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1 **Chemically induced compaction bands:**
2 **Triggering conditions and band thickness**

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5
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8
9 **Key points**

- 10 - Compaction band instabilities due to cataclasis and dissolution in rocks
11 - Strong chemo-poro-mechanical coupling, grain breakage and chemical softening
12 - Regions of instability, compaction band thickness and periodicity

13
14 **Abstract**

15 During compaction band formation various mechanisms can be involved at different scales.
16 Mechanical and chemical degradation of the solid skeleton and grain damage are important
17 factors that may trigger instabilities in the form of compaction bands. Here we explore the
18 conditions of compaction band formation in quartz- and carbonate-based geomaterials by
19 considering the effect of chemical dissolution and grain breakage. As the stresses/deformations
20 evolve, the grains of the material break leading to an increase of their specific surface.
21 Consequently, their dissolution is accelerated and chemical softening is triggered. By accounting
22 for (a) the mass diffusion of the system, (b) a macroscopic failure criterion with dissolution
23 softening and (c) the reaction kinetics at the micro level, a model is proposed and the conditions
24 for compaction instabilities are investigated. Distinguishing the micro-scale (grain level) from the
25 macro-level (Representative Elementary Volume) and considering the heterogeneous
26 microstructure of the REV it is possible to discuss the thickness and periodicity of compaction
27 bands. Two case studies are investigated. The first one concerns a sandstone rock reservoir which
28 is water flooded and the second one a carbonate rock in which CO₂ is injected for storage. It is
29 shown that compaction band instabilities are possible in both cases. (200 words)

30
31
32
33 **Index terms**

34 STRUCTURAL GEOLOGY: Mechanics, theory and modeling

1 NONLINEAR GEOPHYSICS: Bifurcations and attractors, Critical phenomena, Pattern
2 formation

3 COMPUTATIONAL GEOPHYSICS: Modeling

4

5 **Keywords**

6 Compaction band instabilities, Strain localization, Chemo-poro-mechanical coupling, Grain
7 breakage, Rocks, Reservoirs

8

9

10 **1. Introduction**

11 The presence of compaction bands in nature may provide useful information on various
12 geological processes as it is an indication of the stress state history of a geological formation.
13 Compaction bands are usually characterized by a significant reduction of the pore space, which in
14 most of the cases is accompanied by an important reduction in permeability. Thus, compaction
15 bands are also important in reservoir mechanics for oil production and CO₂ storage [*Olsson et al.*,
16 2002; *Holcomb et al.*, 2007; *Rutqvist*, 2012]. Pore collapse, intergranular grain fracturing and
17 sliding, Hertzian cracking at grain contacts, grain crushing, grain attrition, grain-matrix
18 debonding and matrix fracturing are often observed to a less or bigger extent inside the
19 compacting zone [*Baud et al.*, 2004, 2009; *Sternlof et al.*, 2005; *Tondi et al.*, 2006; *Holcomb et*
20 *al.*, 2007; *Aydin and Ahmadov*, 2009; *Zhu et al.*, 2010; *Cilona et al.*, 2012; *Rustichelli et al.*,
21 2012; *Wong and Baud*, 2012]. In parallel, it has been observed that reactive fluids play a
22 significant role in creep and compaction of porous rocks [e.g. *Le Guen et al.*, 2007; *Liteanu and*
23 *Spiers*, 2009; *Rutqvist*, 2012]. Generally, mechanical damage and chemical degradation of the
24 solid skeleton (i.e. of the grains and matrix) are important factors that may trigger instabilities in
25 the form of compaction bands. The objective of the present paper is to explore the possibilities
26 and conditions of compaction band formation due to chemo-mechanical reasons in porous
27 geomaterials.

28
29 In the frame of Continuum Mechanics, compaction bands can be seen as an instability of the
30 underlying mathematical problem. Discrete approaches like the Discrete Element Method have
31 also been proposed in the literature [*Katsman et al.*, 2005; *Katsman and Aharonov*, 2006; *Wang*
32 *et al.*, 2008; *Marketos and Bolton*, 2009, among others] but they are computationally intensive
33 and the identification of the general conditions that lead to strain localization is not
34 straightforward. On the other hand a continuum approach can reveal the conditions for
35 compaction band triggering given the constitutive behavior of the material [*Rudnicki and Rice*,
36 1975; *Vardoulakis and Sulem*, 1995; *Rudnicki*, 2002]. It is commonly observed that strain
37 localization is favored by strain softening and most of the existing theoretical studies focus on
38 pure mechanical reasons for compaction band formation (mechanical softening due to grain and
39 matrix damage and pore collapse). Nevertheless, more recently, the role of chemical softening on

40 slip instabilities has been explored [e.g. *Brantut and Sulem, 2012; Veveakis et al., 2012, 2013*]
41 showing that chemistry may play an important role in shear band instabilities and strain
42 localization. Here we focus on the conditions that lead to the formation of pure compaction bands
43 in quartz- or carbonate-based geomaterials by considering the effect of chemical dissolution.
44 Shear enhanced compaction bands are out of the scope of the present study and their investigation
45 can be a future extension of the proposed model.

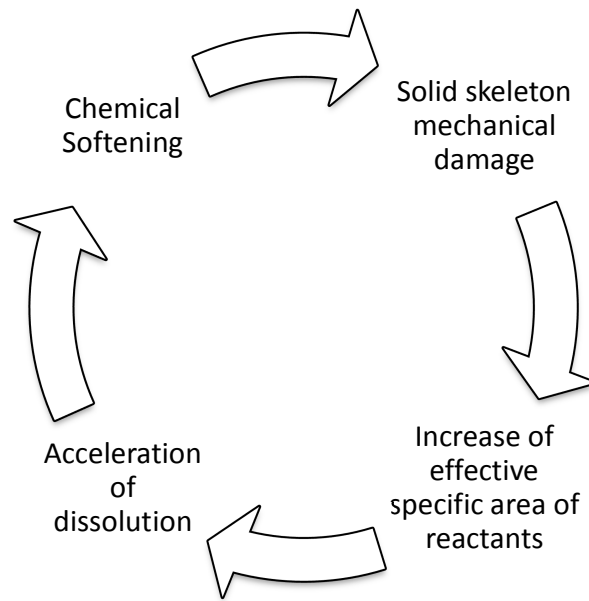
46
47 The effect of chemical dissolution is important in field and in reservoir applications. For instance,
48 the experimental results of Xie et al. [2011] showed that the chemical dissolution of a limestone
49 leads to a significant increase of the porosity (from 23% for the intact rock to 27% for the
50 degraded one). According to the same authors, the plastic pore collapse threshold is also reduced
51 from about 30 to 20 MPa and the chemically degraded materials become more collapsible and
52 more ductile due to the increase in porosity and the degradation of the inter-granular cementation.
53 This evidence is corroborated by other authors [e.g. *Nova et al., 2003; Hu and Hueckel, 2007b;*
54 *Zinsmeister et al., 2013*] for a class of geomaterials and results in a contraction of the elastic
55 domain only due to chemical reasons (chemical softening). In parallel, in a saturated porous
56 geomaterial, the progressive mechanical damage of the solid skeleton during compaction has as a
57 result the increase of the interface area of the reactants (i.e. of the solution with the solid) and
58 consequently the acceleration of the dissolution rate of the solid phase [cf. *Rimstidt and Barnes,*
59 *1980*]. Thus, the solid skeleton is degraded more rapidly (mass removal because of dissolution),
60 the overall mechanical properties of the system diminish (contraction of the elastic domain –
61 chemical softening), deformations increase and the solid skeleton is further damaged
62 (intergranular fractures, debonding, breakage of the porous network etc.). Figure 1 schematically
63 shows this positive feedback process, whose stability is not guaranteed. Actually, as it will be
64 shown in this paper, instabilities in the form of compaction bands may be triggered.

65
66 The impact of chemical phenomena and, in particular, of pressure solution inside deformation
67 bands, has been demonstrated in several cases [*Tondi et al., 2006; Tondi, 2007; Liteanu and*
68 *Spiers, 2009; Cilona et al., 2012; Rustichelli et al., 2012*]. However, the effect of dissolution on
69 compaction band formation is difficult to observe in the field and its exact role is still a subject of
70 investigation. Notice that field observations are made long after the formation of compaction

71 bands, where the poro-mechanical properties of the rock have obviously drastically changed.
72 Exploring the effects of dissolution prior to compaction band formation is also a difficult task in
73 the laboratory as the reproduction of the field conditions is not straightforward. Nevertheless,
74 recent experimental tests with dissolvable surrogate materials and numerical studies with the
75 Discrete Element Method have shown that dissolution of grains causes pronounced changes in
76 the fabric and in the intergranular force transmission that may result in episodic microstructural
77 changes and to strain localization [*Shin, 2009; Shin and Santamarina, 2009; Tran et al., 2012*].
78 The aforementioned indications of chemically induced strain localization instabilities provides
79 the motivation of the present study and justifies the consideration of strong chemo-mechanical
80 couplings in models due to dissolution.

81
82 Two scales are distinguished in the present chemo-hydro-mechanical model. The first scale is
83 related to the macroscopic poromechanical behavior of the geomaterial. At this level the mass
84 balance equation and the stress equilibrium are expressed over the representative elementary
85 volume (REV). The second scale, which will be called here micro-scale, concerns the behavior of
86 a single grain of the geomaterial and its surrounding matrix. Intergranular and matrix fracturing,
87 grain-matrix debonding and the dissolution reaction kinetics refer to this scale. The distinction of
88 the micro- and the macro-scale is presented schematically in Figure 2. The bridging between the
89 macro- and the micro-scale is achieved here through the empirical law of Lade [1996], which
90 relates the effective grain size to the mechanical energy input to the system. In this sense, at least
91 part of the mechanical energy is dissipated through the various micro-mechanisms related to the
92 solid skeleton damage for the creation of new surfaces in the medium (fracturing). The advantage
93 of the aforementioned empirical law over other micromechanical approaches is that it needs the
94 calibration of only one parameter. More sophisticated grain damage models that account for the
95 degradation of the inter-granular cement of rocks might be suitable, but the chosen
96 phenomenological approach is a first step for studying some key features of chemical degradation
97 on compaction banding by avoiding unnecessary complexity. Dissolution reaction kinetics at the
98 micro-level (i.e. at the scale of a single grain) are then up-scaled to the macro-level.

99



100

101 *Figure 1. Positive feedback process due to dissolution and solid skeleton damage (e.g.*
 102 *intergranular fracturing, breakage of the porous network, matrix cracking, grain-matrix*
 103 *debonding etc.)*

104

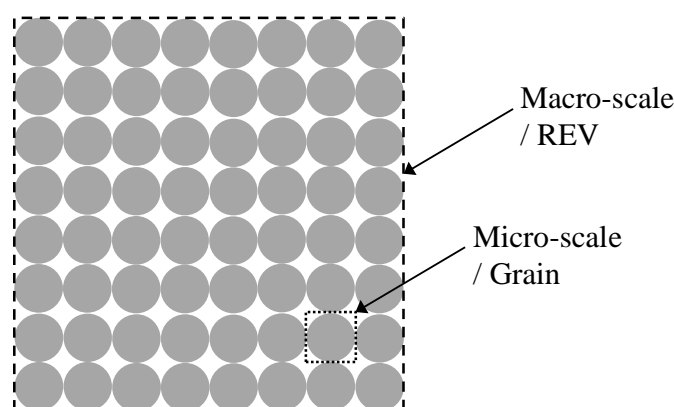
105 As it was already mentioned, the present analysis aims at exploring the possibilities of
 106 compaction band formation due to chemical softening effects only. Therefore, mechanical
 107 softening (or hardening) induced by grain damage or pore collapse is not considered herein in
 108 order to isolate the chemical effects. This means that the effect of grain damage and pore collapse
 109 on the evolution of the yield surface as described for instance by Das et al. [2011] is neglected.
 110 On the other hand the effect of the damage of the solid skeleton on the acceleration of chemical
 111 reaction is taken into account by explicitly introducing the effective specific surface of the grains
 112 in the reaction kinetics law. Of course it is possible to account also for mechanical
 113 softening/hardening due to grain damage and pore collapse but this would somewhat hide the role
 114 of chemical effects explored here.

115

116 The first two sections of the paper are devoted to the formulation of the proposed chemo-poro-
 117 mechanical model by distinguishing the micro- and the macro-level behavior. At the macro-level
 118 (section 2), the constitutive behavior of the material is described within the frame of plasticity
 119 theory, the mass balance equation is derived and the linear momentum balance is set forth in

120 order to account for the different species in the REV. At the micro-level (section 3), the evolution
 121 of the effective grain size due to various micro mechanisms is described and the reaction kinetics
 122 are formulated and upscaled to the macro-level. Next, in section 4, the possibility of compaction
 123 band formation is discussed through a linear stability analysis. A criterion for compaction band
 124 instabilities is proposed and its sensitivity to the various parameters of the model is explored.
 125 Note that because of the strong chemo-poro-mechanical coupling, the application of the classical
 126 localization criterion of Rice [1976] is not straightforward. Finally, in section 5 two examples of
 127 compaction band instabilities are given for a quartzic rock under water flooding conditions and
 128 for a carbonate grainstone in relation with CO₂ injection and storage. All the mathematical
 129 calculations were performed with the symbolic language mathematical package Mathematica©
 130 and they are available to the reader upon request.

131



132

133 *Figure 2. Schematic representation of the REV (macro-scale) and of the grains (micro-scale).*
 134 *The picture is an idealization of the microstructure of a rock. The dissolution rate is*
 135 *homogeneous in the REV when the size and the chemical composition of the grains is*
 136 *homogeneous. In the case of compaction bands, the grains break, their size is not necessarily*
 137 *uniform in the REV and consequently the dissolution rate is not any more homogeneous.*

138

139 **2. Macro-scale**

140 **2.1. Constitutive behavior**

141 Based on experimental research and theoretical considerations, several constitutive laws have
 142 been proposed in the literature that relate the observed macroscopic stresses and strains. Plasticity

143 theory is a standard framework for the mathematical formulation of the underlying mechanical
 144 problem. For a recent review of common plasticity models used for porous rocks we refer to
 145 Wong and Baud [2012].

146
 147 Depending on the applied stress path, the boundary conditions and the geomaterial at hand, a
 148 strain hardening or softening response can be observed due to the various mechanisms, which
 149 take place at the microlevel, i.e. at the grain-scale, and are related to the evolution of the
 150 microstructure of the solid skeleton (e.g. pore collapse) and to grain damage (e.g. grain
 151 fracturing). However, for the reasons exposed in the introduction, only chemical softening will be
 152 considered herein.

153
 154 The removal of minerals from the solid skeleton through chemical processes causes the
 155 mechanical strength of the rock to decrease [e.g. *Hu and Hueckel, 2007a, 2007b*]. Therefore, we
 156 expect the mechanical strength of a rock to be a function of the (residual) mass of the solid

157 skeleton after the dissolution process. Expressing as $\zeta = \zeta(t) = \frac{M_s(t)}{M_s^0}$, $0 \leq \zeta \leq 1$, the ratio of the

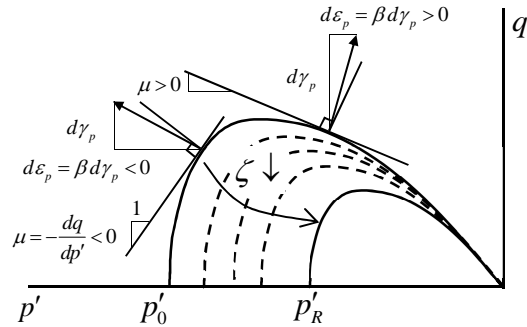
158 current mass, $M_s(t)$, of the constituent 's' over its initial mass, $M_s^0 = M_s(t=0)$, in the REV
 159 before the chemical process starts (initial/reference state), ζ can be seen as a chemical softening
 160 parameter [see also Nova et al., 2003; Hu and Hueckel, 2007b]. Assuming compression negative,
 161 Figure 3. describes qualitatively the strength domain of the geomaterial in the $q - p'$ plane due
 162 to chemical softening, i.e. for decreasing ζ . The scalars p' and q are respectively the Terzaghi
 163 effective mean stress (i.e. the difference between the total mean stress and the pore pressure) and
 164 the shearing stress intensity. The shearing stress intensity q is defined as the square root of the

165 second invariant of the deviatoric part, s_{ij} , of the stress tensor: $q = \sqrt{\frac{1}{2} s_{ij} s_{ji}}$. The Einstein

166 summation convention is adopted. In order to preserve the generality of the approach we consider
 167 a general yield surface of the form:

168
$$f \equiv f(q, p', p'_c) = 0 \quad (1)$$

169 where p'_c is a material parameter (namely the yield stress under isotropic loading) which is
 170 assumed to decrease from p'_0 (initial reference state) to p'_R (residual yield stress once the
 171 chemical reaction is completed) according to the following law $p'_c \equiv p'_R - (p'_R - p'_0)\zeta^\kappa$ (Figure
 172 3). κ is an exponent that can be experimentally determined for the material and the chemical
 173 process at hand.



174
 175 *Figure 3. Chemical isotropic softening due to dissolution of a constituent. ζ describes the ratio*
 176 *of the residual mass of the constituent in the REV over its initial (reference) mass before the*
 177 *dissolution process. Compression is taken negative. $\beta > 0$ corresponds to dilatancy and $\beta < 0$ to*
 178 *contractancy (decrease of porosity).*

179
 180 In the frame of a small strain theory, the strain may be separated into elastic and plastic parts as
 181 follows:

182
$$\varepsilon = \varepsilon^e + \varepsilon^p \quad \text{and} \quad \gamma = \gamma^e + \gamma^p \quad (2)$$

183 where ε denotes the volumetric deformation and γ the shearing strain intensity $\gamma = \sqrt{2e_{ij}e_{ji}}$
 184 with e_{ij} being the deviatoric strain tensor.

185
 186 The elasto-plastic incremental stress-strain relationships are given by:

187
$$\dot{\boldsymbol{\sigma}}' = \mathbf{M}_{ep} \dot{\boldsymbol{\varepsilon}} + \boldsymbol{\Psi} \dot{\zeta} \quad (3)$$

188 where $\boldsymbol{\sigma}' = \begin{pmatrix} p' \\ q \end{pmatrix}$, $\boldsymbol{\varepsilon} = \begin{pmatrix} \varepsilon \\ \gamma \end{pmatrix}$, $\mathbf{M}_{ep} = \left[\mathbf{I} - \frac{\mathbf{M}_e \frac{\partial g}{\partial \boldsymbol{\sigma}} \left(\frac{\partial f}{\partial \boldsymbol{\sigma}} \right)^T}{\left(\frac{\partial f}{\partial \boldsymbol{\sigma}} \right)^T \mathbf{M}_e \frac{\partial g}{\partial \boldsymbol{\sigma}}} \right] \mathbf{M}_e$, $\boldsymbol{\Psi} = -\frac{\mathbf{M}_e \frac{\partial f}{\partial \zeta} \frac{\partial g}{\partial \boldsymbol{\sigma}}}{\left(\frac{\partial f}{\partial \boldsymbol{\sigma}} \right)^T \mathbf{M}_e \frac{\partial g}{\partial \boldsymbol{\sigma}}}$, \mathbf{I} is the identity

189 matrix and $\mathbf{M}_e = \begin{pmatrix} K & 0 \\ 0 & G \end{pmatrix}$ the elasticity matrix. $(\cdot)^T$ denotes the transpose of (\cdot) . K and G are
 190 respectively the elastic bulk and the elastic shear moduli of the geomaterial. g is the plastic
 191 potential which can be identified to the yield surface, f , in case of associate plasticity. For a
 192 general plastic constitutive law, we can write:

193
$$\frac{\partial f}{\partial \boldsymbol{\sigma}} = \begin{pmatrix} \mu \\ 1 \end{pmatrix}, \quad \frac{\partial g}{\partial \boldsymbol{\sigma}} = \begin{pmatrix} \beta \\ 1 \end{pmatrix} \quad (4)$$

194 where β is the dilatancy angle ($\beta > 0$ for dilatant materials) and μ is the internal friction of the
 195 geomaterial that depend on the actual state of stress. Using Eq.(4) we obtain:

196
$$\mathbf{M}_{ep} = \frac{GK}{G + \beta\mu K} \begin{pmatrix} 1 & -\beta \\ -\mu & \beta\mu \end{pmatrix} \quad (5)$$

197 and

198
$$\boldsymbol{\Psi} = \frac{1}{G + \beta\mu K} \begin{pmatrix} \beta K \\ G \end{pmatrix} \frac{\partial f}{\partial \zeta} \quad (6)$$

199 with

200
$$\frac{\partial f}{\partial \zeta} = \frac{\partial f}{\partial p'_c} \frac{dp'_c}{d\zeta} = \frac{\partial f}{\partial p'_c} \kappa (p'_R - p'_0) \zeta^{\kappa-1} \quad (7)$$

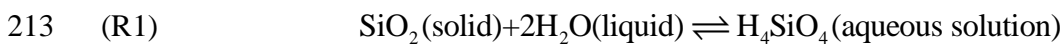
201 It is worth mentioning, that under constant loading, chemical softening would result in the
 202 gradual accumulation of plastic deformations, which are controlled by the dissolution reaction
 203 kinetics. This accumulation of deformations under constant loading can be seen as a creep

204 behavior induced by the chemical process of dissolution. Creep phenomena due to reactive fluids
 205 injection are observed in laboratory tests on porous rocks [*Le Guen et al.*, 2007; *Liteanu and*
 206 *Spiers*, 2009; *Shin and Santamarina*, 2009; *Brantut et al.*, 2013; *Croizet et al.*, 2013].

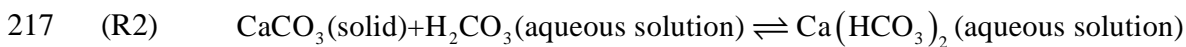
207

208 **2.2. Mass balance**

209 The dissolution of minerals is described by appropriate chemical reactions. According to the
 210 reaction's stoichiometry the minerals of a rock may be degraded and transformed to amorphous
 211 solid or liquid. For example the dissolution/precipitation of quartz in water is described by the
 212 following chemical equation:



214 At equilibrium the silicic acid (H_4SiO_4) is dissolved in the water with a concentration of the
 215 order of few ppm. Another example is the dissolution of calcite with water that is saturated with
 216 carbon dioxide:



218 This equation represents a set of consecutive reactions that take place and are responsible for the
 219 dissolution of carbonate rocks [*Grgic*, 2011].

220 Here we consider any dissolution process of the form:



222 The reaction kinetics of this general equation will be analyzed in the next section.

223

224 At the macro-scale, the REV can be seen as a porous medium. The mass of the solid phase is M_3
 225 (e.g. SiO_2) and the mass of the fluid phase is $M_f = M_1 + M_2$, where M_1 is the mass of the
 226 solvent (e.g. H_2O) and M_2 the mass of the dissolution product (e.g. H_4SiO_4).

227 Following the notation of Coussy [2004] the mass balance equations hold:

228
$$\frac{d^f M_1}{dt} = \frac{d}{dt}(\rho_{f1} n_1 d\Omega_t) = \dot{r}_{1 \rightarrow 2} d\Omega_t \quad (8)$$

229
$$\frac{d^f M_2}{dt} = \frac{d}{dt}(\rho_{f2} n_2 d\Omega_t) = (\dot{r}_{2 \rightarrow 1} + \dot{r}_{2 \rightarrow 3}) d\Omega_t \quad (9)$$

230
$$\frac{d^s M_3}{dt} = \frac{d}{dt}[\rho_s (1-n) d\Omega_t] = \dot{r}_{3 \rightarrow 2} d\Omega_t \quad (10)$$

231 where $\frac{d^\chi}{dt}$ is the particle derivative, ρ_χ is the density and n_χ is the Eulerian porosity referring to
 232 particle χ (f for fluid and s for solid species), $\dot{r}_{\chi \rightarrow \psi}$ represents the rate of mass of χ
 233 transformed to ψ and $d\Omega_t$ is the material volume in the current configuration. By definition:

234
$$\rho_{f1} n_1 + \rho_{f2} n_2 = \rho_f n \quad (11)$$

235 where ρ_f is the density of the solution, which depends on the reactant's concentrations, the
 236 pressure p_f and the temperature. Equations (8) and (9) lead to:

237
$$\frac{d^f}{dt}(\rho_f n d\Omega_t) = \dot{r}_{2 \rightarrow 3} d\Omega_t \quad (12)$$

238 Considering isothermal conditions and constant density for the solid phase, Eqs.(10) and (12)
 239 become:

240
$$-\frac{\partial n}{\partial t} + \nabla_{\mathbf{x}} \mathbf{V}_s - \nabla_{\mathbf{x}} (n \mathbf{V}_s) = -\frac{1}{\rho_s} \dot{r}_{2 \rightarrow 3} \quad (13)$$

241
$$\frac{\partial n}{\partial t} + n \frac{1}{\rho_f} \frac{\partial \rho_f}{\partial p_f} \frac{\partial p_f}{\partial t} + n \frac{1}{\rho_f} \frac{\partial \rho_f}{\partial w_2} \frac{\partial w_2^M}{\partial t} + \frac{1}{\rho_f} \nabla_{\mathbf{x}} (\rho_f) n \mathbf{V}_f + \nabla_{\mathbf{x}} (n \mathbf{V}_f) = \frac{1}{\rho_f} \dot{r}_{2 \rightarrow 3} \quad (14)$$

242 where the Nabla operator refers to the initial configuration (\mathbf{X} is the position vector of the solid
 243 particles in the initial configuration), \mathbf{V}_s and \mathbf{V}_f are respectively the velocities of the solid phase

244 and fluid particles, p_f the fluid pressure and $w_2^M = \frac{M_2}{M_f}$ the average dissolution product to fluid
 245 mass fraction over the REV (macro-scale). Adding Eqs. (13) and (14) and using the chain rule,
 246 the pore pressure diffusion-generation equation is obtained:

$$247 \quad \frac{\partial p_f}{\partial t} = c_{hy} \nabla_{\mathbf{x}}^2 p_f - \frac{1}{\beta^*} \frac{\partial \varepsilon}{\partial t} - c_{p, ch} \frac{\partial w_2}{\partial t} \quad (15)$$

248 where c_{hy} is the hydraulic diffusivity and $\beta^* = n\beta_f$ with $\beta_f = \frac{1}{\rho_f} \frac{\partial \rho_f}{\partial p_f}$ is the compressibility of
 249 the fluid [see also *Ghabezloo and Sulem, 2008*]. For plastic incompressibility of the solid phase
 250 and of the fluid, $\beta^* = \frac{1}{K}$. For the derivation of the pore pressure diffusion-generation equation,
 251 the Darcy law was assumed and the gradient of the density of the fluid was ignored as being
 252 negligible compared to the other terms of the equation. The deformation of the solid skeleton, i.e.
 253 the term $\nabla_{\mathbf{x}} \mathbf{V}_s = \frac{\partial \varepsilon}{\partial t}$, is responsible for the hydro-mechanical coupling. In the right hand side of
 254 the equation the last term expresses the pore pressure change due to dissolution/precipitation
 255 process and $c_{p, ch}$ is the chemical pressurization coefficient, which depends on the evolution of
 256 the fluid density because of the reaction process. Herein, we neglect this effect as it does not
 257 affect the stability of the system and generally its influence is limited [*Stefanou and Sulem,*
 258 *2013*]. Finally, for small strains $\nabla_{\mathbf{x}} \approx \nabla_{\mathbf{x}}$, where \mathbf{x} is the position vector of the solid particles in
 259 the current configuration.

260

261 **2.3. Momentum balance under oedometric conditions**

262 Under oedometric conditions the stress equilibrium is expressed as follows:

$$263 \quad \sigma_{zz,z} = 0 \quad (16)$$

264 where $\sigma_{ij} = \sigma'_{ij} - p_f \delta_{ij}$, δ_{ij} is the Kronecker delta, $\sigma_{zz} = p - \frac{2}{\sqrt{3}} q$, $\gamma = -\frac{2}{\sqrt{3}} \varepsilon_{zz}$, $\varepsilon = \varepsilon_{zz}$,

265 $\varepsilon_{zz} = u_{z,z}$ $\varepsilon_{rr} = \varepsilon_{\theta\theta} = 0$ (for axisymmetric conditions). Using Eq.(3) we obtain:

266
$$\dot{\sigma}'_{zz} = c \left(\Lambda \dot{\epsilon}_{zz} + \frac{\partial f}{\partial \zeta} \dot{\zeta} \right) \quad (17)$$

267 where $\Lambda = K \frac{\left(1 + \frac{2}{\sqrt{3}}\beta\right)\left(1 + \frac{2}{\sqrt{3}}\mu\right)}{\frac{2}{\sqrt{3}} - \beta\frac{K}{G}}$ and $c = \frac{\frac{2}{\sqrt{3}} - \beta\frac{K}{G}}{1 + \beta\mu\frac{K}{G}}$. $c\Lambda$ expresses the change of the

268 effective vertical stress due to compaction and $c\frac{\partial f}{\partial \zeta}$ its change due to chemical degradation.

269 3. Micro-scale

270 3.1. Cataclasis, solid skeleton damage and evolution of the effective grain size

271 A rock can be seen as an assemblage of bonded grains, which during deformation may break into
 272 smaller grains with rupture of bonds. Following Lade et al. [1996] the grain size reduction is
 273 related to the total energy input. In this sense, at least part of the mechanical energy is dissipated
 274 through various micro-mechanisms for the creation of new surfaces in the medium (grain
 275 fracturing). “Hertzian cracks” at the grain contacts (intergranular fracturing, grain crushing, grain
 276 attrition etc.), “microcracking” of the cementitious matrix [Aydin and Ahmadov, 2009; Cilona et
 277 al., 2012] and grain debonding [Castellanza and Nova, 2004] are some important mechanisms
 278 that lead to the reduction of the effective grain size of the geomaterial and consequently to the
 279 increase of the dissolution front.

280
 281 Under this framework, it is convenient to define an effective grain size, D , whose evolution
 282 reflects the effect of grain damage. According to Lade et al. [1996], for a given grain size
 283 distribution, this effective grain size is commonly taken equal to D_{10} (10% of the system weight
 284 consists of grains below D_{10} diameter). Generally, the choice of the appropriate effective grain
 285 size depends on the exact grain size distribution and the material at hand. For instance in the case
 286 of shear bands created in the laboratory D_{10} seems a good measure for the effective grain size
 287 reduction [El Bied et al., 2002]. On the other hand, in the case of compaction bands observed in
 288 the field, D_{50} seems a better choice [Cilona et al., 2012].

289

290 Lade et al. [1996] proposed the following expression for the evolution of grain crushing of a
 291 granular material in terms of the mechanical work input:

$$292 \quad D = D_0 \left(\frac{a}{a + E_T} \right) \quad (18)$$

293 where D_0 is the effective grain size of the initial gradation and E_T is the total energy input per
 294 specimen unit volume (including the energy due to isotropic compression and shearing). a is a
 295 fitting parameter, which will be called here grain damage sensitivity parameter that expresses the
 296 extent of the grain size change due to energy input. For large values of parameter a , as compared
 297 to E_T , the grain size is constant. As discussed by Lade et al. [1996], the total energy seems to be a
 298 more relevant parameter for describing grain breakage compared to the effective mean stress or
 299 the void ratio. As an example, for Cambria sand $a = 0.747\text{MPa}$ [Lade et al., 1996]. For a rock,
 300 the parameter a should also depend on the grain size distribution [cf. Cheung et al., 2012].
 301 Considering that the specific effective surface of a grain S (effective grain surface area to
 302 volume ratio) is inversely proportional to the grain diameter, it is natural to assume the same type
 303 of relationship:

$$304 \quad S = S_0 \left(1 + \frac{E_T}{a} \right) \quad (19)$$

305 It is worth emphasizing that the grains of rocks are often cemented together and that the
 306 consideration of the rock as a granular material, with well-defined grain to pore fluid interactions
 307 is rather an idealization of reality. Nevertheless, as it was mentioned above, the dissipation during
 308 mechanical loading is mainly attributed to internal friction mechanisms (reorganization of grains,
 309 grain dislocation/disclination) and microcracking of both the grains and of the cementitious
 310 matrix (Hertzian cracking, microcracking, debonding etc.). In this sense, Eq.(19), could also be
 311 used in order to describe the increase of the specific surface of the grains of a cemented granular
 312 material like a porous rock. Parameter a may be calibrated either through appropriate
 313 micromechanical models that take into account the micro-cracking of the matrix and grain
 314 breakage [e.g. Das et al., 2011] or through adequate experimental tests. More specifically, the
 315 value of the grain damage parameter may be determined experimentally through acoustic

316 emissions, X-ray tomography or other experimental techniques [Wong and Baud, 2012] that will
 317 correlate the effective grain size due to grain damage and the mechanical energy input.

318
 319 Finally, it should be mentioned that grain crushing results in a reduction of porosity and generally
 320 to a reduction of permeability [Walsh and Brace, 1984; Sulem and Ouffroukh, 2006]. Moreover,
 321 due to dissolution, the specific area of the grains at the microscale is increased and the existing
 322 microcracks propagate further (subcritical growth, stress corrosion cracking). Therefore, the grain
 323 crushing sensitivity parameter, a , is not necessarily constant in time and may depend on the
 324 reaction kinetics, the stress level and the nature of the saturating fluid. However, in the linear
 325 stability analysis presented in the next section, the permeability change due to porosity reduction
 326 is of second order and can be neglected for the onset of strain localization. Nevertheless, for
 327 performing numerical simulations of rocks with the proposed chemo-poro-mechanical model it is
 328 possible to determine the evolution of these parameters based on empirical and/or theoretical
 329 models.

330

331 **3.2. Reaction kinetics**

332 According to Rimstidt and Barnes [1980] the rate of reaction between the fluid and solid phase
 333 during quartz dissolution (or precipitation, R1) is directly proportional to the interfacial area
 334 between the solid and the liquid phase. The dissolution rate is commonly assumed to be
 335 proportional to the mineral surface area exposed to the aqueous solution [Cubillas *et al.*, 2005].
 336 Here we generalize this idea for any dissolution process of the form of R3. The rate of production
 337 (or consumption) of moles of the dissolution product is considered to obey the following rate
 338 equation:

$$339 \quad \frac{\partial n_2}{\partial t} = Ak(1 - \Omega) \quad (20)$$

340 where n_2 is the number of moles of the product of the dissolution process (e.g. the H_4SiO_4 or the
 341 $\text{Ca}(\text{HCO}_3)_2$), A the available interfacial area of the solid with the solution, k the dissolution
 342 rate constant per unit area of the interface and Ω is the degree of saturation. If $\Omega = 1$ the reaction

343 is at equilibrium, if $\Omega < 1$ dissolution takes place and if $\Omega > 1$ precipitation happens. Ω is
 344 expressed in terms of the reaction quotient, Q , and the equilibrium constant, K_{eq} , as follows:

$$345 \quad \Omega = \frac{Q}{K_{eq}} \quad (21)$$

346 It is worth mentioning that in order to account for the intergranular dissolution-diffusion micro-
 347 mechanisms that take place in the thin aqueous film that is developed at the grain contacts, a
 348 chemical potential that depends on the stress state is often considered (pressure solution [Rutter,
 349 1983]). As a result, the reaction kinetics (i.e. Eq. (20), equilibrium constant, reaction rate) might
 350 be functions of the stress state. However, intense cataclasis (stress induced cracking, grain
 351 breakage, microcracking, debonding etc.) is often observed inside the zone of deformation bands
 352 [Baud *et al.*, 2009; Cilona *et al.*, 2012]. Therefore, in the stability analysis performed in the next
 353 section, the reaction kinetics will not directly depend on the stress state, but will be a function of
 354 the available interaction surface of the reactants (dissolution of grains), which depends on the
 355 stress state because of grain damage.

356
 357 Assuming that the reaction activity coefficients for the reactants are close to unity and that
 358 $w_2 \ll 1$, Eq.(20) and (21) give (Appendix A.1):

$$359 \quad \frac{\partial w_2}{\partial t} = k^* \frac{S}{e} \left(1 - \frac{w_2}{w_2^{eq}} \right) \quad (22)$$

360 where $k^* = \frac{\mu_2}{\rho_f} k$, $e = \frac{n}{1-n}$ is the void ratio and w_2^{eq} the mass fraction of dissolution product to
 361 the fluid mass at chemical equilibrium. It should be emphasized that Eq.(22) is written at the
 362 microscale and that w_2 , S and e represent local quantities, which are not necessarily
 363 homogeneous over the REV (Figure 2). On the other hand, the constitutive law and the mass
 364 balance represent quantities defined over the REV and therefore the aforementioned local
 365 quantities have to be upscaled from the micro- to the macro-level.

366

367 **3.3. Chemical softening and upscaling**

368 At the macroscale the rate of the chemical softening parameter ζ holds for $w_2 \ll 1$ (Appendix
369 A.2):

$$370 \quad \frac{\partial \zeta}{\partial t} = -\frac{\mu_3 \rho_f}{\mu_2 \rho_s} e^\zeta \frac{\partial w_2^M}{\partial t} \quad (23)$$

371 The average mass fraction of the dissolution product over the REV (macro-level) is related to the
372 local mass fraction w_2 (Figure 2) as follows:

$$373 \quad w_2^M = \frac{1}{V_T} \int_{V_T} w_2 dV \quad (24)$$

374 Assuming that $w_2 = w_2(z, t)$ (oedometric conditions) is a function that can be expanded into
375 Taylor series up to the second order in z :

$$376 \quad w_2(z_l, t) \approx w_2(z, t) + z_l \left. \frac{\partial w_2(z, t)}{\partial z} \right|_z + \frac{1}{2} z_l^2 \left. \frac{\partial^2 w_2(z, t)}{\partial z^2} \right|_z \quad (25)$$

377 where z_l is the coordinate in a local coordinate system with origins at the center of the REV, z ,
378 injecting equation (25) to equation (24) and integrating over the REV we obtain:

$$379 \quad w_2^M \approx w_2 + \ell_c^2 \frac{\partial^2 w_2}{\partial z^2} \quad (26)$$

380 where $\ell_c = \ell_{REV} \sqrt{\frac{1}{24}} \approx \frac{\ell_{REV}}{5}$ appears as a characteristic internal length and ℓ_{REV} is the size of the
381 REV in the z direction. Note that if the mass fraction is homogeneous all over the REV, then
382 $w_2^M = w_2$.

383 **4. Compaction band formation**

384 Equations (15), (16) and (22) describe the evolution of the system under oedometric conditions,
385 while equation (26) links the macro- and micro-scale. Introducing the dimensionless quantities:

$$\begin{aligned}
\hat{w}_2^{(M)} &= \frac{w_2^{(M)}}{K_{eq}}, \quad \hat{p}_f = \frac{p_f}{\sigma_n}, \quad \hat{\sigma}_{ij} = \frac{\sigma_{ij}}{\sigma_n}, \quad \hat{p} = \frac{p}{\sigma_n}, \quad \hat{q} = \frac{q}{\sigma_n}, \quad \hat{E}_T = \frac{E_T}{\sigma_n}, \quad \hat{a} = \frac{a}{\sigma_n}, \quad \hat{\beta}^* = \beta^* \sigma_n \\
\hat{u}_i &= \frac{u_i}{D_0^{50}}, \quad \hat{L} = \frac{L}{D_0^{50}}, \quad \hat{S} = S D_0^{50}, \quad \hat{\rho}_\chi = \frac{\rho_\chi}{\rho_{H_2O}}, \quad \hat{t} = \frac{t}{T}, \quad \hat{c}_{hy} = \frac{T}{(D_0^{50})^2} c_{hy}
\end{aligned}
\tag{27}$$

with D_0^{50} the median grain diameter in the reference/initial state and choosing $T = \frac{K_2^{eq} D_0^{50}}{k^*}$ as characteristic time, the aforementioned equations become:

$$\hat{\sigma}_{zz,\hat{z}} = 0 \tag{28}$$

$$\frac{\partial \hat{p}_f}{\partial \hat{t}} = \hat{c}_{hy} \frac{\partial^2 \hat{p}_f}{\partial \hat{z}^2} - \frac{1}{\hat{\beta}^*} \frac{\partial \varepsilon}{\partial \hat{t}} - \hat{c}_{p, ch} \frac{\partial \hat{w}_2^M}{\partial \hat{t}} \tag{29}$$

$$\frac{\partial \hat{w}_2}{\partial \hat{t}} = e^{-1} \hat{S} (1 - \hat{w}_2) \tag{30}$$

$$\hat{w}_2^M \approx \hat{w}_2 + \hat{\ell}_c^2 \frac{\partial^2 \hat{w}_2}{\partial \hat{z}^2} \tag{31}$$

In the following the hats over the variables will be dropped in order to simplify the notations.

It has to be mentioned that the application of the well-known localization (bifurcation) criterion of Issen & Rudnicki [Rudnicki and Rice, 1975; see also Rice, 1976; Bigoni and Hueckel, 1991; Issen and Rudnicki, 2000] is not straightforward in the present case where a chemo-poro-mechanical coupling takes place. For this reason a linear stability analysis is performed in the next section.

4.1. Linear stability analysis

Let u_z^h , p_f^h and w_2^h be the solutions of the above equations that lead to homogeneous deformation in space and $\tilde{u}_z(z, t)$, $\tilde{p}_f(z, t)$ and $\tilde{w}_2(z, t)$ perturbations such that:

405

$$\begin{aligned}
 u_z(z,t) &= u_z^h + \tilde{u}_z(z,t) \\
 p_f(z,t) &= p_f^h + \tilde{p}_f(z,t) \\
 w_2(z,t) &= w_2^h + \tilde{w}_2(z,t)
 \end{aligned}
 \tag{32}$$

406 The spatial dependence of the perturbations is decomposed into Fourier modes with wavelength
 407 λ . Assuming zero fluid flux and no reaction evolution at the boundaries of the model:

408

$$\tilde{u}_z(z,t) = Ue^{st} \sin\left(\frac{z}{\ell}\right), \quad \tilde{p}_f(z,t) = Pe^{st} \cos\left(\frac{z}{\ell}\right) \quad \text{and} \quad \tilde{w}_2(z,t) = We^{st} \cos\left(\frac{z}{\ell}\right)
 \tag{33}$$

409 where $\lambda = 2\pi\ell = L/N$, N is equal to an integer satisfying the zero fluid flux and reaction
 410 evolution at the boundary $z = \pm L/2$, and s is the growth coefficient of the perturbation
 411 (Lyapunov exponent). $s > 0$ means local instability.

412
 413 Introducing the above perturbations to Eqs.(28)-(30) and neglecting the higher order terms we
 414 obtain the system:

415

$$\tilde{\sigma}_{zz,z} = 0
 \tag{34}$$

416

$$\frac{\partial \tilde{p}_f}{\partial t} = c_{hy} \frac{\partial^2 \tilde{p}_f}{\partial z^2} - \frac{1}{\beta^*} \frac{\partial \tilde{\varepsilon}}{\partial t}
 \tag{35}$$

417

$$\frac{\partial \tilde{w}_2}{\partial t} = \frac{1-w_2^h}{e^h} \tilde{S} - \frac{S^h}{e^h} \left[1 + \eta^h (1-w_2^h) \right] \tilde{w}_2
 \tag{36}$$

418

$$\tilde{w}_2^M \approx \tilde{w}_2 + \ell_c^2 \frac{\partial^2 \tilde{w}_2}{\partial z^2}
 \tag{37}$$

419 where $\eta^h = \frac{\mu_3}{\mu_2} \left(1 + e^h \frac{\rho_f}{\rho_s} \right)$. The superscript 'h' denotes the homogeneous state of the system. \tilde{S}
 420 represents the perturbation of the specific surface because of the perturbations introduced in
 421 Eq.(32) and because of Eq.(19). All the above quantities are dimensionless (the hats were
 422 dropped for simplicity).

423

424 Substituting Eqs.(33) in the above equations, we obtain the following system of equations in
425 matrix form:

$$426 \quad \mathbf{A} \cdot \mathbf{X} = \mathbf{0} \quad (38)$$

$$427 \quad \text{where } \mathbf{A} = \begin{pmatrix} \frac{A_{11}}{\ell^2} & -\frac{1}{\ell} & \frac{A_{13}}{\ell} \left(1 - \frac{\ell_c^2}{\ell^2}\right) \\ -\frac{s}{\ell} \frac{1}{\beta^*} & s + \frac{c_{hy}}{\ell^2} & 0 \\ -\frac{A_{32} S^h}{\ell} & 0 & s + S^h \varphi \end{pmatrix},$$

$$428 \quad A_{11} = -K \frac{\left(\frac{2\beta}{\sqrt{3}} + 1\right) \left(\frac{2\mu}{\sqrt{3}} + 1\right)}{1 + \frac{K}{G} \beta \mu}, \quad A_{13} = -\left(\frac{2}{\sqrt{3}} - \frac{K}{G} \beta\right) \frac{\partial f}{\partial \zeta} \zeta^h e^h \frac{\rho_f \mu_3}{\rho_s \mu_2}, \quad A_{32} = \frac{1 - w^h}{ae^h},$$

$$429 \quad \varphi = \frac{1}{e^h} [1 + \eta^h (1 - w^h)] \quad \text{and} \quad \mathbf{X} = \begin{pmatrix} U \\ P \\ W \end{pmatrix}.$$

430

431 The system has non trivial solutions when the determinant of \mathbf{A} is zero, i.e. $Det[\mathbf{A}] = 0$, or
432 equivalently:

$$433 \quad s^2 + C_1 s + C_0 = 0 \quad (39)$$

$$434 \quad \text{with } C_0 = \frac{S^h \beta^* c_{hy}}{\ell^2} \frac{\varphi A_{11} + A_{13} A_{32} \left(1 - \frac{\ell_c^2}{\ell^2}\right)}{A_{11} \beta^* - 1} \quad \text{and}$$

$$435 \quad C_1 = S^h \varphi + \frac{A_{11} c_{hy}}{A_{11} \beta^* - 1} \frac{1}{\ell^2} + S^h \frac{A_{13} A_{32}}{A_{11} \beta^* - 1} \left(1 - \frac{\ell_c^2}{\ell^2}\right).$$

436

437 If the real part of a root of the above quadratic equation (characteristic polynomial) is positive
438 then the system is unstable.

439

440 **4.2. Conditions for instability**441 The roots s_1 and s_2 of the above quadratic equation satisfy:

442
$$s_1 s_2 = C_0 \text{ and } s_1 + s_2 = -C_1 \quad (40)$$

443 Therefore if $C_0 < 0$ then at least one root with positive real part exists. For $A_{11} < 0$ and $w^s < 1$ 444 (start of the reaction), $C_0 < 0$ gives:

445
$$\varphi A_{11} + A_{13} A_{32} \left(1 - \frac{\ell_c^2}{\ell^2} \right) > 0 \quad (41)$$

446 or

447
$$\frac{\partial f}{\partial \zeta} \zeta^h \left(1 - \frac{\ell_c^2}{\ell^2} \right) < C_{cr} \quad (42)$$

448 where $C_{cr} = -\frac{\Lambda \rho_s \mu_2}{\Xi \rho_f \mu_3}$ and $\Xi = \frac{1 - w^h}{\varphi a}$. This expression accounts for chemical softening due to449 dissolution and includes the characteristic length of the REV. When dissolution occurs, $w^h < 1$ 450 and $\Xi > 0$. Moreover, in compression, $d\varepsilon_{zz}^p = d\varepsilon^p < 0$ and therefore $\beta < 0$. Consequently, Λ is451 positive and C_{cr} is negative. Λ expresses the increase of the effective compressive stress452 because of compaction (see Eq.(17)). $\frac{\partial f(\zeta)}{\partial \zeta} < 0$ expresses the contraction of the elastic domain453 (chemical softening) because of dissolution. Ξ represents the effect of dissolution on the stability454 of the system due to grain damage. The bigger the value of Ξ is (i.e., high effect of dissolution

455 due to grain damage) the more unstable the system becomes, as the instability condition (42) is

456 satisfied for a larger spectrum of μ and β values (the instability region in the $q-p$ plane is457 larger). The role of Ξ will be explored in the next paragraph458 In the special case where $C_{cr} = 0$ ($\mu = -\frac{\sqrt{3}}{2}$ or $\beta = -\frac{\sqrt{3}}{2}$ leading to $\Lambda = 0$), Eq. (42) becomes:

459
$$\frac{\partial f}{\partial \zeta} \left(1 - \frac{\ell_c^2}{\ell^2} \right) < 0 \quad (43)$$

460 and the system is unstable if $\ell > \ell_c$. In other words, the instability travels in space with a finite
 461 wavelength ($\lambda > 2\pi\ell_c \approx 1.2\ell_{REV}$) resulting in a compaction band of finite thickness. This will be
 462 further investigated in the next paragraphs. Notice that in the absence of mechanical softening
 463 and internal length ($\ell_c = 0$) the above condition for compaction band instabilities ($\mu = -\frac{\sqrt{3}}{2}$ or
 464 $\beta = -\frac{\sqrt{3}}{2}$) coincides with the condition derived by Issen & Rudnicki [2000].

465
 466 Finally, the above sufficient condition for instability should be completed with the following
 467 condition in order to avoid the consideration of flutter instabilities, which are characterized by the
 468 fact that the roots of the characteristic polynomial have an imaginary part:

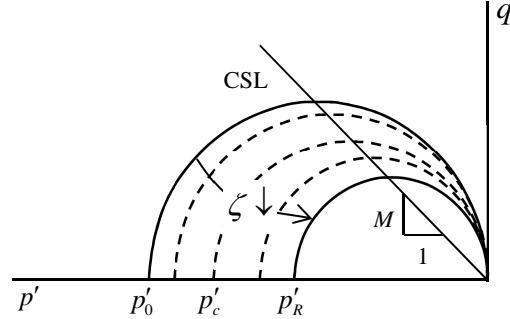
469
$$C_1^2 - 4C_0 \geq 0 \quad (44)$$

470 The reason for excluding this kind of oscillatory instabilities (blowing-up oscillations) is that they
 471 seem unphysical for the chemo-mechanical problem at hand.

472
 473 **4.3. Regions of stability and parameters sensitivity**

474 As an example of a cap yield surface we consider a modified Cam-clay yield surface with an
 475 associate flow rule. Certainly, modified Cam-clay with associate flow rule is not the best
 476 constitutive model for rocks and in order to obtain quantitative results an extensive experimental
 477 research has to be conducted. Nevertheless, it is expected, that a general cap model, (Figure 3)
 478 will be overall appropriate [Baud *et al.*, 2006; Wong and Baud, 2012]. Herein, we choose the
 479 modified Cam-clay plasticity model with associate flow rule due to its relatively simple form (i.e.
 480 elliptic yield cap). In particular the adopted yield surface is (Figure 4):

481
$$f \equiv q^2 + M^2 p'(p' - p'_c) = 0 \quad (45)$$



483 *Figure 4. Modified Cam-clay model with chemical softening. Compression negative. CSL stands*
 484 *for the critical state line, which is assumed constant.*

485 In this way the qualitative behavior of the system may be studied pointing out the effects of
 486 chemical softening to compaction band formation. For the example at hand we consider the
 487 following dimensionless parameters: $p'_0 = -1$, $p'_R = -0.2$, $p_f = 0.4$, $M = 1$, $\kappa = 1$, $K = G = 100$

488 , $S = 1$, $c_{hy} = 6 \times 10^7$, $\frac{\mu_3}{\mu_2} = 0.6$, $\frac{\rho_f}{\rho_s} = 0.4$. In this paragraph we assume that $\ell_c = 0$, or in other

489 words we consider that the rock microstructure is homogeneous (see section 3.3). This
 490 assumption does not influence the conditions for the onset of the compaction band formation (see
 491 Eqs. (42) or (43)), which is the purpose of the current paragraph. In the next paragraph the
 492 heterogeneity of the REV will be considered and its influence to compaction bands
 493 thickness/periodicity will be explored.

494
 495 According to Eq.(42) compaction band triggering depends on Ξ , which represents the tendency
 496 of the system for compaction band formation. With higher values of Ξ , the region of instability
 497 in $q-p$ plane becomes larger. Figure 5 and Figure 6 show the dependency of Ξ on the grain
 498 fracturing coefficient a for $n = 0.25$ and on the porosity n for $a = 0.1$ respectively. Notice that
 499 when the solution is saturated, $w_2^h = 1$ (i.e. the dissolution process stops), compaction bands can

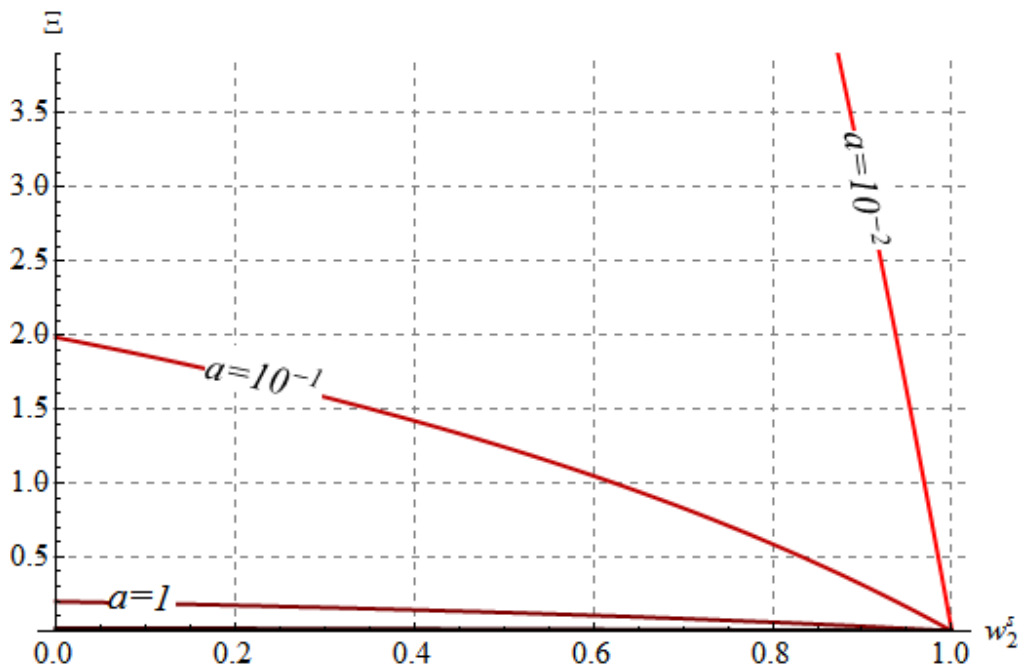
500 be triggered only if $\beta = -\frac{\sqrt{3}}{2}$ or $\mu = -\frac{\sqrt{3}}{2}$. This is consistent with the Issen and Rudnicki

501 condition for compaction bands [Issen and Rudnicki, 2000]. The same holds when no grain
 502 damage occurs ($a \rightarrow \infty$) or when the medium is not porous ($n \rightarrow 0$), as no reaction takes place

503 and chemical softening is impossible. According to Figure 6, in the case of layered materials,
 504 compaction bands would preferentially develop in the more porous layers.

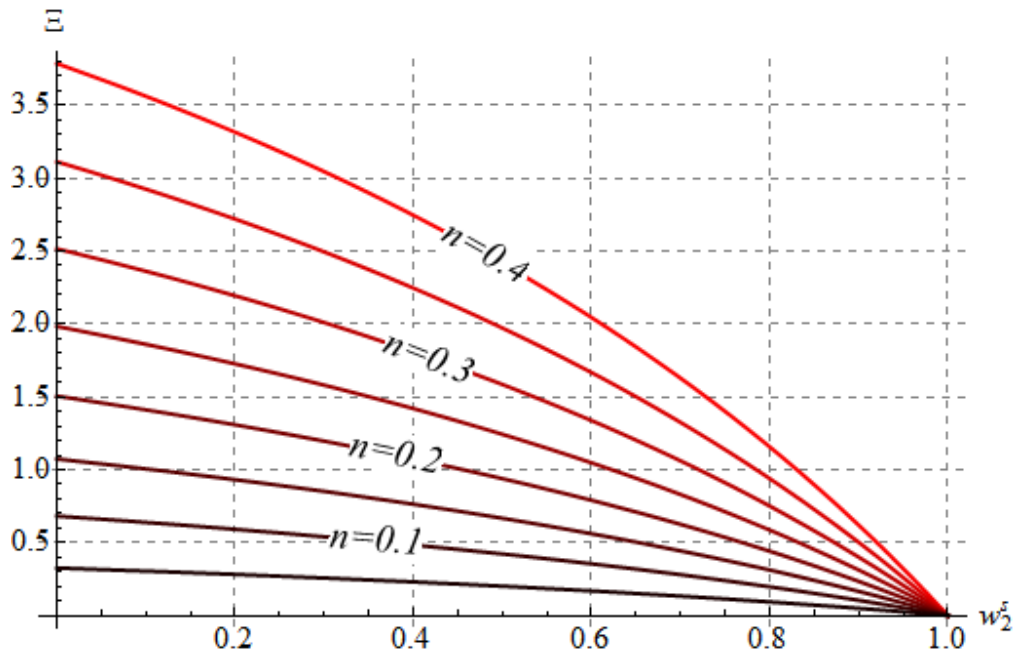
505
 506 Figure 7, Figure 8 and Figure 9 show the region of instability in the $q-p$ plane for various
 507 values of the grain damage coefficient, a , for $w_2^h = 0$ and for $n = 0.25$. If at a certain time the
 508 stress state in the $q-p$ plane is inside this instability region then compaction bands instabilities are
 509 triggered. Notice that because of the strong chemo-mechanical coupling of the proposed model,
 510 the instability manifold is larger than the instability manifold defined by the Issen & Rudnicki
 511 [2000] compaction band criterion (see Figure 7 to Figure 9) and its size depends on the grain
 512 damage parameter a . Moreover, compaction bands can be triggered even for $\beta + \mu > -\sqrt{3}$.
 513 Figure 10 shows the growth coefficient s as a function of the wavelength of the perturbation for
 514 $a = 10^{-2}$, $w_2^h = 0$, $n = 0.25$, $p = 1.2$, $q = 0.4$ and $\zeta = 1$. s is positive and the system is unstable.

515



516

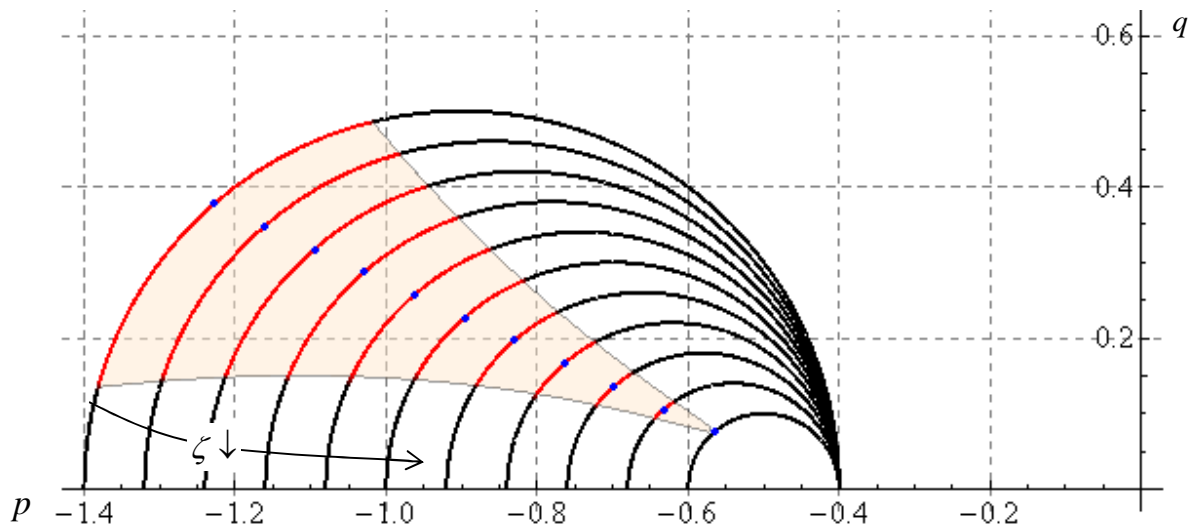
517 *Figure 5. Tendency for compaction bands for $n = 0.25$. The more crushable the grains are, i.e.*
 518 *a small, the bigger the coefficient Ξ and the larger the region of instability is in $q-p$ plane.*



519

520 Figure 6. Tendency for compaction bands for $a = 0.1$. The more porous the geomaterial is, the
 521 bigger the coefficient Ξ and the larger the region of instability in $q - p$ plane are.

522

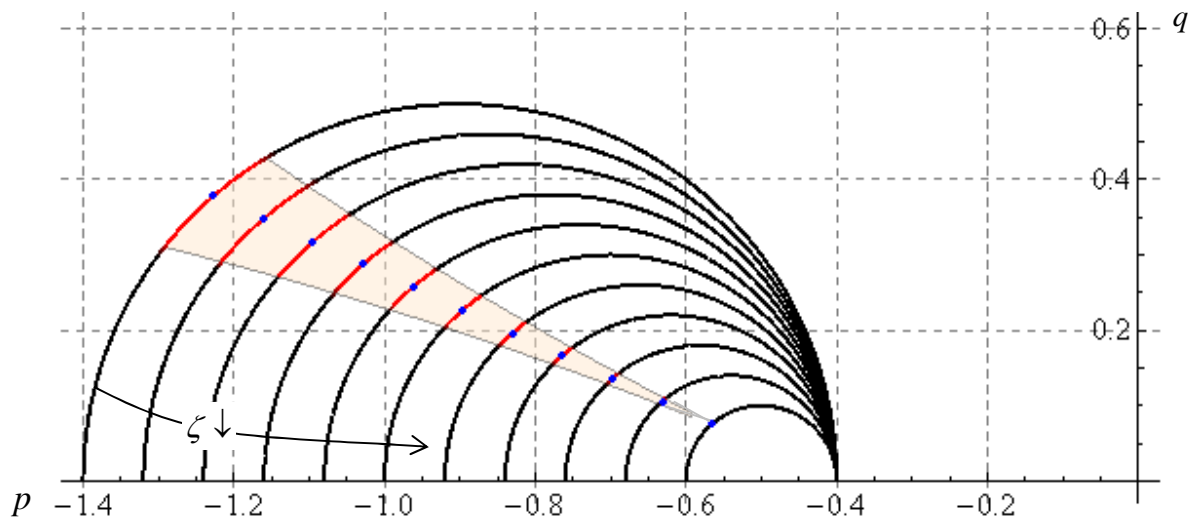


523

524 Figure 7. Instability region (shaded) for compaction bands under oedometric conditions for
 525 $a = 10^{-3}$ ($\Xi = 200$). The points (in blue) represent the Issen & Rudnicki criterion for compaction
 526 band instabilities. Different yield surfaces are drawn for different levels of chemical degradation
 527 ζ .

528

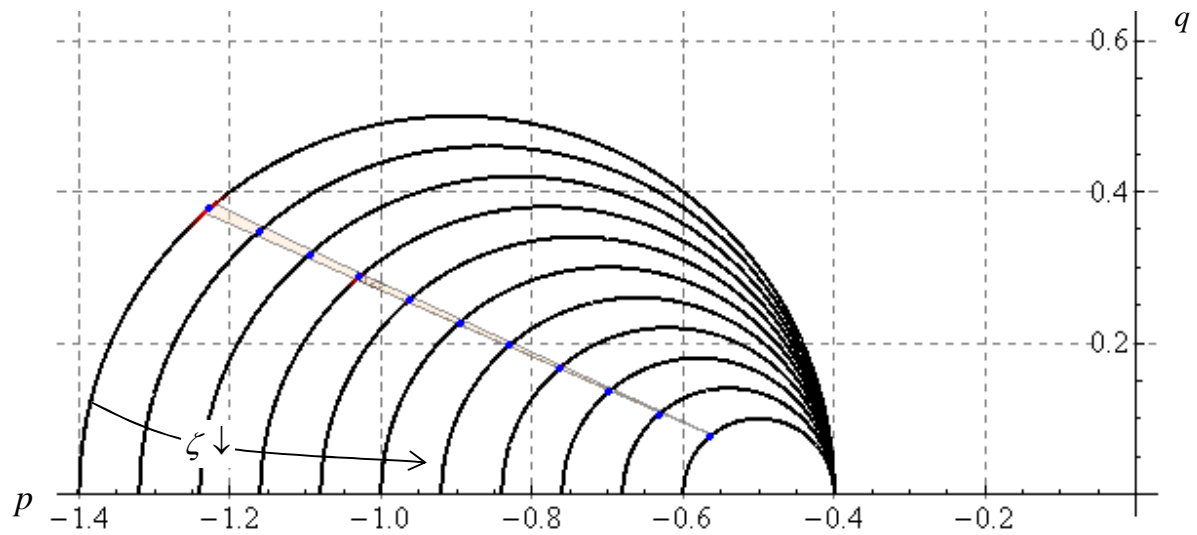
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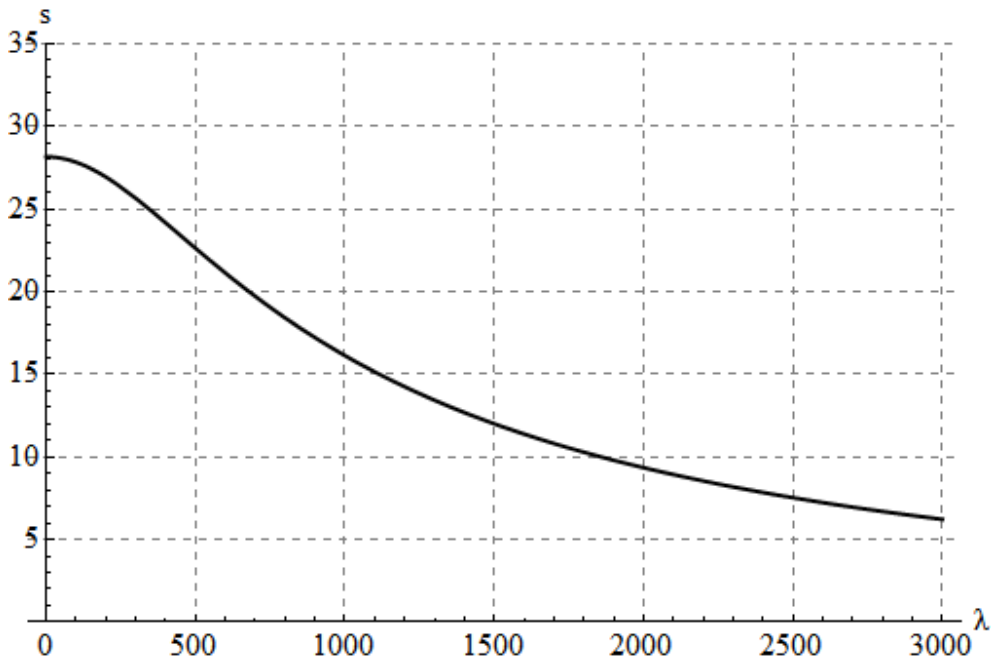
531 *Figure 8. Instability region (shaded) for compaction bands under oedometric conditions for*
 532 *$a = 10^{-2}$ ($\Xi = 20$). The points (in blue) represent the Issen & Rudnitsky criterion for*
 533 *compaction band instabilities. Different yield surfaces are drawn for different levels of chemical*
 534 *degradation ζ .*

535



536

537 *Figure 9. Instability region (shaded) for compaction bands under oedometric conditions for $a = 1$*
 538 *($\Xi = 2$). The points (in blue) represent the Issen & Rudnitsky criterion for compaction bands.*
 539 *Different yield surfaces are drawn for different levels of chemical degradation ζ .*



540

541 *Figure 10. Instability growth coefficient s in terms of the perturbation wavelength λ for $\ell_c = 0$.*
 542 *The growth coefficient is positive and the system is unstable.*

543

544 **4.4. Wave length selection**

545 Figure 10 shows that the perturbation that travels fastest (maximum growth coefficient -
 546 dominant wave length) has zero wavelength. In other words, the dominant wavelength, λ_{\max} , is
 547 zero. This means that the deformation would be localized in a zone of an infinitesimal thickness
 548 as time increases (see Eq.(33)) or equivalently that the compaction bands would be
 549 infinitesimally close to each other ($spacing = \lambda_{\max} = L_b/N = 0$ and so for given $L = L_b$, $N \rightarrow \infty$).
 550 However, experimental evidence shows that this is hardly the case. Due to the heterogeneous
 551 microstructure of geomaterials, compaction is localized to a thin band of finite thickness (several
 552 grains). Consequently, the evolution of the reaction cannot be homogeneous over the REV (
 553 $\ell_c \neq 0$, Eq.(26)). For example, assuming, that the REV has a size of $\ell_{REV} = 20$ (twenty grains),
 554 then $\ell_c \simeq 4$. The size of the REV is rather a statistical quantity and depends on the material at
 555 hand. For the aforementioned characteristic length, the linear stability analysis shows that the
 556 perturbation that travels fastest has a wave length equal to approximately $\lambda_{\max} = 160$ grains

557 (Figure 11) leading to a thickness of 80 grains for the compaction band ($\frac{\lambda_{\max}}{2} = 80D_0^{50}$) or
 558 equivalently to a finite minimum compaction band spacing ($spacing = \lambda_{\max} = L_b/N$ for given
 559 $L = L_b$). The derived compaction band thickness refers to the compressive part of the
 560 perturbation which is half of the wavelength. For $D_0^{50} = 0.1\text{mm}$ the thickness of the band is 8
 561 mm.

562
 563 In the literature, field and laboratory observations show that the thickness of deformation bands is
 564 variable. Tondi [2007] presents the example of a quite thick compactive shear band of the order
 565 of 300 grains. On the contrary, in Baud et al. [2004], the thickness of the compaction band
 566 observed is of the order of two grains. In the present analysis the compaction band thickness
 567 depends on the chemo-mechanical properties of the material and of the heterogeneity of the
 568 microstructure. However, it should be mentioned that the present model should not be considered
 569 as predictive and that the calculated compaction band thickness has only a qualitative character.
 570 In particular, the derived compaction band thickness corresponds to the thickness of the
 571 compacting zone at the onset of the localization. This process zone may decrease further during
 572 compaction. For example, strain softening because of grain crushing/fracturing and pore collapse,
 573 which was excluded here in order to isolate the effects of dissolution, can decrease further the
 574 compaction band thickness. In order to simulate the evolution of the compaction band thickness,
 575 a post-bifurcation analysis is necessary which exceeds the scope of the present paper.

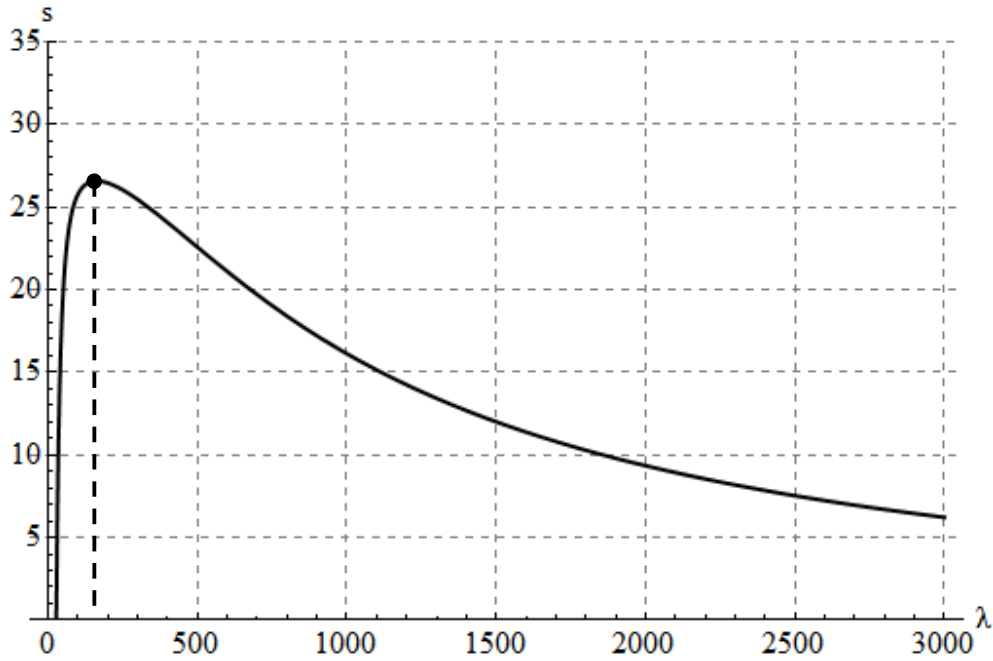
576
 577 The dominant wavelength, λ_{\max} , depends on the hydraulic diffusivity of the rock. In Figure 12
 578 we trace the Lyapunov exponent, s , versus the wavelength λ for different orders of magnitude of
 579 the hydraulic diffusivity. The lowest considered value of the dimensionless hydraulic diffusivity
 580 c_{hy} is 6×10^3 and the highest is 6×10^{10} . These values correspond to permeability values ranging
 581 from 10^{-13}m^2 to 10^{-20}m^2 . Figure 12 shows that a lower permeability of the material leads to a
 582 smaller dominant wavelength. In other words, the present model leads to thinner compaction
 583 bands for more impermeable materials. From the physical point of view, at the onset of the
 584 instability, chemical dissolution leads locally to an abrupt increase of the plastic deformations
 585 and an abrupt reduction of the effective vertical stress. In oedometric conditions, this drop of the

586 vertical effective stress leads to a fast increase of the pore pressure inside the compaction band in
587 order to equilibrate the total applied vertical stress, which is assumed constant. Therefore, the role
588 of hydraulic diffusivity is crucial, as a more permeable material would need a larger zone in order
589 for the pore pressure to rapidly increase and equilibrate the constant total vertical stress. On the
590 contrary, in a less permeable material, this zone will be narrower leading to thinner compaction
591 bands. Moreover, the faster the dissolution reaction is, the more rapidly the pore pressure has to
592 build-up in order to equilibrate the total vertical stress. Consequently, for a given value of the
593 hydraulic diffusivity parameter, the chemical softening rate controls the thickness of the
594 localization zone.

595
596 The chemical softening rate depends on the grain damage parameter a and on the dissolution
597 rate k^* . Figure 13 shows the dependency of the compaction band thickness on the grain damage
598 parameter for the numerical example presented in the previous section. As expected, stronger
599 grain damage (i.e. lower values of parameter a) leads to thinner bands (a higher specific surface
600 leads to a higher chemical softening rate). It should be emphasized, though, that this is only a
601 chemical softening effect and that the possible introduction of additional mechanical softening
602 through grain breakage would enhance this trend. Similarly, in Figure 14 we present the
603 dominant wavelength in terms of the grain damage parameter for various orders of magnitude of
604 the dissolution rate coefficient. We observe that a faster reaction rate leads to a narrower
605 compaction band at the onset of the localization. This is due to the fact that higher values of k^*
606 increase the chemical softening rate (Eq.(22) and (23)). Alternatively, the impact of the
607 dissolution rate to strain localization can be investigated through the non-dimensional expression
608 of the hydraulic diffusivity parameter. According to equation (27) by increasing the dissolution
609 rate (faster reaction) the non-dimensional hydraulic diffusivity decreases, which leads to thinner
610 compaction bands as discussed above (Figure 12).

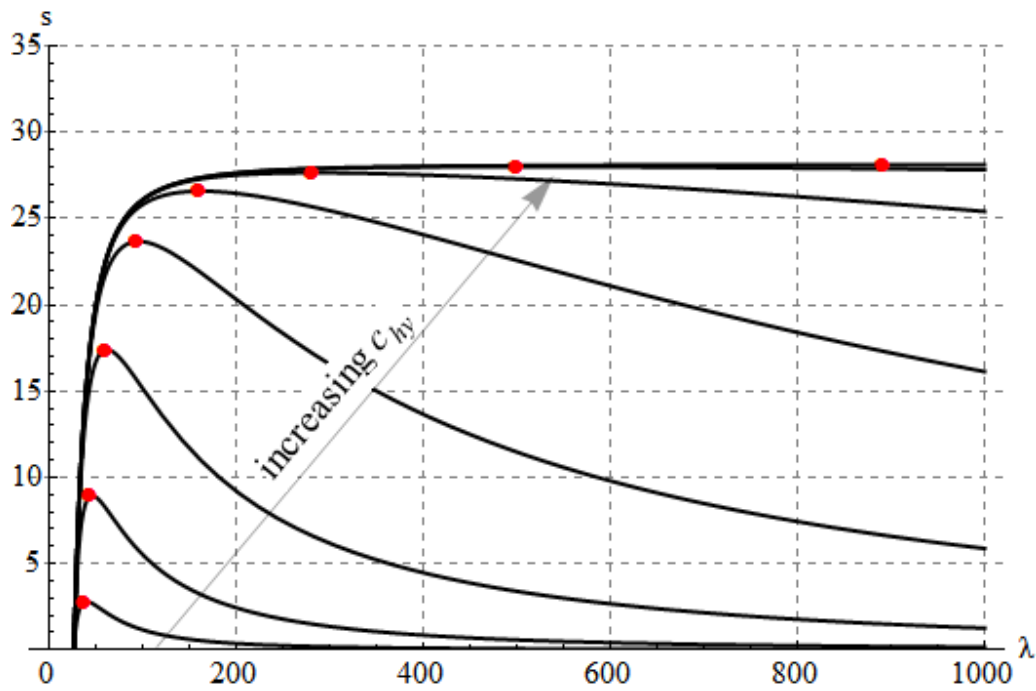
611
612 These mathematical results have to be experimentally verified. In absence of experimental
613 evidence, one could however remark that it is consistent with what is observed for shear bands
614 where the localization zone decreases with increasing mechanical or chemical softening
615 [Vardoulakis and Sulem, 1995; Sulem et al., 2011; Brantut and Sulem, 2012; Veveakis et al.,
616 2012, 2013]. In the present case, the dissolution increases the chemical softening of the material.

617 The deformation rate is linked to the dissolution rate so that a faster dissolution induces a higher
 618 softening rate.



619

620 *Figure 11. Growth coefficient, s , in terms of the perturbation wave length for $a = 10^{-2}$ and*
 621 *$\ell_c \simeq 4$. The dominant wave length corresponds to the value of λ for which the growth*
 622 *coefficient is maximum. The thickness of the compaction band is proportional to the dominant*
 623 *wave length.*



624

625 *Figure 12. Influence of hydraulic diffusivity on the dominant wavelength for $a = 10^{-2}$ and $\ell_c \approx 4$.*
 626 *The red dots represent the maximum value of the Lyapunov exponent for a given hydraulic*
 627 *diffusivity. The more impermeable the medium is the more localized the compaction instability*
 628 *becomes (smaller wavelength).*

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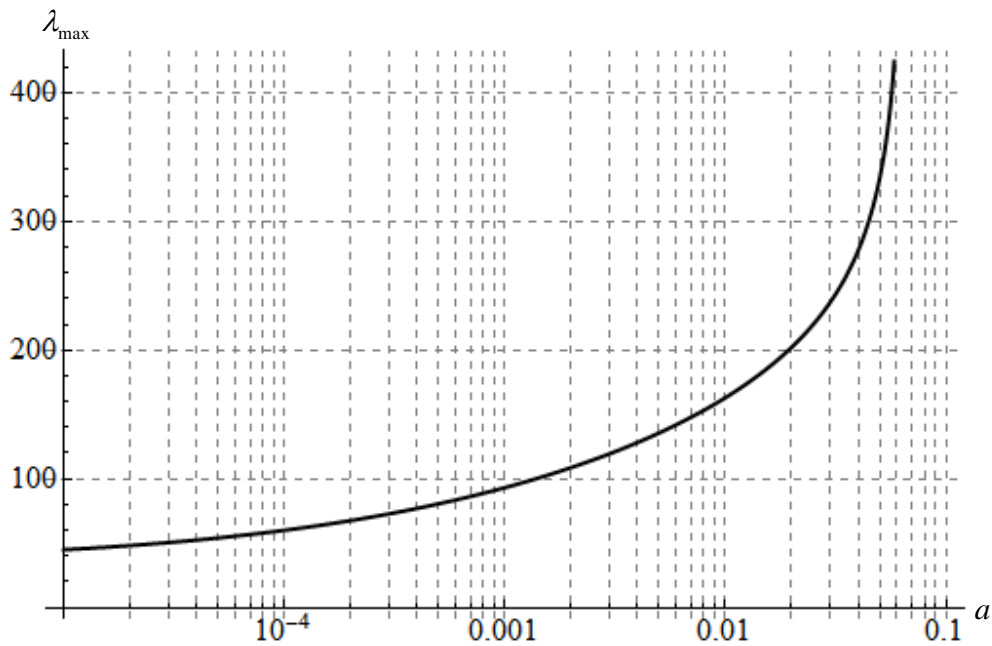
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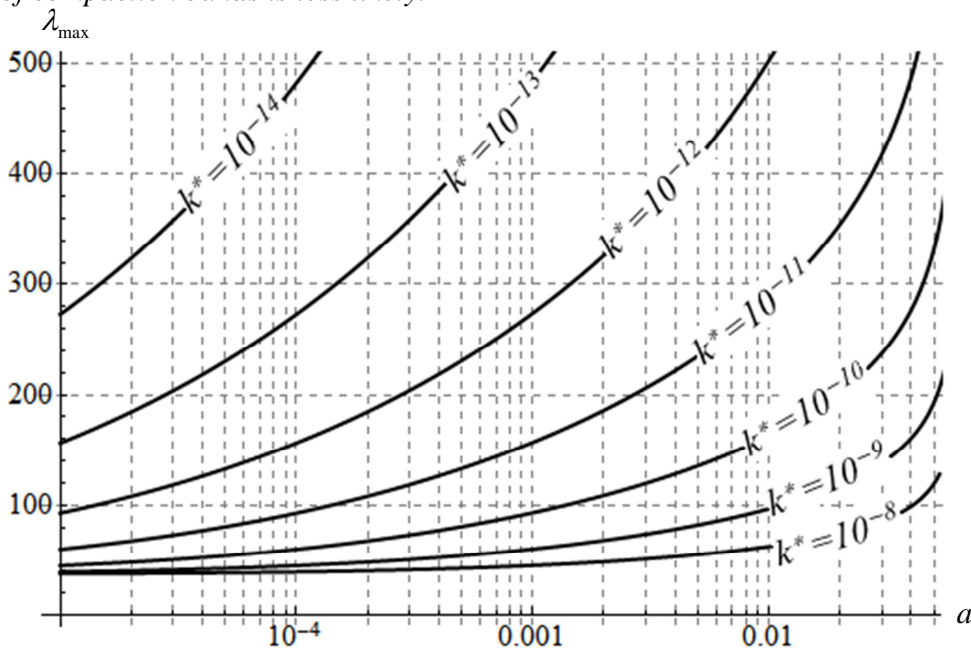
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642

643 *Figure 13. Dominant wave length (wave length selection) in terms of the grain damage*
 644 *parameter a . As the grains become less crushable, λ_{\max} tends to infinity and therefore the*
 645 *formation of compaction bands is less likely.*



646

647

648 *Figure 14. Dominant wave length (wave length selection) in terms of the grain damage*
 649 *parameter a for various values of the dissolution rate (k^* in m/s). The faster the reaction is the*
 650 *narrower the compaction band zone is.*

651 **5. Case studies**652 **5.1. Compaction banding of a sandstone reservoir**

653 The scenario of water flooding of a sandstone reservoir at 4km depth is studied in this paragraph.

654 This example could reflect the conditions of enhanced oil recovery by injecting water into the

655 reservoir to maintain the pore pressure. In offshore operations the injected water is commonly

656 seawater, which is different from the formation water. The injection of water can trigger the

657 dissolution of the quartz-based rock (R1) which in turn may trigger compaction band instabilities.

658 At this depth it is assumed that the water pressure is $p_f \approx 40\text{MPa}$ and the total vertical stress is659 $\sigma_n \approx 100\text{MPa}$. Some typical values for sandstone are given in Table 1.

660

661 *Table 1. Indicative material properties of the sandstone.*

Quantity	Value	Dimensionless value
Hydraulic diffusivity, c_{hy}	$2 \cdot 10^{-3} \text{ m}^2 \text{ s}^{-1}$	$6.25 \cdot 10^{11}$
Grain diameter, D_0^{50}	0.1 mm	1
Effective grain surface area to volume ratio, S	50 mm^{-1}	1
Bulk modulus, K	10 GPa	100
Shear modulus, G	7.5 GPa	75
Porosity, n	0,20	-
CSL slope, M	1	-
Initial yield stress, p'_0	60 MPa	0.60
Residual yield stress, p'_R	15 MPa	0.15
Chemical Softening exponent, κ	2	-
Grain damage parameter, a	0.5 MPa	0.005
μ_3/μ_2	0,6	-
Fluid density, ρ_f	1 g cm^{-3}	1
Solid density, ρ_s	2.65 g cm^{-3}	2.65
Dissolution rate, k	$10^{-8} \text{ mol s}^{-1} \text{ m}^{-2}$ ($k^* \approx 10^{-12} \text{ m/s}$)	

662

663 Before the reaction takes place the stress state is represented by the point A in the $q-p$ plane

664 (Figure 15). Elastic loading under oedometric conditions is assumed for the initial stress state, i.e.

665 point A (stress path line no. 1) and $\sigma_{zz} = -\sigma_n = -100\text{MPa}$. At time $t = 0$ the reservoir is flooded
 666 with water. The water injected in the reservoir is continuously renewed in such a way that
 667 practically open flow conditions hold ($w_2(t > 0) = 0$). As a result the system is not in chemical
 668 equilibrium and dissolution occurs. Consequently, the material is progressively degraded due to
 669 chemical softening and the chemical softening parameter decreases from its initial value $\zeta_0 = 1$.
 670 When $\zeta = \zeta_A \approx 0.9$ the material yields, plastic strains are accumulated and grain damage occurs
 671 (Eq.(19)). By neglecting in this phase the influence of the dissolution on the increase of the
 672 specific area of the grains of the rock (i.e. effective grain surface area to grain volume ratio, S , is
 673 assumed quasi-constant) an estimation of the time needed for yielding is possible. Equations (22)
 674 and (23) are combined to give, for sub-spherical grains:

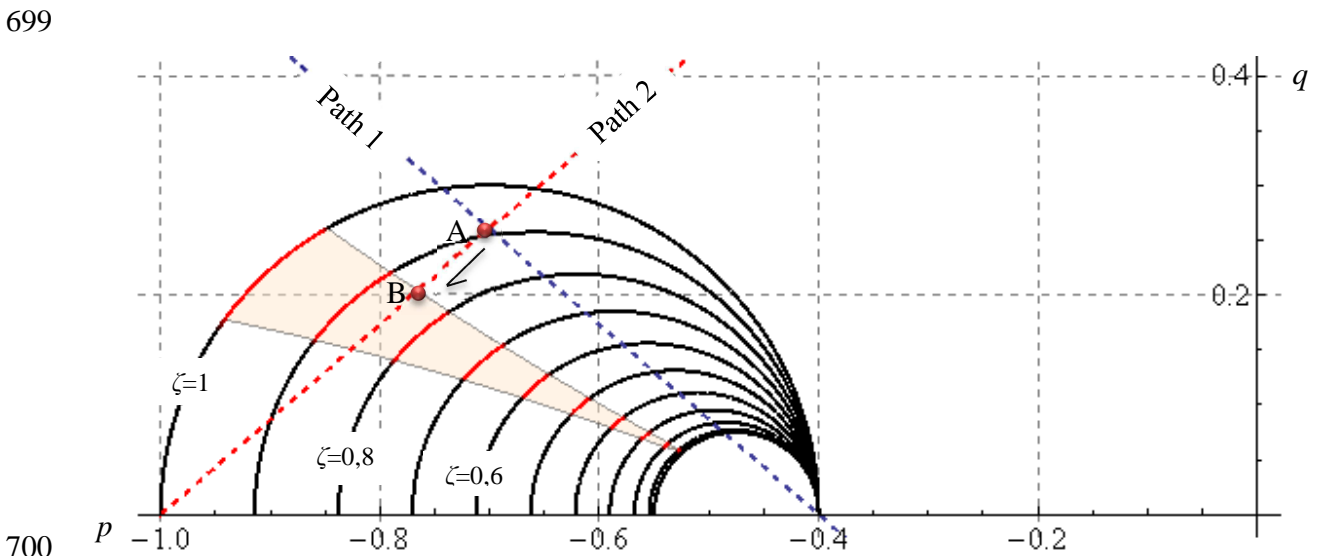
$$675 \quad t_A \approx \frac{\mu_2 \rho_s}{\mu_3 \rho_f} \frac{1}{S k^*} \ln \zeta_A^{-1} \approx 3.5 \text{ months} \quad (46)$$

676 It should be mentioned that if closed flow conditions were considered ($w_2(t = 0) = 0$,
 677 $w_2(t > 0) \neq 0$) then the dissolution would be slower and several injection cycles would be
 678 needed for yielding as the solution saturates quite quickly (equilibrium constant: $\log K_{eq} \approx -3.15$
 679 at 80°C) without provoking significant chemical softening.

680
 681 After reaching point A, the yield surface continues to contract due to dissolution and the stress
 682 state evolves until point B. At point B the system becomes unstable and compaction band
 683 instabilities are triggered. The minimum thickness of the compaction bands is equal to $\pi \ell_c$
 684 (Eq.(42)), but this can be larger depending on the exact stress path, the chemo-mechanical
 685 parameters (paragraph 4.4) and the nonlinear behavior of the system in the post bifurcation
 686 regime. Contrary to the post bifurcation regime, where the deformations are abruptly localized
 687 and the conditions can be considered undrained, drained conditions can be assumed from point A
 688 to point B if we consider that the time scale of the pore fluid diffusion is much faster than the
 689 time scale of the chemical softening process. Therefore, we can admit that the stress path after
 690 yielding follows Path 2 (Figure 15). This path is derived from the fact that the vertical stress is
 691 kept constant $\sigma_{zz} = -\sigma_n$ under oedometric conditions (see Eq.(16)).

692

693 The instability happens when $\zeta = \zeta_B \approx 0.85$. From point A to point B the specific surface of the
 694 grains is smoothly increased as microcracking, debonding and grain crushing take place
 695 (homogeneous accumulation of plastic deformations, Eq.(19)). Assuming for simplicity that
 696 $S_B \approx S_A$, the time needed for instability ($\zeta_0 \rightarrow \zeta_B$) is of the order $t_B \approx 5$ months. After this
 697 point the deformations are localized into a zone of finite thickness (Eq.(42)) and excessive grain
 698 damage occurs (onset of instability – compaction band formation).



700

701 *Figure 15. Instability region (shaded) for compaction bands under oedometric conditions for a*
 702 *sandstone at 4 km depth. Different yield surfaces are drawn for different levels of chemical*
 703 *degradation, ζ . The stress path 1 represents the elastic loading of the material under*
 704 *oedometric conditions, while the stress path 2 represents the path that the material follows after*
 705 *yielding.*

706

707 **5.2. Compaction banding induced by CO_2 injection in carbonate grainstones**

708 The formation of compaction bands in carbonate rocks is hardly evidenced both in field and in
 709 laboratory [Baud *et al.*, 2009]. Nevertheless, more recently, Cilona *et al.* [2012] observed
 710 compaction bands in porous carbonate grainstones both in field and in laboratory. Their
 711 observations extend the previous evidence of compaction bands in porous carbonate rocks by
 712 Baxevanis *et Papamichos* [2006] and Tondi *et al.* [2006]. In the aforementioned works, Hertzian
 713 cracks at grain contacts, microcracking, debonding and pore collapse that result in an overall
 714 effective grain size reduction were shown to develop inside compaction bands. As it was

715 mentioned in paragraph 3.2, these mechanisms are essentially taken into account in the proposed
 716 model through the hyperbolic fit of the evolution of the effective grain size with the mechanical
 717 energy input into the system (Eqs. (18), (19)).

718
 719 In this paragraph we attempt to explore the possibility of compaction bands formation in
 720 carbonate grainstones because of CO₂ injection at 1.8 km depth. The set of reactions that take
 721 place because of the CO₂ injection are summarized through the stoichiometry of equation R2. In
 722 Table 2 we present some typical values for the chemo-mechanical parameters of a porous
 723 carbonate rock. At this depth, we assume that the water pressure is $p_f \approx 18\text{MPa}$ and the total
 724 vertical stress is $\sigma_n \approx 45\text{MPa}$. The instability region, the elastic and plastic oedometric stress
 725 paths as well as the initial stress state (Point A) are shown in Figure 16. Following the same
 726 reasoning as in the previous case study, a CO₂ solution is injected in the rock mass. The rock
 727 dissolves and its mechanical strength diminishes (open fluid flow conditions). When
 728 $\zeta = \zeta_A \approx 0.9$ the rock starts to yield and a homogeneous plastic strain field is developed. This
 729 phase of deformation under constant applied loading (i.e. the overburden) corresponds to the
 730 creep behavior that is observed due to CO₂ injection [*Le Guen et al.*, 2007; *Liteanu and Spiers*,
 731 2009; *Rutqvist*, 2012]. At point B (Figure 16), $\zeta = \zeta_B \approx 0.63$, the system bifurcates and
 732 compaction bands are formed. Again the minimum thickness (or spacing) of the compaction
 733 bands is equal to $\pi\ell_c$ (Eq.(42)), but this can be different depending on the stress path, the
 734 chemo-mechanical parameters and the nonlinear behavior of the system in the post bifurcation
 735 regime (mechanical softening).

736
 737 Under open flow conditions, an estimation for the time needed for reaching point B is $t_B \approx 5$ days
 738 (Eq.(46) assuming a constant value for S until the instability and sub-spherical grains). Notice,
 739 that this period of time is far smaller as compared to the previous example due to the faster
 740 dissolution rate of calcite.

741
 742 For CO₂, injection open flow conditions would correspond to a zone outside the gas plume where
 743 the formation fluid is saturated with CO₂, but is not in chemical equilibrium with the rock so that

744 carbonate dissolution occurs continuously ($w_2 < w_2^{eq}$). Rohmer and Seyedi [2010], using an
 745 hydro-mechanical model on a reservoir case study, show that the dissolution front might extend
 746 few kilometers around the injection well after 10 years of continuous injection.

747

748 Table 2. Indicative material properties of a carbonate grainstone.

Quantity	Value	Dimensionless value
Hydraulic diffusivity, c_{hy}	$10^{-3} \text{ m}^2 \text{ s}^{-1}$	$3 \cdot 10^5$
Grain diameter, D_0^{50}	0.2 mm	1
Effective grain surface area to volume ratio, S	25 mm^{-1}	1
Bulk modulus, K	5 GPa	110
Shear modulus, G	5 GPa	110
Porosity, n	0.25	-
CSL slope, M	0.9	-
Initial yield stress, p'_0	30 MPa	0.67
Residual yield stress, p'_R	0 MPa	0
Chemical Softening exponent, κ	1	-
Grain crushing parameter, a	1 MPa	0.011
μ_3/μ_2	0.62	-
Fluid density, ρ_f	1 g cm^{-3}	1
Solid density, ρ_s	2.65 g cm^{-3}	2.65
Dissolution rate, k	$10^{-6} \text{ mol s}^{-1} \text{ m}^{-2}$ ($k^* \approx 1.6 \cdot 10^{-10} \text{ m/s}$)	

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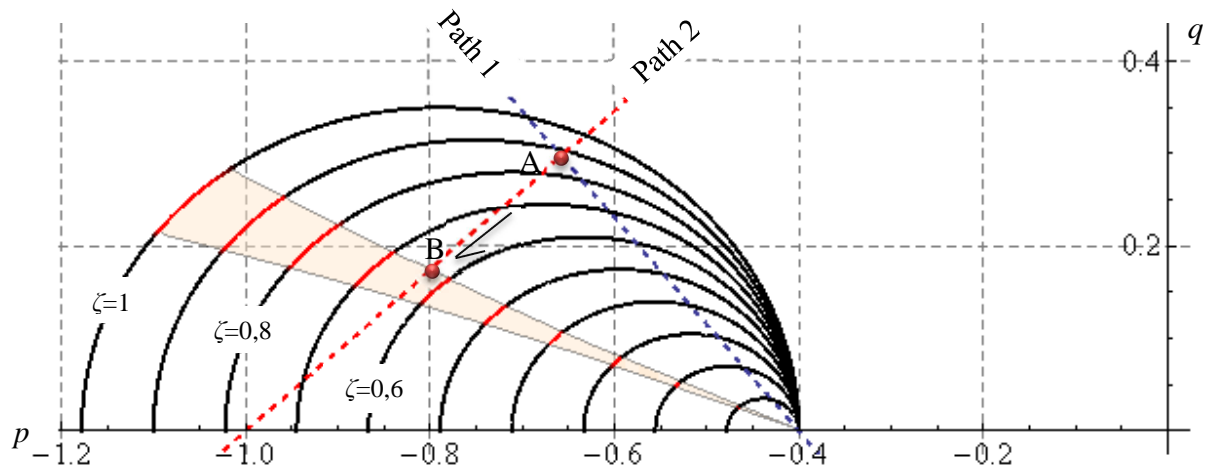
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761

762 *Figure 16. Instability region (shaded) for compaction bands under oedometric conditions for a*
 763 *carbonate grainstone at 1.8 km depth. Different yield surfaces are drawn for different levels of*
 764 *chemical degradation, ζ . The stress path line no. 1 represents the elastic loading of the material*
 765 *under oedometric conditions, while the stress path line no. 2 represents the path that the material*
 766 *follows after yielding.*

767

768 6. Conclusions

769 Grain fracturing and attrition, “microcracking” of the cementitious matrix, debonding and pore
 770 collapse are some important micro-mechanisms that lead to the reduction of the effective grain
 771 size of a geomaterial and consequently to the increase of the dissolution front in reactive
 772 environments. The proposed chemo-poro-mechanical model accounts for the aforementioned
 773 mechanisms. Part of the mechanical energy supplied to the system is dissipated through various
 774 micro-mechanisms for the creation of new surfaces in the microstructure. In this sense, a
 775 phenomenological approach based on the work of Lade et al. [1996] is used to relate the effective
 776 grain specific surface and the total energy input per specimen unit volume. Following this
 777 approach, which describes the above dissipative mechanisms through a single parameter, and in
 778 the frame of dissolution process, the chemo-poro-mechanical model presented herein provides a
 779 strong coupling between mechanics and reaction kinetics (Figure 1).

780

781 The purpose of the present paper was to explore the possibility of chemically induced compaction
 782 band instabilities. Therefore, in order to isolate the chemical effects, mechanical softening

783 induced by grain damage was not considered in the present form of the proposed chemo-poro-
784 mechanical model. The stability of the system is studied through a linear stability analysis, which
785 revealed the critical parameters for which compaction bands may occur in quartz- and carbonate-
786 based rocks. Porosity and grain damage sensitivity (parameter a) are important parameters that
787 considerably influence the triggering of compaction band instabilities due to dissolution. For a
788 specific rock, the determination of the exact value of the grain damage sensitivity parameter is
789 possible and may include classical mechanical tests, grain-size analyses and/or microstructural
790 observations and measurements (e.g. X-ray tomography, acoustic emissions). The other
791 parameters of the model (elastic constants, yield surface, chemical softening factor and reaction
792 kinetics) can also be determined experimentally.

793
794 Due to the existing heterogeneity of the microstructure (e.g. different grain sizes and constituents
795 in the REV) the dissolution rate may not be homogeneous over the REV. The size of the REV is
796 a finite statistical quantity that depends upon the geomaterial at hand. Therefore, a rock is
797 characterized by at least one characteristic length, which by appropriately up-scaling the micro-
798 level to the macro-level (separation of scales and averaging) can be adequately expressed at the
799 macroscale, i.e. incorporated into the proposed chemo-poro-mechanical model. The linear
800 stability analysis shows that because of this characteristic length, the deformation is localized into
801 narrow bands of finite thickness and that these bands are periodically distributed in space. This is
802 in accordance with many field and laboratory observations where there is evidence that
803 compaction bands are localized into a thin band (several grains thickness) and are quasi-
804 periodically arranged in space. In field and laboratory observations, the thickness of compaction
805 bands is variable. In the present analysis the compaction band thickness depends on the chemo-
806 poro-mechanical properties of the material and the heterogeneity of the microstructure.
807 According to the present model, the thickness and the periodicity of compaction bands are related
808 to the grain damage parameter, the hydraulic diffusivity and the dissolution rate. The more
809 crushable the grains are, the lower the permeability is and the higher the dissolution rate is, the
810 narrower the compaction band is. Nevertheless, the results presented here have a qualitative
811 character. The study of the precise evolution of the system and of the compaction band thickness
812 after the onset of localization (post-bifurcation regime) exceeds the scope of the present paper

813 and will be pursued further in a future work by integrating the non-linear chemo-poro-mechanical
814 system of equations.

815

816 Finally, two scenarios of chemically induced compaction band instabilities were investigated.
817 The first one concerns the water flooding of a quartzic rock reservoir, while the second one
818 concerns a carbonate grainstone in which a CO₂ solution is injected. The effect of open and
819 closed flow conditions was discussed, the instability regions were determined and a rough
820 estimation of the critical time for compaction band triggering was made. Two phases are
821 distinguished. The first phase is characterized by a creep behavior of the rock, while in the
822 second phase compaction band instabilities are triggered. The present study focused on pure
823 compaction bands, but the concept of the present model can be extended to other types of
824 deformation bands, such as shear bands. The formation of deformation bands in reservoirs due to
825 chemical processes and other non-linearities can be related to important changes in permeability,
826 to the creation of reinforced zones (compactive behavior) or, in the contrary, to the creation of
827 new faults and fractures (shear behavior). However, in order to quantify the implications of such
828 phenomena to the sustainability of reservoir projects, large scale simulations are necessary that
829 will take into account the aforementioned phenomena and the precise injection characteristics.

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832 their fruitful comments and suggestions, which improved the paper. Moreover, they would like to
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834 SEED-0003).

835 **8. Appendix**

836 **8.1. Reaction kinetics in terms of mass fraction and specific area**

837 At the microscale the mass fraction is:

838
$$w_2 = \frac{m_2}{m_f} \text{ or } w_2 = \frac{\mu_2}{\rho_f V_f} n_2 \quad (\text{A1})$$

839 where m_f is the mass of the fluid phase and m_2 is the mass of the dissolution product at the
 840 microscale. Assuming that $w_2 \ll 1$ or equivalently that the density of the fluid, ρ_f , remains
 841 practically constant during the reaction process, we obtain that:

842
$$\frac{\partial n_2}{\partial t} \approx \frac{\rho_f}{\mu_2} e V_s \frac{\partial w_2}{\partial t} \quad (\text{A2})$$

843 Furthermore, assuming that the activity coefficients for the reactants are close to unity the
 844 reaction quotient is expressed in terms of the reaction activities of the reactants as follows:

845
$$Q \approx \frac{a_2}{a_1} \text{ or } Q \approx \frac{n_2}{n_1} \quad (\text{A3})$$

846 Assuming again that $w_2 \ll 1$, we obtain:

847
$$Q \approx w_2 \frac{\mu_1}{\mu_2} \quad (\text{A4})$$

848 Consequently the mass fraction at equilibrium is related to the equilibrium constant as follows:

849
$$w_2^{eq} \approx \frac{\mu_2}{\mu_1} K_{eq} \quad (\text{A5})$$

850 Using the above expressions, Eq.(20) yields:

851
$$\frac{\partial w_2}{\partial t} \approx k \frac{\mu_2}{\rho_f} \frac{S}{e} \left(1 - \frac{w_2}{w_2^{eq}} \right) \quad (\text{A6})$$

852 where we set $S = \frac{A}{V_s}$.

853 **8.2. Chemical softening parameter and mass fraction**

854 By definition $\zeta = \frac{M_s}{M_s^0}$ and using the stoichiometry of the dissolution reaction we obtain:

855
$$\frac{\partial \zeta}{\partial t} = \frac{1}{M_s^0} \frac{\partial M_s}{\partial t} = -\frac{\mu_3}{M_s^0} \frac{\partial n_2}{\partial t} \quad (\text{A7})$$

856 Using Eq.(A2) at the macroscale (REV) we get for $w_2^M \ll 1$:

857

858
$$\frac{\partial \zeta}{\partial t} \approx -\frac{\mu_3}{\mu_2} \frac{M_f}{M_s^0} \frac{\partial w_2^M}{\partial t} = -\frac{\mu_3}{\mu_2} \frac{\rho_f}{\rho_s} e \frac{M_s}{M_s^0} \frac{\partial w_2^M}{\partial t} \quad (\text{A8})$$

859 or

860
$$\frac{\partial \zeta}{\partial t} \approx -\frac{\mu_3}{\mu_2} \frac{\rho_f}{\rho_s} e \zeta \frac{\partial w_2^M}{\partial t} \quad (\text{A9})$$

861

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1018 **List of Captions**

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1021	debonding etc.).....	6
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