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COUPLED CHEMO-MECHANICS OF EVOLVING PERMEABILITY IN GEOMATERIALS

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ABSTRACT: *Flow of fluids in geomaterials may be strongly affected by chemo-mechanical processes, which are either externally induced or spontaneous. This is the case when dissolution, transport of the dissolved mineral and its precipitation in the neighborhood pore space causes a decrease of what is upscaled as Darcian permeability. Factors affecting the process are studied at the micro-scale using a model of a system of vessels of variable length and opening formed in the neighborhood of a stressed contact between two damage-affected grains. It appears that permeability is mildly affected by the contact area increase, and for most of the duration, by the precipitation of the mineral solute, until the inter-grain pores are almost completely clogged by the precipitate, when the permeability decreases very fast by orders of magnitude. Rigid chemo-plasticity model is employed to simulate the enhancement to dissolution induced by formation of new inter-phase interfaces at the walls of micro-cracks, represented by dilatant plastic strain. Such a process is widely believed to occur in oil/gas bearing sediments. Dissolution leads to chemo-plastic softening of the material. Couplings, feedbacks and feedforwards between mechanical, transport and geochemical processes caused by intergranular damage and dissolution are discussed.*

1 INTRODUCTION

Geomaterials respond to some environmental circumstances through generation of a series of self-regulatory mechanisms of damage, deformation, erosion, and chemical processes or reactions: e.g. osmosis, dissolution and precipitation. These mechanisms are coupled at different scales. Several natural geomechanical processes, as sediment compaction, rock weathering or landsliding appear to include such sequences of mechanisms. A better understanding of the involved processes will improve the chances to control and/or engineer better the geo-materials and geo-structures subject to such processes.

We investigate mechanisms that control the rate of damage to geomaterials induced by geochemical processes at the level of intergranular contact in sediment compaction eventually critically affecting its permeability. Our focus is on irreversible changes expressed via chemo-plasticity studied numerically at three scales: those of grain, grain- assembly- and continuum-scale. The proposed scenario includes: damage in the near-contact area via microfracturation; activation of the microfracture wall surfaces as sites for dissolution of minerals entering in contact with permeating water; mass removal due to dissolution causing material softening; diffusion and advection of the dissolved mineral across the grain toward the external pore space; diffusion within the pore fluid of the dissolved minerals; and finally precipitation of minerals onto free grain surfaces. This paper presents an extension of our previous work (Hu and Hueckel, 2007) to the effect of the above processes on sediment fluid

transmissibility. Such effects are crucial in petroleum extraction engineering and compaction of clay barriers.

Rigid chemo-plasticity is used to simulate the contact phenomena in the grain at the microscale, coupled with the reactive-diffusive transport of the dissolved mineral across the grain. Upscaled variables at mesoscale simulate the change of permeability of the grain system as a result of the mineral precipitation. The resulting redistribution of mass within the pore space is controlled by microscopic and mesoscopic chemical gradients, which in turn are controlled by microscopic damage variables. Partial masses of the same mineral are shown to play a different role in the sediment stiffness changes, changes in porosity and in permeability at the macro- and meso-scale requiring them to be linked to different processes (dissolution and precipitation) derivable only at the microscale. Self-regulation is exercised via stress reduction by the increasing contact surface area, and possibly by the generation of pore pressure, due to the dissolution flux via the increasing reaction site area controlled by damage and eventually a permeability decrease.

2 OUTLINE OF BASIC MECHANICAL, HYDRAULIC AND GEOCHEMICAL PROCESSES

The natural process of normal consolidation of sediments is believed to be a superposition of several coupled processes regarding the sediment solid matrix and pore fluids. The processes involved are: deformation of sediment grains, including plastic deformation, especially near contact between grain asperities and corresponding locally smooth surface of another grain; dilatant damage induced by the plastic yielding; infiltration of the dilatant zone by pore water; activation of damage (microcracking) related internal interfaces between pore liquid and solid; dissolution of minerals at these interfaces; diffusion of the dissolved species within the grain away from the reaction sites; intergrain diffusion of the dissolved minerals in the pore water and precipitation of those on the free surface of adjacent or remote grains.

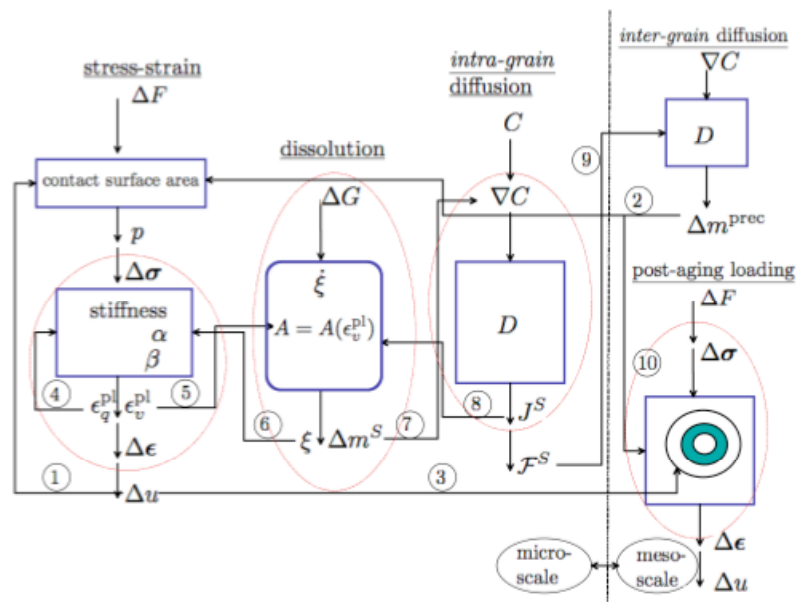


Fig. 1: Schematic of feedbacks and feedforwards in coupled processes of mineral reaction assisted sediment compaction and stiffening (for identification labels see Hueckel and Hu, 2009)

The above sequence of processes is common to other phenomena such as aging (see Mitchell and Solymar, 1984; Hueckel et al., 2001; Hueckel et al., 2005), or pressure solution

in petroleum reservoir modeling (Rutter, 1976; Lehner, 1995). Hu and Hueckel (2007) have developed a sequence of three-scale models to simulate aging, and identified leading feedbacks and feedforwards of interscale coupling to evaluate the evolution of meso-scale stiffness. A summary of the feedbacks and feedforwards are redrawn in Fig. 1. In this paper we apply that model to address the evolution of permeability, and feedbacks involved in it.

The three-scale model of sediment compaction is described in detail by Hu and Hueckel (2009). In what follows only main concepts are presented. The models at the microscale include stress-strain relationship, dissolution law, and intra-grain diffusion. At the meso-scale the models include intergranular diffusion with precipitation and a resulting increase of material stiffness. The macro-scale is not shown in the figure. The micro-scale medium is a porous material of the solid grain minerals. Its porosity is the internal grain porosity, and the mechanical properties of the material are those of the solid grain mineral. Stress and strain (positive in compression) are microstress and microstrain at the scale of a fraction of the grain (Fig. 2).

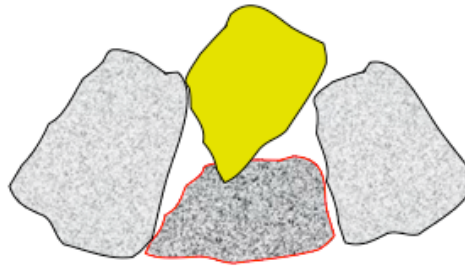


Fig. 2. Schematic of a rigid grain indentation into an adjacent rigid-plastic grain

The material of individual grain is taken as rigid plastic. To describe mathematically a near contact failure one needs to identify a micro-stress (σ_{ij}) yield locus, $f(\sigma_{ij}) \leq 0$, within which no strain (ε_{ij}) occurs, whereas at yielding, the strain rate is entirely irreversible

$$f < 0, \text{ and for } f = 0, \dot{f} < 0: \dot{\varepsilon}_{ij} = 0; \\ \text{whereas for } f[\sigma_{ij}, p_c] = 0 \text{ and } \dot{f}(\dot{\sigma}_{ij}, \dot{p}_c) = 0: \dot{\varepsilon}_{kl} = \dot{\varepsilon}_{kl}^{irr} \neq 0 \quad (1)$$

The superimposed dot over a symbol denotes a time rate.

Dilatancy-damage at the sub-grain scale is a critical variable for the entire concept as a vehicle of the chemo-mechanical coupling. It is linked to micro-cracking, which plays multiple roles. First, it provides a connected network across the damaged part of the grain that becomes instantly permeated with water as a result of suction induced by dilatancy. Second, micro-crack walls form new solid-fluid interface, which constitutes a source of dissolution of mineral species, that is solid mass removal. Third, irreversible micro-slips occur across micro-cracks. As a result of micro-slips and mass removal, the yielding behavior of the material is affected by two competing plastic hardening mechanisms: deviatoric strain-hardening and mass removal softening. Hence, p_c which is the apparent preconsolidation stress, i.e. isotropic size characteristics of the yield locus, depends on two hardening parameters, which are mechanical (ε_q^{irr}) or chemical (ξ) in nature (Hueckel, 2002).

$$p_c = p_c(\boldsymbol{\varepsilon}_q^{irr}, \xi), \text{ where } \boldsymbol{\varepsilon}_q^{irr} = \left| \left(\frac{2}{3} \dot{\boldsymbol{\varepsilon}}_{ij}^{irr} \dot{\boldsymbol{\varepsilon}}_{ij}^{irr} \right)^{1/2} \right| \text{ and } \dot{\boldsymbol{\varepsilon}}_{ij}^{irr} = \dot{\boldsymbol{\varepsilon}}_{ij} - \frac{1}{3} \dot{\boldsymbol{\varepsilon}}_{kk}^{irr} \boldsymbol{\delta}_{ij} \quad (2)$$

Chemical softening parameter ξ is an accumulated relative mass removal of a single mineral species that dominates the material strength, computed with respect to the original total mass of that particular species. The relationship between change in the apparent preconsolidation stress p_c and a reaction progress variable is established empirically. A mineral in the example, dominating the strength of quartz sand is silica. Dissolution reaction of silica in water may be measured through a change in activity of its product which is silicic acid H_4SiO_4 formed in the aqueous solution. The rate of silica dissolution is determined by Rimstidt and Barnes (1980) formula as proportional to the specific surface area of the solid-fluid interface A , normalized with respect to 1 m^2 to yield a non-dimensional quantity \tilde{A}

$$\frac{da_{\text{H}_4\text{SiO}_4}}{dt} = \tilde{A} \gamma_{\text{H}_4\text{SiO}_4} (k_+ a_{\text{SiO}_2} a_{\text{H}_2\text{O}}^2 - k_- a_{\text{H}_4\text{SiO}_4}) \quad (3)$$

where a_i are activities, and γ_i , activity coefficients, of i -th species, while k_+ and k_- are rate constants of respectively forward and backward reactions. \tilde{A} is a dimensionless specific interfacial surface area, as above, per unit mass of pore fluid. For details see Hu and Hueckel, (2007). ξ is a reaction progress $\xi=1$ variable constrained by the inequalities: $0 \leq \xi \leq 1$; when, the reaction is completed, that is all silica is removed from the material.

In the current context the damage occurs within a single grain and consists in opening of microcracks. A new scalar variable, \tilde{a} , represents the amount of the added interface surface area per unit volume of the grain medium. It is linked to the relative reaction area, \tilde{A} , $\tilde{a} = \tilde{A} \frac{n_g \rho_w}{\rho^0}$, where $\rho^0 = 1 \text{ kg/m}^3$, ρ_w is the density of water, n_g is porosity of the grain solid.

The new internal interface surface area generated by the micro-cracking per unit volume is proposed to be proportional to the volumetric strain. Hence,

$$\tilde{A} = \tilde{A}(\boldsymbol{\varepsilon}_v^{irr}) = \phi |\boldsymbol{\varepsilon}_v| + \phi_c; \quad \boldsymbol{\varepsilon}_v < 0 \quad (4)$$

where ϕ is a constant, whereas ϕ_c represents the specific surface area of pre-existing voids. For $\boldsymbol{\varepsilon}_v > 0$, $\phi = 0$, which aims at excluding the compressive strain for which there are no micro-cracks and hence no change in the dissolution surface area. Therefore

$$\dot{\xi} = (\Xi |\boldsymbol{\varepsilon}_v| + \Xi_o) \gamma_{\text{H}_4\text{SiO}_4} k_+ a_{\text{SiO}_2} a_{\text{H}_2\text{O}}^2; \quad \Xi = s\phi |\boldsymbol{\varepsilon}_v|; \Xi_o = \text{const}; \quad \boldsymbol{\varepsilon}_v < 0 \quad (5)$$

Notably, precipitation at the dissolution site is neglected.

The irreversible strain rate mode is determined by the associated flow rule, whereas its magnitude by the plastic multiplier $\dot{\lambda}$ resulting from the extended Prager's consistency condition, $\dot{f}(\boldsymbol{\sigma}_{ij}, \dot{\boldsymbol{\varepsilon}}_q^{irr}, \dot{\xi}) = 0$ (see e.g. Hueckel, 2002). Therefore, the strain rate is a function of the rates of stress and reaction progress

$$\dot{\epsilon}_{ij}^{irr} = \frac{1}{H} \left(\frac{\partial f}{\partial \sigma_{mn}} \dot{\sigma}_{mn} + \frac{\partial f}{\partial \xi} \dot{\xi} \right) \frac{\partial f}{\partial \sigma_{ij}}; \quad H = - \frac{\partial f}{\partial \epsilon_q^{irr}} \left[\frac{2}{3} \frac{\partial f}{\partial s_{ij}} \frac{\partial f}{\partial s_{ij}} \right]^{\frac{1}{2}}; \quad s_{ij} = \sigma_{ij} - \frac{1}{3} \sigma_{kk} \delta_{ij} \quad (6)$$

In particular, at constant stress, an irreversible strain rate is generated proportional to the reaction rate. The yield locus in the form of a set of linear functions for single principal stress components has the advantage of a very simple kinematics, yet capturing the essence of straining,

$$\frac{\sigma_1}{\sigma_{01}} = 1 + \alpha \epsilon_q - \beta \xi \geq 0, \quad \frac{\sigma_2}{\sigma_{02}} = 1 + \alpha \epsilon_q - \beta \xi \geq 0; \quad \sigma_{01} \geq 0; \quad \sigma_{02} \leq 0 \quad (7)$$

where α and β are constants, σ_3 remains undetermined. As discussed earlier, ξ is the relative mass removal (its rate is described by eq. (5)). At this point the precipitation term is ignored, as it would lead to a constitutive non-linearity, making it impossible to solve the equation system semi-analytically.

The intra-grain diffusion of silicic acid is limited to the damaged zone, as shown in Fig. 3a. It is described (in radial coordinates with axial symmetry here) by a linear reactive diffusive transport law combined with a mass balance law, with a reaction term (last term of the RHS) through which transport is coupled to deformation via eq. (6):

$$\frac{\partial x_{\text{H}_4\text{SiO}_4}}{\partial t} = D \left(\frac{\partial^2 x_{\text{H}_4\text{SiO}_4}}{\partial r^2} + \frac{1}{r} \frac{\partial x_{\text{H}_4\text{SiO}_4}}{\partial r} \right) + \frac{k_+ (\Xi \epsilon_v + \Xi_0)}{s} \quad (8)$$

where $x_{\text{H}_4\text{SiO}_4}$ is molar fraction of aqueous silica in the fluid phase within the grain. Assuming the pore fluid to be a dilute solution, the molar fraction of any of its species k , has mass content $m_{kF} = M_{kF} / V_0$, linked to molar fraction, x_{kF} through V_0 which is the reference volume of the entire grain medium. D is the solute diffusion coefficient.

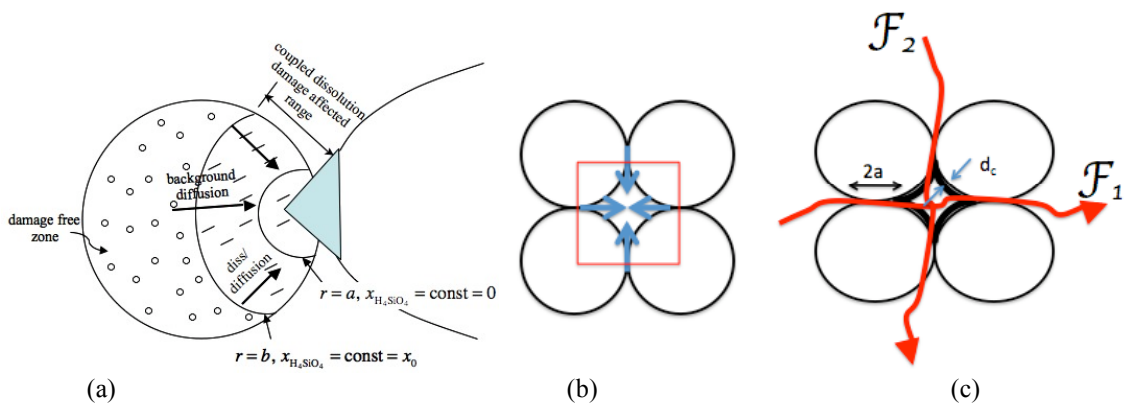


Figure 3. (a) A cartoon showing intra-grain diffusion of the dissolved species; (b) in-grain pore with the flux of the dissolved silica; (c) pore fluid flow through a dissolution altered meso-scale inter-grain pore system

The above system of constitutive equations is to be supplemented by equilibrium equations and kinematics expression to yield displacements. Hu and Hueckel (2007) provided a solution to the coupled chemo-mechanical deformation-diffusion using an axisymmetrical approximation proposed for the contact problem by Johnson (1985).

3 CHEMO-MECHANICALLY INDUCED EVOLUTION OF PERMEABILITY

In a similar way to the evolution of stiffness, the evolution of permeability is evaluated at the meso-scale, at which we consider as a REV a quadruplet of 1/4th grains, with equal radiuses, R , as shown in Fig. 3b.

There are two processes that affect the inter-grain pore system as shown in Fig. 3b: the increase of the contact size between two stressed grains due to chemo-plastic deformation of the grain and the precipitation of the solute within the pore. In terms of porosity variation, the effect of contact area increase is more significant, as seen in Fig. 4a. However, it is an open question if the same is true for the permeability evolution.

The current radius of contact may be approximated as $a=a_0+u_a$ here a_0 is the initial radius of the contact, while u_a is the vertical indentation advancement. Fig. 5 shows the evolution of the radius a in terms of its variable part of u_a as a function of time depending on the dissolution rate constant k_+ .

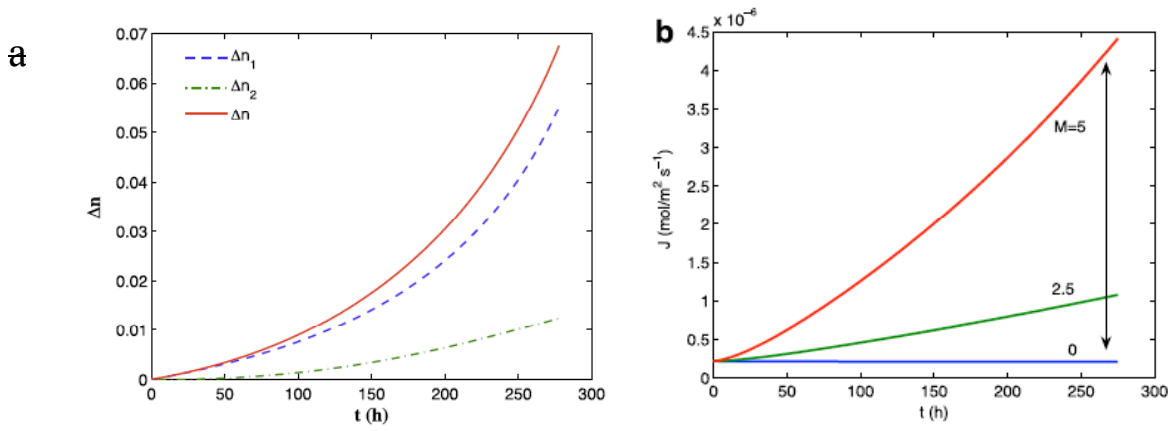


Figure 4. (a) Inter-grain porosity change (Δn), originated from deformation of the grain (Δn_1) and from precipitation induce grain coating; (b) Flux of dissolved silica from a grain as a function of intra-grain mass transfer coefficient $M = \Xi k_+ a^2 / D x_0$, where x_0 is the initial concentration of silicic acid within the grain

The rate of the formation of coating can be calculated as a change in the coating thickness, \dot{d}_c over an area, dS of the free surface, as certain number of moles of silica are precipitated in the water volume, dV ,

$$\dot{d}_c = \frac{VOL_{\text{pre}}}{dS} = \frac{MOL_{\text{pre}} \nu_{\text{SiO}_2}}{dS} = \frac{k_- x_{\text{H}_4\text{SiO}_4} \cdot dV \cdot \nu_{\text{SiO}_2}}{\nu_{\text{H}_2\text{O}} dS} = \frac{k_- x_{\text{H}_4\text{SiO}_4} \bar{\omega}_p \nu_{\text{SiO}_2}}{\nu_{\text{H}_2\text{O}}} \quad (9)$$

where $\bar{\omega}_p = V_{\text{void}} / S_{\text{free}} = (2/\pi - 1/2)R$, ν_{SiO_2} and $\nu_{\text{H}_2\text{O}}$ are the molar mass of SiO_2 and H_2O , respectively. $x_{\text{H}_4\text{SiO}_4}(\theta, t)$ is the distribution of precipitating silica determined from a steady state solution of diffusion-precipitation transport equation, where θ , $-\pi/4 < \theta < \pi/4$ is an angular coordinate along the grain surface, with a grain mineral mass flux $\pm f_p(t) = \mp J_a \pi a / 4 \bar{\omega}_p$, as a boundary conditions at the contact points, while $J_a(t)$ is the mass flux across the inner boundary of the grain, $r = a$ (see Hu and Hueckel, 2007 for details). As a result of linearization of eq. (9), the coating rate becomes uniformly distributed, i.e. independent of θ .

$$\dot{d}_c = \frac{J_a a v_{\text{SiO}_2}}{R v_{\text{H}_2\text{O}}} \quad (10)$$

The relative coating rate \dot{d}_c/R is plotted against time in Fig.5b.

The two variables shown in Fig. 5 control the overall change in permeability of the pore system as visualized in Fig. 4c. Notably their dependence with time is different. The change in the contact area appears to accelerate with time, whereas the coating rate increases nearly linearly, after an initial period.

To evaluate the effect of the two factors, a simple formula based on Poisseuille flow through two connected in-series planar tubes of different thickness, will be studied following Hueckel et al. (1997) by examining meso-scale structural changes. Given, that gravity plays an essential role in stress pattern, in which horizontal contacts are usually acted upon by twice as big grain force than the vertical contacts, we shall limit our consideration to conduit denoted as \mathcal{F}_1 . Explicit data are needed to calculate the permeability evolution as listed in Table 1.

Table 1: Characteristic dimensions of the flow conduit

name	symbol	value
grain size	R	1mm
maximum damage zone	b	0.7mm
size of the indent radius	$a_0=b/10$	0.07mm
asperity size=contact slit	δ_c	0.05mm
initial (nominal) pore opening	δ_{p0}	0.86mm

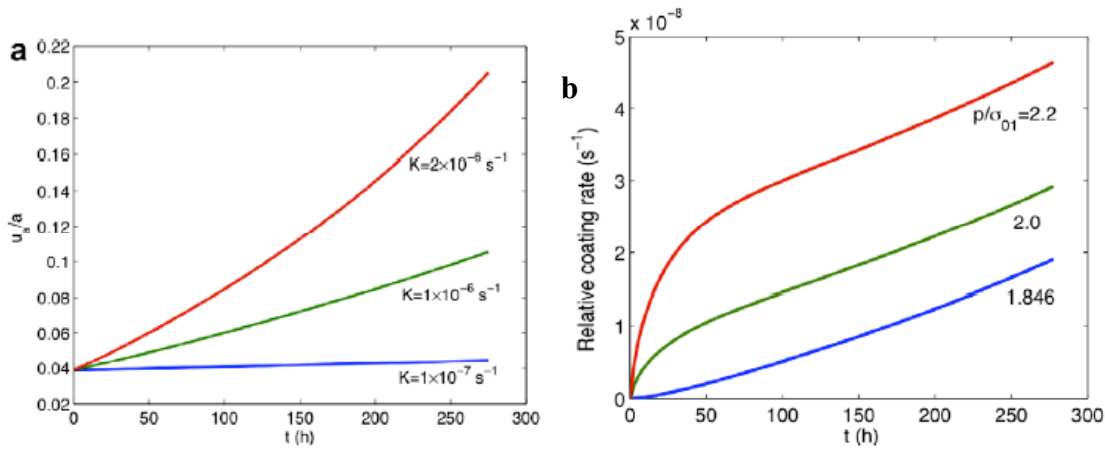


Fig. 5: (a) Increment of the contact area (also grain penetration) as function of time and rate constant; (b) relative coating rate of the walls of inter-grain pore (also of free surfaces of grains) for different values of constant inter-grain pressure. Normalized with respect to grain size, R (calculated for $k_+=1 \times 10^{-12} \text{ s}^{-1}$).

The flow is envisioned as occurring through a 2D flat vessel composed of the intergranular slit communicating with the adjacent inter-grain pore, as shown in Fig. 6. The intergrain slit has a very limited opening, equivalent to a size of an asperity, or a piece of mineral debris locked in the contact space. As such it will be assumed not to change during the process of deformation, and enlarging of the contact extent.

where K^t is a coefficient describing the evolution of the intrinsic permeability due to chemo-mechanical coupling in the medium, v_i denote the molar mass of species “i”. As may be easily anticipated from the form of the expression for K^t , the evolution of the permeability will be not much sensitive to the effect of pore coating, until well into an advanced stage when the size of the pore approaches the size of the contact slit, as seen in Fig. 7.

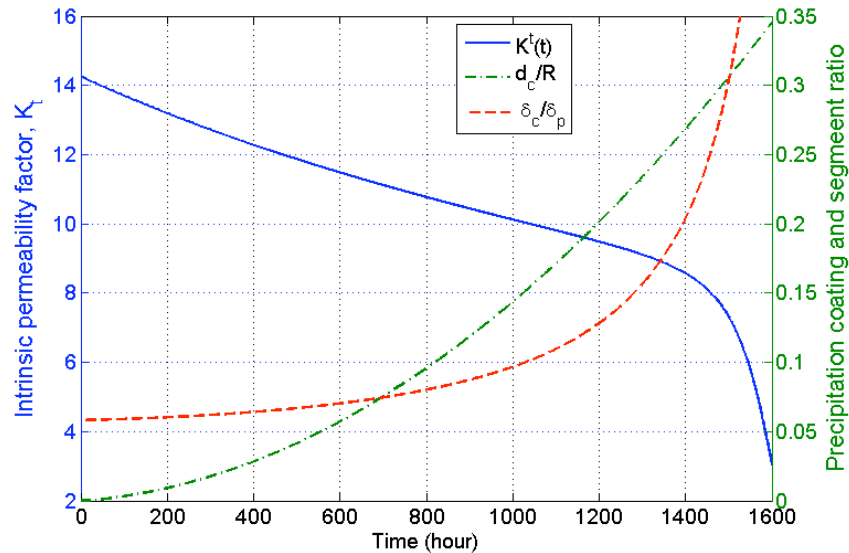


Fig.7: Evolution of permeability: $K^t(t)$ -intrinsic permeability factor (left scale); ratio of the thickness of contact slit to the pore size, δ_c/δ_p (right scale); integrated precipitation pore coating d_c/R

The intrinsic permeability factor (time dependent) is seen to decline slightly (30%) in the first 1400 hours mainly due to the increase of the contact area, whereas it seems to be unaffected by the pore wall coating by precipitating mineral. This abruptly changes in a short time, when the pore is practically clogged by the precipitate, and hence its size becomes of the order, and then even smaller than the intergranular contact slit. So, within a few days the permeability drops over one order of magnitude, and more. It is therefore concluded that precipitation is felt very quickly as far as the increase of stiffness is concerned (Hu and Hueckel⁶), while it is not felt in terms of permeability until it reaches a critical moment when permeability drastically drops in a short time. A practical conclusion from the simulation concerns the critical time to clogging that may be easily estimated from the mass dissolution rate needed to produce a solute to fill almost completely the pore.

4 CONCLUSIONS

From the presented analysis it can be seen that the strongest feedforward to permeability evolution is provided by dissolution and damage, hence the chemo-plastic process. Notably, another implication of feedback (1), Fig. 1, regarding the increase of the contact area is not as significant (at least if the hypothesis of asperity is valid). Nevertheless, its effect on reducing the contact stress (not quantified here), may lead to a retardation of the dissolution enhancement. Further reduction of stress may come from generation of a substantial pore pressure, as permeability drastically decreases. Pore pressure, at a constant overburden, reduces the effective stress, and hence the intergranular forces. This slows the process of the mass removal and therefore of chemical softening. In particular circumstances lowering of the effective stress may interrupt temporarily the plastic yielding, introducing elastic pauses,

during which however the dissolution continues, until the decreasing yield limit reaches anew the effective stress state. This feedback mechanism is not included in Fig. 1, which is limited to micro- and meso-scale feedbacks. The presented model is clearly idealized, many constitutive functions are taken as linear, so the actual processes may be oversimplified. Nevertheless, the discussed feedbacks and feedforwards capture the essence of the coupled processes in their basic properties.

In regard to the permeability evolution it is found that while precipitation affects the increase of stiffness relatively early in the process (Hu and Hueckel, 2007), it does not affect permeability until the process reaches a critical moment when permeability drastically drops in a short time. A practical conclusion regards the critical time to clogging that may be easily estimated from the mass dissolution rate needed to produce a solute to fill the pore.

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