density of the liquid
133 phase, R_v is the gas constant of water vapor and D_v the vapor permeability coefficient.

134 The exponent n_l ranges from 1 to 6 as suggested by [19] and D_l is the liquid conductivity
135 of pure water [30]:

$$D_l = \left[3.8 \left(\frac{A}{\phi_0 \rho_w^l} \right)^2 10^{3(S_w^l - 1)} \right] \frac{\partial c_w}{\partial h} \quad (8)$$

136 being A the water adsorption coefficient, ρ_w^l the mass density of liquid water and S_w^l the
137 degree of saturation of the solution. Moreover, in agreement with [31] the expression $g_\omega =$

138 $\frac{\rho_{ws}^l}{\rho_w^l}(1 - 0.03m)$ is assumed, being m the molality.

139 For the salt diffusion part, D_s is the diffusion coefficient for pure water and $f_s = (S_w^l)^{n_s}$ is a
140 correction factor that takes into account the actual cross section available for diffusion, and
141 n_s is the saturation exponent, here set equal to one.

142 As anticipated in Section 2, when the salt precipitates the porosity tends to decrease due
143 to the voids occupied by the salt. This phenomenon is taken into account in the model by
144 introducing the effective porosity:

$$\phi_{eff} = \phi_0(1 - S_s^s) \quad (9)$$

145 where S_s^s is the crystallized salt saturation degree. Moreover, as described in Section 2,
146 the porosity change due to salt crystallization can alter also the tortuosity and, hence, the
147 salt diffusion. Several expressions which relate tortuosity and porosity can be found in the
148 literature [32, 33, 34, 35, 36, 37, 38, 39]. Here, we refer to the relationship provided in
149 [33, 37, 40]:

$$\tau = 1 - p \ln(\phi_{eff}) \quad (10)$$

150 Varying coefficient p , the above expression can be applied to a bed of uniform spheres as
151 well as to overlapping, non-uniform spheres, see Figure 5.

152 In this model, the instantaneous equilibrium between liquid and vapour water is assumed.
153 Accordingly, the moisture content at a certain temperature can be expressed as a function
154 of the relative humidity. This relation, known as sorption/desorption isotherm, can be
155 considered as a material property and can be obtained by experimental testing, see for
156 instance [41] and the reference therein. The sorption isotherm is commonly determined for
157 pure water, but it is influenced by the dissolved salt due to of the changing contact angle
158 and the surface tension of the solution [42], and can be conveniently expressed in terms of
159 saturation degree of the solution S_{ws}^l as a function of the equivalent relative humidity. In
160 particular, the analytical expression for sorption/desorption curves proposed in [30] is here
161 adopted:

$$S_w^l = \frac{\psi - 1}{\psi - h} h \quad (11)$$

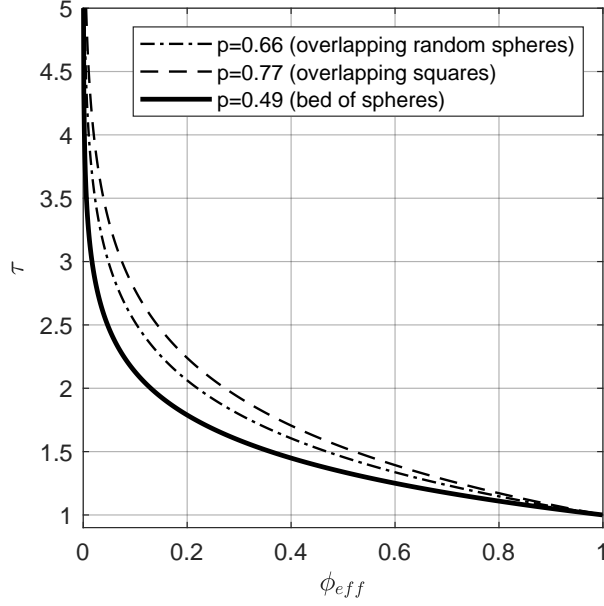


Figure 5: Effect of effective porosity variation on tortuosity for different values of p

162 where ψ is the sorption isotherm parameter. Then, the relation for sorption curves takes
 163 into account for the presence of precipitated salt inside the pores and, indirectly, the effect
 164 of the presence of dissolved salt, see also [22][43] for further details.

165 3.3. Crystallization and dissolution equations

166 The salt crystallization or dissolution depends on the supersaturation ratio which is
 167 defined by the ratio ω/ω_{sat} , being ω_{sat} the dissolved salt concentration at saturation.

168 In particular:

$$\begin{cases} \frac{\omega}{\omega_{sat}} > \alpha_0 \Rightarrow \text{crystallization} \\ \frac{\omega}{\omega_{sat}} < 1 \Rightarrow \text{dissolution} \end{cases} \quad (12)$$

169 where α_0 is the crystallization threshold ($\alpha_0 > 1$ for first crystallization, $\alpha_0 = 1$ for further
 170 crystallization). In general, the threshold of supersaturation ratio for primary crystallization
 171 relies on the properties of the porous material and on the type of salt. The evolution equation
 172 which describes the salt precipitation/dissolution kinetics, i.e. quantifies the amount of salt
 173 which precipitates, can be written as:

$$\frac{\partial c_s^s}{\partial t} = \pi r_p^2 \rho_s^s \frac{n}{V_{tot}} K_c \left| \frac{\omega}{\omega_{sat}} - 1 \right|^P \quad (13)$$

174 where a constant amount of salt nuclei n in the solution, as well as an isotropic distribution
 175 of cylindrical pores and cylindrical nuclei of the same radius of the pores (r_p), is assumed.
 176 In Equation (13), ρ_s^s is the density of the crystallized salt, K_c is the growth rate coefficient,
 177 V_{tot} the pore volume and P is the crystallization process order that relies on the properties
 178 of the porous material and on the type of salt. As noted in [44] in the case of nitrates, the
 179 crystallization-dissolution process could be described using an improved model incorporating
 180 Pitzer's equations.

181 3.4. Crystallization pressure

182 When the salt crystallizes inside the pores it exerts a pressure on the solid matrix that
 183 can be evaluated with the well-known relation introduced by Steiger in [13]:

$$p_s = \frac{vRT}{V_s} \left(\ln \frac{\omega}{\omega_{sat}} + \ln \frac{\gamma}{\gamma_{sat}} \right) \quad (14)$$

184 in which v is the total number of ions released due to complete dissociation of salt, R is the
 185 ideal gas constant, T is the temperature, V_s is the molar volume of precipitated salt, γ is
 186 the mean activity coefficient of the dissolved salt and γ_{sat} is the mean activity coefficient of
 187 the dissolved salt at saturation. As noted in [45], when the porous material is characterized
 188 by high values of pore size (e.g. in natural stones), Eq. (14) could be extended to account
 189 for the influence of pore size on the solution properties.

190 4. Mechanical model

191 As for the mechanical model, a continuum body is considered. The compatibility equa-
 192 tion is written as:

$$\boldsymbol{\varepsilon} = \frac{1}{2}(\nabla \mathbf{u}^T + \nabla \mathbf{u}) \quad (15)$$

193 being \mathbf{u} the displacement vector and $\boldsymbol{\varepsilon}$ the strain tensor. Given the small velocities of the
 194 phases, the equilibrium reads:

$$div(\boldsymbol{\sigma}) + \mathbf{F} = 0 \quad (16)$$

195 where $\boldsymbol{\sigma}$ is the stress tensor and \mathbf{F} the vector of volume forces, which, if no external
 196 distributed loads are prescribed, can be written as:

$$\mathbf{F} = \mathbf{g}\rho_{eff} \quad (17)$$

197 being \mathbf{g} the gravitational acceleration vector and ρ_{eff} the effective mass density defined as:

$$\rho_{eff} = \rho_m + c_w^g + c_w^l + c_s^l + c_s^s \quad (18)$$

198 being ρ_m the mass density of the solid matrix.

199 Isotropic material behavior based on the plastic-damage model developed by Lee and Fenves
 200 [27] is considered. Particularly, this constitutive law aims to phenomenologically represent
 201 the mechanical response of quasi-brittle materials, such as brick. In this framework, the
 202 constitutive relationship is expressed as:

$$\boldsymbol{\sigma} = (1 - D)\mathbb{C}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}_p) - \boldsymbol{\sigma}_s \quad (19)$$

203 where D is the scalar degradation damage variable, \mathbb{C} is the initial undamaged elastic stiffness
 204 tensor, $\boldsymbol{\varepsilon}_p$ is the plastic part of the strain tensor and $\boldsymbol{\sigma}_s$ accounts for the stress induced by
 205 salt crystallization.

206 To account for the different behavior of the quasi-brittle material in tension and compression
 207 (i.e. cracking in tension and crushing in compression) within a plastic-damage framework,
 208 two independent scalar damage variables ($0 \leq d_t < 1$ for tensile regime and $0 \leq d_c < 1$
 209 for compressive regime) are considered in the constitutive model. Indeed, it should be
 210 herein highlighted that a combination of cracking/crushing failure modes can be expected
 211 also due to the stress induced by salt crystallization. Accordingly, the damage state of a
 212 single material point can be overall represented by the single degradation damage variable
 213 D expressed as:

$$D = 1 - (1 - d_t)(1 - d_c) \quad (20)$$

214 which combines tensile and compressive damage states.

215 It can be easily verified that D in (20) fulfills the condition $0 \leq D < 1$ and is equal to either d_t

216 when $d_c = 0$ (tensile case) or d_c when $d_t = 0$ (compressive case). This constitutive law adopts
 217 the concepts of isotropic degradation damage, strain decomposition and effective stress, i.e.
 218 from both damage mechanics and plasticity theories, see Eq. (21). A nonassociative flow
 219 rule, obtained by a Drucker-Prager type plastic potential is considered, is considered to
 220 govern the dilatancy and to define the plastic strain rate. The flow rule is controlled by
 221 the dilatancy angle ψ , generally assumed equal to 10° in agreement with physical evidence
 222 and previous computational approaches, and a smoothing constant ϵ typically assumed
 223 equal to 0.1. Furthermore, a multiple-hardening Drucker-Prager type surface is adopted
 224 as yield surface. This surface is governed by the ratio f_{b0}/f_{c0} between the biaxial initial
 225 compressive strength f_{b0} and the uniaxial initial compressive strength f_{c0} and a constant ρ ,
 226 which represents the ratio of the second stress invariant on the tensile meridian to that on
 227 the compressive meridian at initial yield. Generally, $f_{b0}/f_{c0} = 1.16$ and $\rho = 2/3$ for quasi-
 228 brittle materials, such as brick. The general parameters adopted for brick are collected in
 229 Table 1.

| ϵ | ψ | f_{b0}/f_{c0} | ρ |
|------------|------------|-----------------|--------|
| 0.1 | 10° | 1.16 | $2/3$ |

Table 1: General parameters for the brick plastic response

230 As regards the stress induced by salt crystallization, collected in $\boldsymbol{\sigma}_s$, it should be noted
 231 that, basing on the so called Law of Partial Pressures [46], $\boldsymbol{\sigma}_s$ can be evaluated as:

$$\boldsymbol{\sigma}_s = \mathbf{I}\sigma_s = \mathbf{I}bS_s^s p_s \quad (21)$$

232 where b is the Biot's coefficient and \mathbf{I} is the identity operator.

233 5. Updating of multiphase model parameters

234 We now specify how the damage induced by the salt crystallization affects the multi-
 235 phase model parameters. In particular, as anticipated in Section 2, the attention is herein
 236 focused on the tortuosity which is herein assumed to be dependent on the actual mechanical

237 damage D . Accordingly, Equation (7) is updated along with the current state of damage,
 238 leading to an updating of the multiphase model. For the tortuosity update, we assume a
 239 law analogous to that used for lithium-ion battery electrodes, where the underlying micro-
 240 structural tortuosity controls the macroscopic charge capacity, average lithium-ion diffusiv-
 241 ity, and macroscopic resistivity of the cell [47]. Then, the Equation (7) is updated by means
 242 of the following definition of the effective tortuosity parameter:

$$\tau_{eff}(D) = \tau \cdot \beta_{\tau}(D) \quad (22)$$

243 where β_{τ} is the tortuosity damage function that describes how tortuosity evolves along with
 244 damage in the porous structure.

245 In order to infer the β_{τ} function, we suppose here the porous material as a bed of spheres
 246 (Figure 6). If we consider that for a specific region of interest (see Figure 6a), we reach
 247 a level of crystallization pressure capable to induce damage, then, we can assume a local
 248 change of τ according to the evolution of the simplified pore structure. With this in mind,
 249 we consider the scenario illustrated in Figures 6a-6d: for increasing damage, β_{τ} increases
 250 until the damage reaches a certain threshold value D^* ($\beta_{\tau} = \beta_{\tau,1}$ for $D = D^*$) and then
 251 decreases till specific value $\beta_{\tau,2}$ defined by the evolution of the particle size.

252 A possible evolution of β_{τ} , along with the damage variable D , is described in Figure 6(e):
 253 we use the smooth-stepping function to modulate the value of β_{τ} along the variation of the
 254 damage variable D with reference to the damage threshold level D^* :

$$\beta_{\tau}(\beta_{\tau,1}, \beta_{\tau,2}, D, D^*) = \begin{cases} 1 + (\beta_{\tau,1} - 1)\left(\frac{D}{D^*}\right)^2\left(3 - \frac{2D}{D^*}\right), & \text{for } D = [0, D^*] \\ \beta_{\tau,1} - \left(\frac{D-D^*}{1-D^*}\right)^2\left(3 - 2\left(\frac{D-D^*}{1-D^*}\right)\right)(\beta_{\tau,1} - \beta_{\tau,2}), & \text{for } D = [D^*, 1] \end{cases} \quad (23)$$

255 In general, we can consider that $\beta_{\tau,2}$ could be precomputed in order to fulfill the following
 256 constrain:

$$\tau_{eff} = \tau\beta_{\tau} = 1 \quad \text{if } D = 1 \quad (24)$$

257 6. Computational method for the staggered solution

258 We now detail the implementation of the staggered framework sketched in Figure 3 for
 259 the solution of the coupled multiphysics problem.

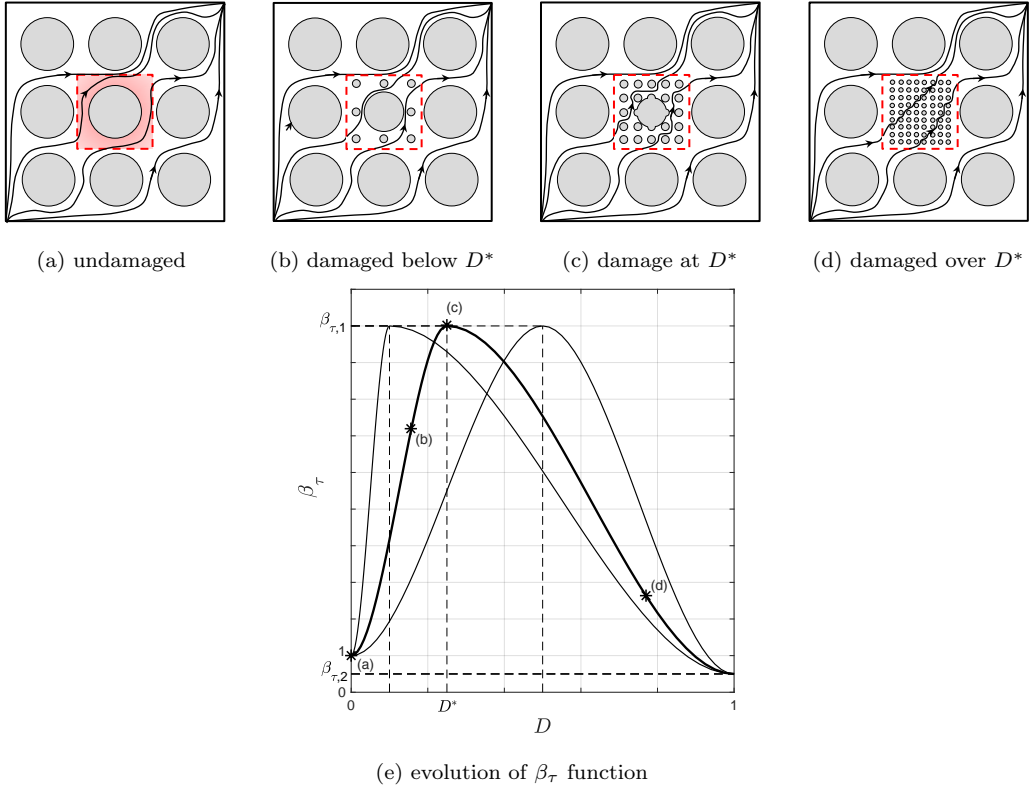


Figure 6: Tortuosity update due to mechanical damage.

260 6.1. Space discretization

261 In order to implement the model, the domain is discretized in space using the finite
 262 element method for the salt transport and crystallization problem and for the mechanical
 263 problem. The proposed staggered procedure allows for the use of different meshes when
 264 exchanging informations between MPM and MM.

265 6.2. Fields transfer operation strategy

266 According to the pseudo-code reported in Algorithm 1 and illustrated in Figure 3 (steps
 267 1, 2, 3, and 4 of Figure 3 are highlighted in Algorithm 1 too), the proposed staggered
 268 technique could admit some variants depending on the different use of the Fields Transfer
 269 Operation (FTO) between the two sets of field equations. These operations (namely step 2
 270 and step 4 of the algorithm 1) can be run at every time step in each field, or can be opti-
 271 mized based on the different evolution times of MPM and MM. In particular, the effect of

Algorithm 1: Pseudo-code of the required steps to run the staggered solution

Input: $\phi_0, \tau_0, \omega_{sat}, \Delta t, D^*, D$ and set $n = 1$

Output: $c_s^s, S_s^s, \sigma, D, \phi_{eff}, \tau_{eff}$

① **MPM** - *Compute/Update* $\phi_{eff}, \tau_{eff}(\phi_{eff}, D), p_s$

if $t = n\Delta t$ **then**

$n = n + 1$

② **FTO** \rightarrow *from MPM to MM*

$\sigma_s = \mathbf{I}\sigma_s = \mathbf{I}bS_s^s p_s$

③ **MM**

Compute/Update D

④ **FTO** \rightarrow *from MM to MPM*

Compute/Update $\beta_\tau(\beta_{\tau,1}, \beta_{\tau,2}, D, D^*)$

else

$t = t + 1$

end

Repeat *Steps 1 to 4*, till $t = t_{final}$

272 the choice of the time interval Δt between two FTO has been investigated, see Appendix A.2.

273

274 7. Numerical examples

275 The multiphase model is applied to evaluate the effects of the mechanical coupling on the
 276 salt transport and crystallization processes in a benchmark consisting in a fired clay masonry
 277 panel exposed to specific boundary conditions, inspired by the benchmark analyzed in [20].
 278 To this aim, we analyze a portion of the panel representative for a generic region of interest,
 279 as illustrated in Figure 7: we consider a generic transverse section of the panel (see cutting
 280 plane Π_0) and focus the attention on a small portion of it depicted in Figure 7.

281 In the following, we use the same mesh to solve the fields variables of both models but
 282 with finite elements of different order: the primary variables of the multiphase model, h
 283 and ω , are interpolated based on Lagrangian shape functions adopting standard 9-node

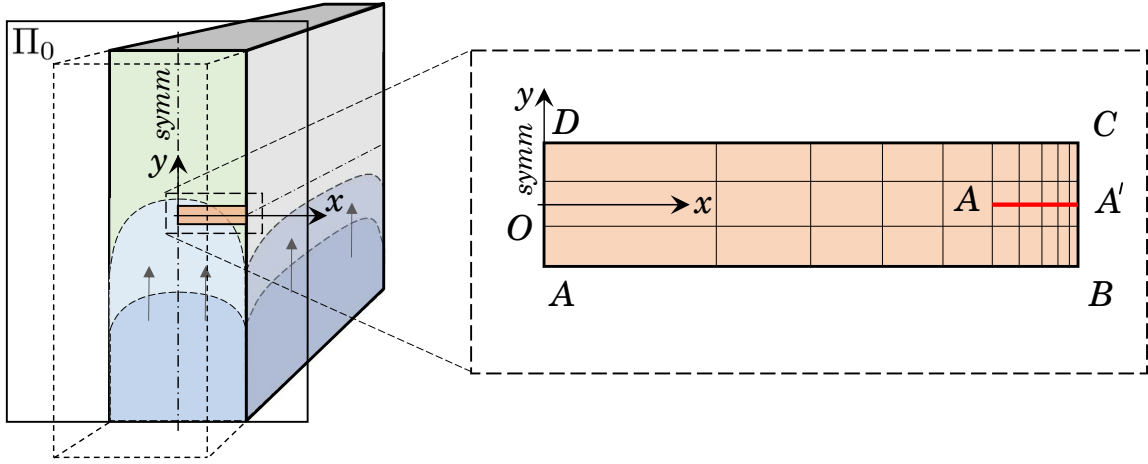


Figure 7: Illustration of the boundary conditions over a region of interest on the cutting plane Π_0 : \overline{DC} and \overline{AB} no flux, $u_y = 0$; \overline{AD} prescribed relative humidity \bar{h} and prescribed mass concentration of dissolved salt $\bar{\omega}$ and symmetry boundary condition for the displacement field; \overline{BC} allowed evaporation with prescribed environmental humidity. The cutting line $A - A'$ highlighted in red color.

284 elements; the mechanical primary variable, the displacement, is also interpolated based on
 285 standard Lagrangian shape functions but adopting 4-node elements. Mesh is constructed
 286 using $N_x = 3$ and $N_y = 100$ elements along the directions x and y , respectively, using a
 287 0.1 element ratio biasing along the x direction (a mesh sensitivity analysis has been also
 288 conducted and the main results are collected in Appendix A.1).

289 The time slot Δt , that rules the FTO, has been set equal to $\Delta t = 6$ hours (the sensitivity
 290 to the FTO time interval has been tested for several Δt values and the main results are
 291 collected in Appendix A.2).

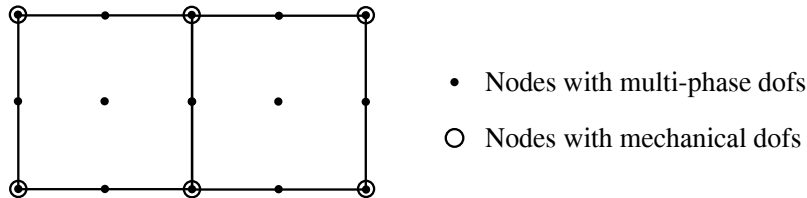


Figure 8: An example mesh illustrating the location of finite element degrees of freedom. Note that linear element and quadratic element can be employed for modeling the two problems.

292 *7.1. Benchmarks definition*

293 Three specific numerical setups are considered and they are organized as follows:

294 **BM#1** In this case, no tortuosity update is considered [22] (i.e. the tortuosity is set
295 constant $\tau = 1$). FTOs are set according to Figure 3(a), using therefore a one-way
296 coupling framework.

297 **BM#2** An extension of the MPM in [22] is used in this case to solve the problem, updating
298 the tortuosity as function of the effective porosity with the relation provided for bed
299 of spheres defined using $p = 0.49$ in Equation (10). Hence, the constitutive law in
300 Equation (7) is updated in time through the definition of the salt diffusion coefficient:
301 an increase of tortuosity will correspond to a drop of the diffusive flux of dissolved salt.
302 The results are illustrated tracking in time variables as for BM#1 plus the tortuosity
303 variable $\tau(\phi_{eff})$. FTOs are set according to Figure 3(a), using therefore a one-way
304 coupling framework.

305 **BM#3** This case implements the full coupling of the MPM and MM along the evolution in
306 time: the tortuosity is updated along with the effective porosity and the mechanical
307 damage using Equations (22) and (23) setting $\beta_{\tau,1} = 2$, $\beta_{\tau,2} = 1$ and $D^* = 0.2$. Results
308 are illustrated tracking in time variables as for BM#2. FTOs are set according to
309 Figure 3(b), using therefore a two-way coupling framework.

310 For each benchmark we consider 96 hours simulations along with the following Environmen-
311 tal Boundary Conditions (EBC) aimed at simulating a portion in time of an ideal weathering
312 real cycle: constant environmental humidity $h_{env} = 50\%$ on \overline{BC} (see Figure 7), initial porous
313 material humidity equal to 50%, on \overline{AD} prescribed humidity \bar{h} that rises from 50% to 99,6%
314 in few hours and prescribed mass concentration of dissolved salt $\bar{\omega} = 0.04$ (see Figure 7).

315 As regards the MPM, material properties are set according to [48], and sodium chloride
316 (NaCl) is chosen as salt present in the saline solution. Summary of the model parameters is
317 reported in Table 2. The crystallization process order P appearing in the growth rate law in

318 Equation (13) is set equal to one, as typically used for sodium chloride salt [48]. Neverthe-
 319 less, a further numerical investigation has been conducted, see Appendix A.3, to test the
 320 robustness of the proposed procedure in case of different salts with different crystallization
 321 process order P that rules the growth rate law in Equation (13). Obtained results show a
 322 significant stability of the proposed method.

323 The time discretization is carried out by means of the backward finite difference method.
 324 A standard iterative strategy based on the Newton–Raphson method is applied to solve the
 325 MPM non-linear system of equations [49].

326 The MM is defined using the following mechanical material parameters for masonry
 327 $E = 1.8GPa$ and $\nu = 0.2$. Being the model formulated in the context of non-associated
 328 plasticity [27], the plastic potential is defined by the dilatancy angle ψ , assumed equal to
 329 10° as generally done for masonry, as well as by a smoothing parameter ϵ assumed equal to
 330 0.1. In addition, the strength domain is specified by the ratio f_{b0}/f_{c0} between the biaxial
 331 f_{b0} and uniaxial f_{c0} initial compressive strengths, assumed equal to 1.16, and by the shape
 332 constant ρ_D , assumed equal to $2/3$ [27]. Compressive strength is set equal to $f_c = 2.0MPa$
 333 and the tensile strength is set equal to $f_t = 0.12MPa$.

334 7.2. Results discussion

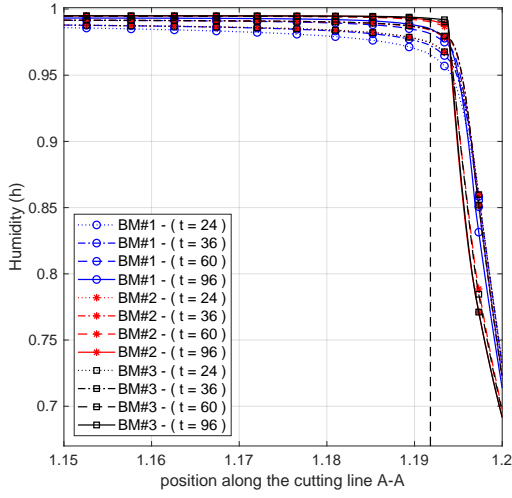
335 The discussion of the results is carried out by a direct comparison of the outcomes for
 336 benchmarks BM#1, BM#2 and BM#3 in Figure 9 and Figure 10. In these figures, results
 337 are illustrated tracking in time the following fields variables: humidity h , supersaturation
 338 ratio ω/ω_{sat} , degree of saturation of crystallized salt S_s^s , tortuosity τ , stress σ_s and degra-
 339 dation damage variable D along the cutting line $A - A'$ (where salt crystallization mainly
 340 occurs), as described in Figure 7. As expected, the rapid variation of the humidity near the
 341 evaporating surface (\overline{BC} boundary in Figure 7), shown in Figure 9(a), leads to an increase
 342 of the supersaturation ratio, see Figure 9(b). In particular, by inspecting Figure 9(b) it
 343 is possible to note that, for all the setups, after $t = 24$ hours, the triggering condition of
 344 Equation (12) will cause salt precipitation near the surface \overline{BC} . This is confirmed by Figure
 345 10(a) and (c) showing an increase of the saturation degree S_s^s and of the stress σ_s in the

| | Quantity | Value | Units | Source |
|----------------|---|------------------------|------------------|--------------|
| K_c | Growth rate coefficient | 0.03 | $\mu m/s$ | [50][51] |
| n | Nuclei in solution | 4×10^{-6} | $(\mu m^3)^{-1}$ | [52] |
| ϕ_0 | Initial porosity | 26% | — | [22] |
| ω_{sat} | Concentration of dissolved salt at saturation | 0.264 | kg/kg | [22][24][53] |
| D_v | Vapor permeability coefficient | 0.0039 | m^2/h | [30] |
| α_0 | Crystallization threshold | 1 | — | [52] |
| ρ_s^s | Salt NaCl density | 2160 | kg/m^3 | Literature |
| r_p | Mean pore radius | 0.700 | μm | [22] |
| A | Water adsorption coefficient | 0.185 | $kg/m^2/s^{0.5}$ | [22] |
| K_s | Salt diffusion Coefficient | 0.499×10^{-9} | m^2/s | [22] |

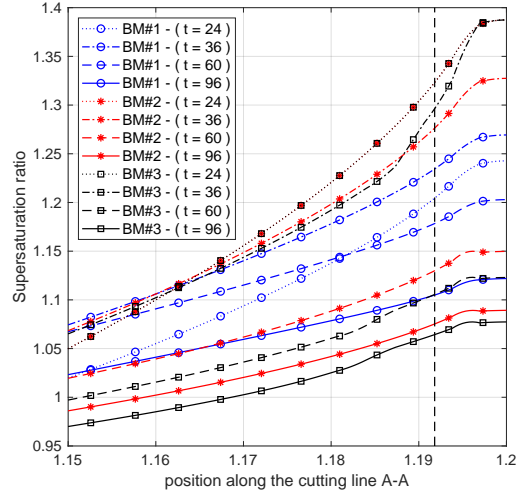
Table 2: Summary of the model parameters used in the numerical tests.

346 same zone (the stress σ_s are computed ad a post-process of the MPM results using Equation
347 (21)). In particular, for the BM#1 the stress concentrates near $x = 1.1918$ (highlighted
348 through a vertical dotted line in Figures 9 and 10) with a maximum magnitude equal to
349 $\sigma_s = 10.0MPa$ for $t = 60$ hours. Furthermore, it is interesting to note that for BM#1 after
350 $t = 60$ hours the S_s^s increases whether, due to the decreasing supersaturation ratio, the σ_s is
351 decreasing too. Moreover, it is worth to note that for BM#1 the single degradation damage
352 variable D , shown in Figure 10(d), remains equal to zero during the whole simulated time.

353 Differently, the effective description of the tortuosity, introduced for BM#2 and BM#3,
354 leads to an update of the various quantities (the stress is now characterized by an increased
355 peak value of $16.5MPa$ and of $18.3MPa$ at $t = 36$ hours for the BM#2 and BM#3 setups,
356 respectively) but, above all, to the activation of the mechanical damage. In particular,
357 inspecting Figure 10(c) and (d), it can be noted that for BM#2 and BM#3 after $t = 36$
358 hours the stress decreases, due to damage activation (in the zone near $x = 1.1918$, D passes
359 from 0.18 at time $t = 36$ hours to about 0.92 at $t = 96$ hours). Furthermore, it appears
360 worth to note that, for BM#3, the tortuosity drops where the damage variable exceeds the



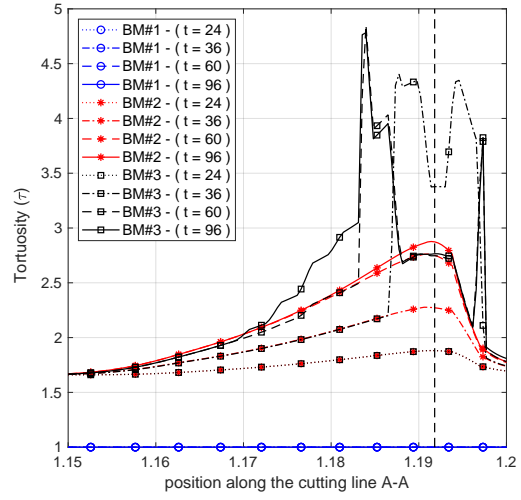
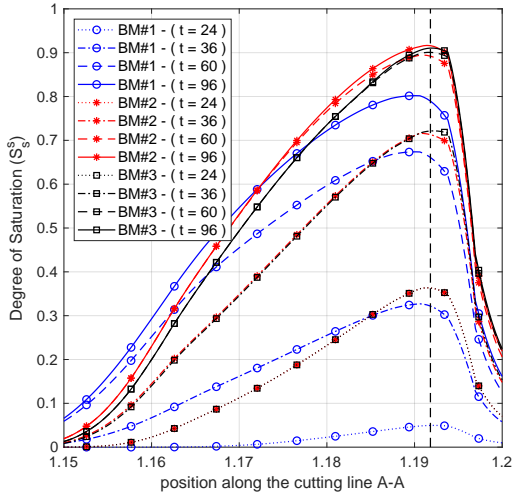
(a)



(b)

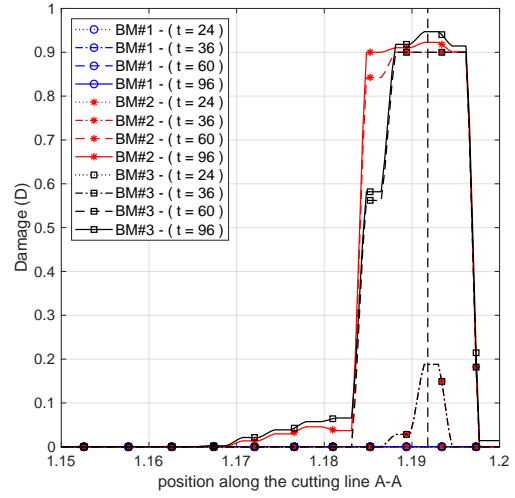
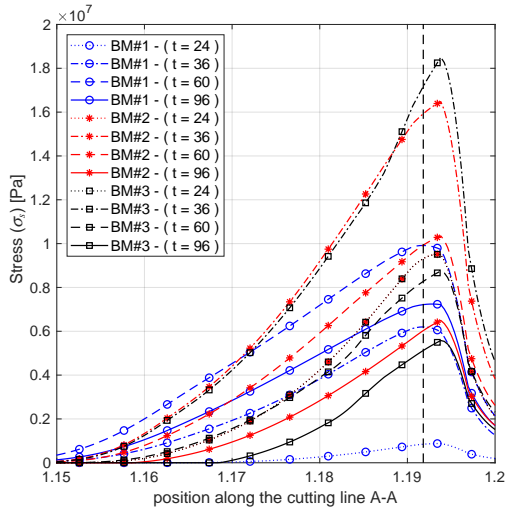
Figure 9: Benchmarks results comparison: illustration of fields variation for $t = [24, 36, 60, 96]$; humidity h (a), supersaturation ratio ω/ω_{sat} (b).

361 threshold $D^* = 0.2$, according to the full coupling of the FTO that admits the combined
 362 evolution of fields. Finally, it should be noted that for time interval between 36 and 96 hours
 363 the values of the tortuosity firstly increase and then drop to values provided by Equations
 364 (24) where the damage variable D is close to one, see Figure 10(b). In this regard, is worth
 365 underlining that some of the curves perfectly overlap in the first part of the simulated time
 366 due to the time required for the accumulation of salt, stress, damage, etc.



(a)

(b)



(c)

(d)

Figure 10: Benchmarks results comparison: illustration of fields variation for $t = [24, 36, 60, 96]$; degree of saturation S_s^s (a), the tortuosity τ (b), the stress σ_s (c) and damage variable D (d).

367 **8. Conclusions**

368 A staggered multiphysics framework for the numerical simulation of salt crystallization-
369 induced damage in porous building materials, such as masonry, was presented and discussed.
370 This framework is based upon a multiphase model to account for salt transport and crys-
371 tallization within a porous the porous material and a mechanical model to account for the
372 mechanical damage of the material. The solution algorithm is composed of a two-way data
373 exchange between the multiphase and the mechanical models. Firstly, crystallization pres-
374 sure information is passed to the mechanical model to analyze the mechanical response of
375 the material. Secondly, the mechanical outcomes (e.g. damage distribution) are used to
376 update some multiphase model properties (e.g. tortuosity) allowing simulations also be-
377 yond the onset of damage. Few simple geometry-based relationships are discussed to update
378 multiphase model properties along with damage.

379 Numerical examples are used to show the capability of the proposed staggered framework
380 for simulating complex interactions among salt transport, salt crystallization, and damaging
381 within the porous material. Numerical results highlight the potential influence of updating
382 multiphase model properties along with damage to conduct simulations also beyond the
383 onset of damage. Further investigations could be also devoted to the role of the pore size
384 since, in general, the maximum tortuosity does not correspond to the minimal porosity.
385 Accordingly, the tortuosity would not only be a function of the porosity but also of the pore
386 structure of the porous material.

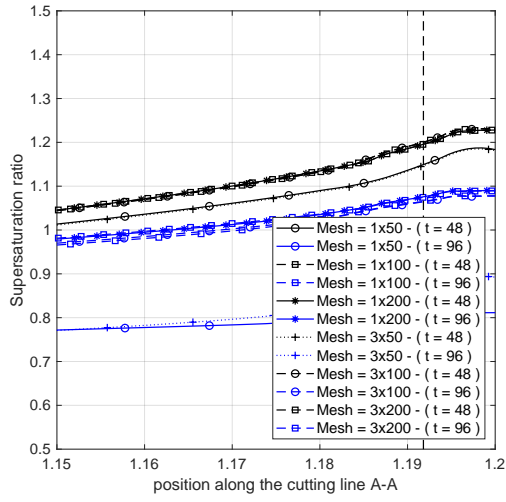
387 **Acknowledgement**

388 The support from the European project CRYSTINART through the Joint Programming
389 Initiative on Cultural Heritage (JPI-CH) is hereby acknowledged.

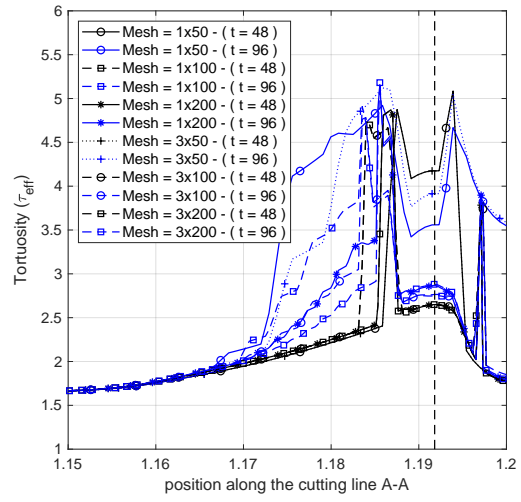
390 **Appendix A. Sensitivity analyses**

391 *Appendix A.1. Mesh refinement*

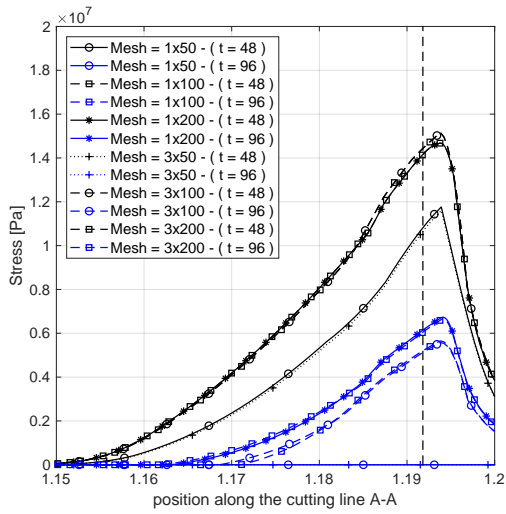
392 BM#3 is used with uniform mesh refinements in both directions ($N_x \times N_y$) using $N_x =$
393 $[50, 100, 200]$ and $N_y = [1, 3, 5]$. Figures A.11, illustrates the results obtained for super-
394 saturation ratio ω/ω_{sat} , the tortuosity τ , the stress distribution $\bar{\sigma}_s$ and damage variable D
395 respectively, using $N_y = 1$ and $N_y = 3$. As expected, Figure A.11 shows a variation in time
396 for variables depending on the mesh density close to the surface, i.e. where the stress varia-
397 tion is severe. Nevertheless, according to these results the solution obtained using $N_x = 100$
398 (independently from the value of N_y) are comparable. This aspect is mainly due to the
399 specific definition of the EBC which defines a 1D-like problem.



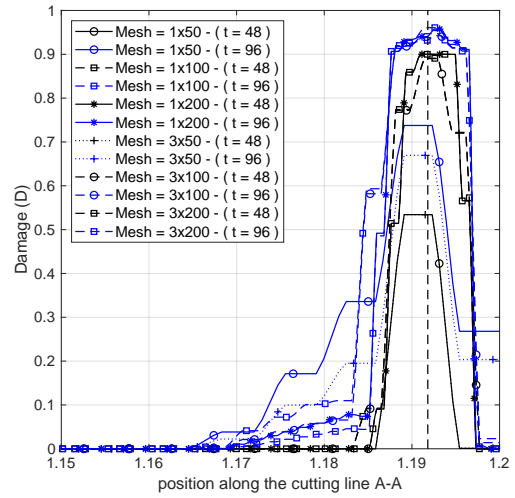
(a)



(b)



(c)



(d)

Figure A.11: Benchmark#3 - mesh sensitivity: Damage, Stress and Tortuosity - Mesh 1x50, 1x100, 1x200 and Mesh 3x50, 3x100, 3x200 - Time Slot $\Delta t = [48, 96]$ hours, $D^* = 0.2$

400 *Appendix A.2. Time slot staggered sequence*

401 The sensitivity of the FTO time interval is tested for several Δt values. Results are
 402 shown in terms tortuosity $\tau(\phi_{eff}, D, D^*)$, averaged stress $\bar{\sigma}_s$ and the single degradation
 403 damage variable D plots: along the cutting line $A - A'$ for $t = [48, 96]$ hours Figure A.12(a),
 404 and for different sampling at $x = 1.1918$ Figure A.12(b) for $\Delta t = [1, 2, 6, 12, 24]$ hours.
 405 By inspecting Figure A.12, it is possible to note that the evolution of the averaged stress is
 406 slightly depending on the definition of the time interval Δt , on all the solutions are matching
 407 the reference solution obtained using $\Delta t = 1$.

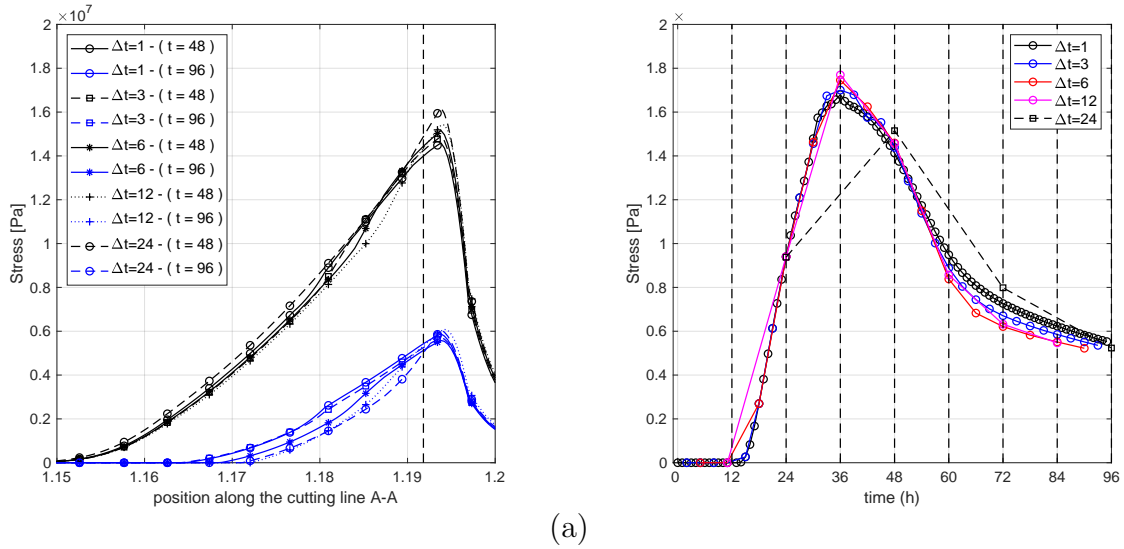


Figure A.12: Staggered time intervals sensitivity - stress field evaluated using FTO time intervals equal to $\Delta t = [1, 3, 6, 12, 24]$ along the time evolution at $x = 1.1918$ (a) and along the $A - A'$ section for $t = [48, 72]$ hours (b).

408 The influence of the choice of time sampling on the effective tortuosity is shown in Figure
 409 A.13. By inspecting Figure A.13 it is possible to note that the evolution of the tortuosity is
 410 characterized by quick variation in time. Accordingly, tortuosity tends to rapidly increasing
 411 as soon as the single degradation damage variable is different from zero, but it also rapidly
 412 decreases when the single degradation damage variable reaches the damage threshold D^* ,
 413 this rapid variation takes 6-8 hours in total when measured at point $x = 1.1918$, see Figure
 414 A.13(a) for $\Delta t = 1$ hours. Therefore, models that are transferring information between fields

415 using $\Delta t \geq 6$ cannot record this rapid variation. As a matter of fact, solutions obtained with
 416 $\Delta t = [12, 24]$ are diverging from the reference solution ($\Delta t = 1$) whether solutions obtained
 417 with $\Delta t = [3, 6]$ are matching the reference solution. Similar evidence can be described for
 418 the resulting damage D in Figure A.14.

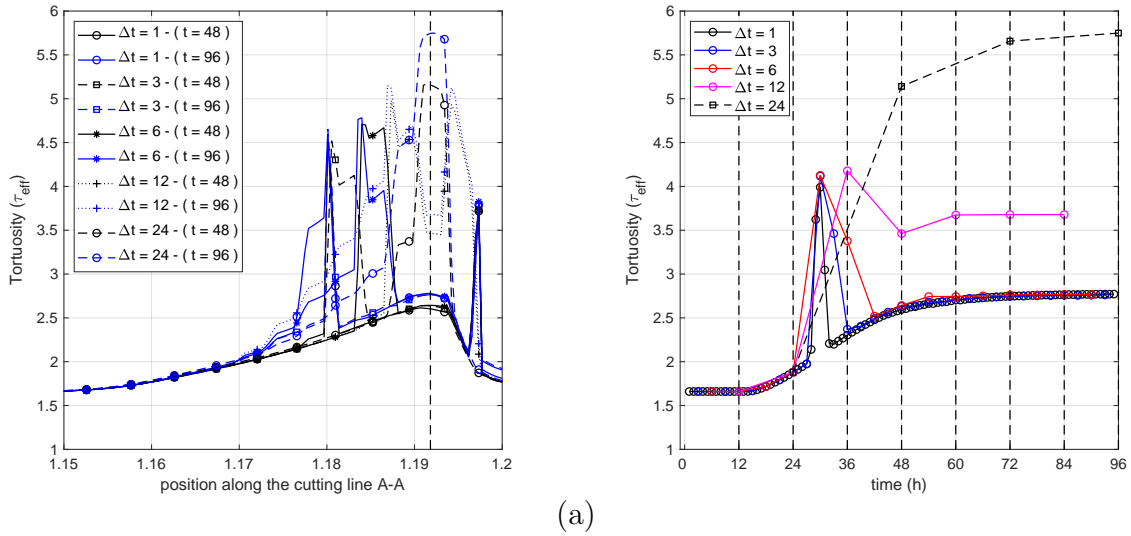
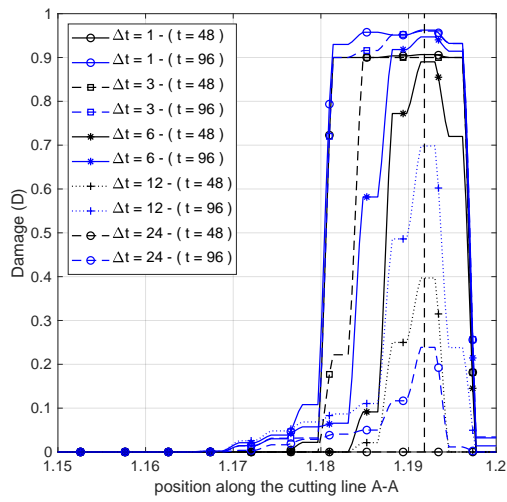
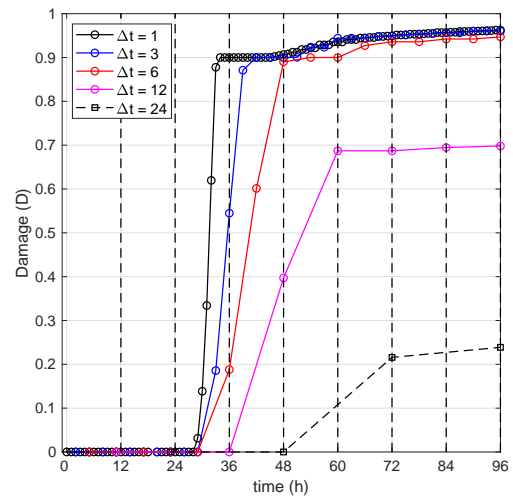


Figure A.13: Staggered time intervals sensitivity - tortuosity field evaluated using FTO time intervals equal to $\Delta t = [1, 3, 6, 12, 24]$ along the time evolution at $x = 1.1918$ (a) and along the $A - A'$ section for $t = [48, 72]$ hours (b).



(a)

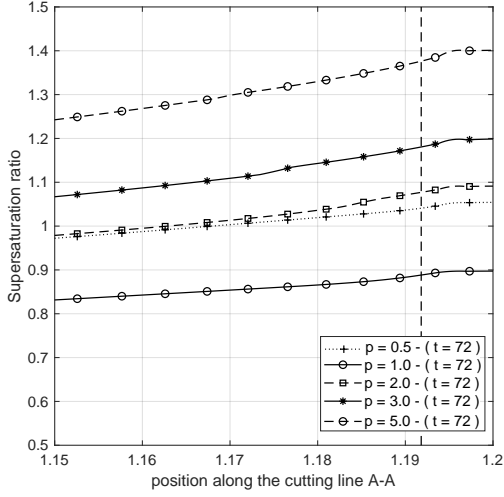


(b)

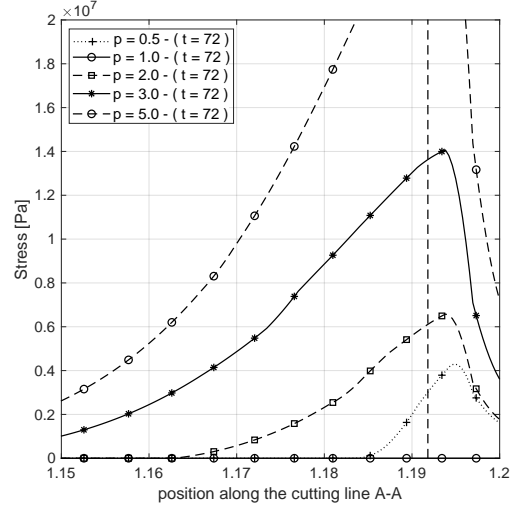
Figure A.14: Staggered time intervals sensitivity - damage field variable evaluated using FTO time intervals equal to $\Delta t = [1, 3, 6, 12, 24]$ along the time evolution at $x = 1.1918$ (a) and along the $A - A'$ section for $t = [48, 72]$ hours (b).

419 *Appendix A.3. Kinetic law power law exponent*

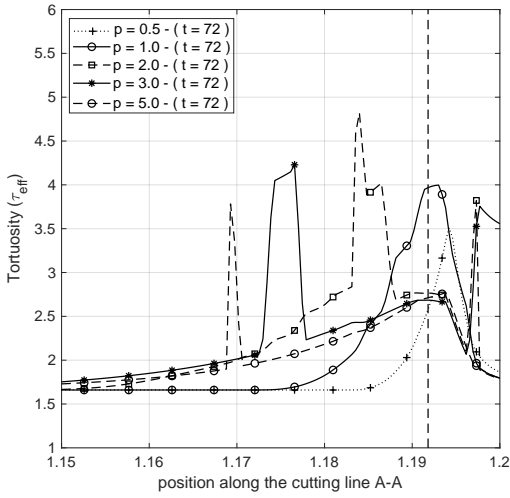
420 The power P appearing in the growth rate law in Equation (13), is the crystallization
421 process order depending on the properties of the porous material and the salt present in the
422 saline solution. We investigated five cases with the process order equal to: $P = [0.5, 1.0,$
423 $2.0, 3.0, 5.0]$ for the BM#3 setup to test the robustness of the procedure, see Figure A.15
424 which refers to $t = 72$ hours. The parameter P depends on the salt type and influences the
425 solution properties, crystal volume and crystallization pressure along with the shape and
426 magnitude of the damaged area, see Figure A.15. As can be noted, the numerical approach
427 appears robust and does not show stability problems by changing P .



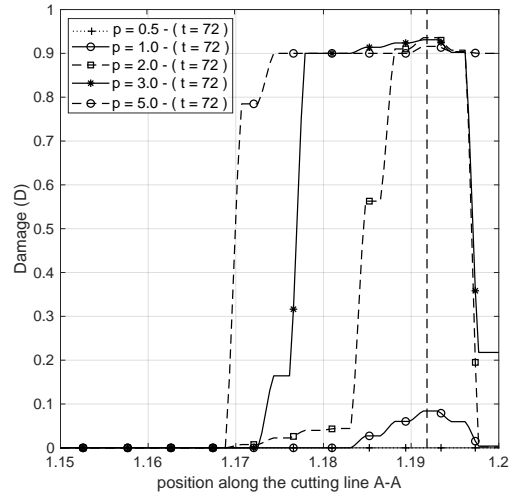
(a)



(b)



(c)



(d)

Figure A.15: Kinetic law power exponent: illustration of fields variation at $t = 72$ for $p = [0.5, 1.0, 2.0, 3.0, 5.0]$: supersaturation ratio ω/ω_{sat} (a), the stress distribution $\bar{\sigma}_s$ (b), the tortuosity τ (c) and damage variable D (d) using a mesh formed by $N_x = 3$ and $N_y = 100$ elements and $\Delta t = 6$ hours.

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