Earth and Planetary Science Letters 288 (2009) 138–148

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

journal homepage: www.elsevier.com/locate/epsl

# A generalized formulation of interfacial tension driven fluid migration with dissolution/precipitation

Yasuko Takei [a,*], Saswata Hier-Majumder [b]

[a] Earthquake Research Institute, University of Tokyo, Tokyo, 113-0032, Japan
[b] Department of Geology, University of Maryland, 237 Regents Drive, College Park, MD-20742, USA

ARTICLE INFO

*Article history:*
Received 11 January 2009
Received in revised form 21 August 2009
Accepted 8 September 2009
Available online 13 October 2009

Editor: L. Stixrude

*Keywords:*
interfacial tension
dissolution/precipitation
fluid infiltration
two-phase flow

ABSTRACT

We present an extended formulation for the interfacial tension driven melt migration by taking into account dissolution/precipitation and diffusive matter transport through the liquid phase. Our results indicate that the melt migration is caused by two mechanisms. In the first mechanism, a change in melt fraction is accommodated by compaction/decompaction of solid matrix, and in the second mechanism, a change in melt fraction is accommodated by dissolution/precipitation. The latter mechanism is newly introduced in this study. As spatial scale decreases, the dominant mechanism changes from compaction/decompaction to dissolution/precipitation, and when the second mechanism is dominant, evolution of melt fraction is governed by a nonlinear diffusion equation. Therefore, critical scale of this transition is called “diffusion length $\delta_d$” below which melt fraction evolves primarily by diffusion. Diffusion length $\delta_d$ is usually smaller than the compaction length $\delta_c$. Important roles of the new mechanism are discussed on the basis of existing experimental data on melt infiltration, shear-induced melt segregation into melt-rich bands, and rehomogenization of melt in the bands due to static annealing. Melt distribution in the mantle is briefly discussed on the basis of the new model.

## 1. Introduction

Interfacial tension plays an important role in determining the grain scale geometry of melt and aqueous fluid (e.g., von Bargen and Waff, 1986; Watson and Brenan, 1987; Hier-Majumder, 2008). Also, interfacial tension driven fluid migration, or fluid infiltration, can drive melt and aqueous fluid over distances much greater than grain scale (e.g., Watson, 1982). Although interfacial tension is important at smaller scales than buoyancy force, it affects the small scale porosity structure, and hence affects the permeability of the two-phase system. Therefore, interfacial tension can significantly affect the buoyancy-driven flow. Experimental studies on the interfacial tension driven fluid migration constrain physical and chemical properties relevant to this process (e.g., Watson, 1982; Riley et al., 1990; Riley and Kohlstedt, 1991; Nakamura and Watson, 2001).

Interfacial tension driven fluid migration was first formulated by Stevenson (1986) within the framework of two-phase flow. It was predicted that fluid distribution tends to be homogenized or localized depending on whether the dihedral angle is smaller or larger, respectively, than 60°. The qualitative result of his model explains well the experimental result that liquid phase with small dihedral angles (<60°) infiltrates into dry rocks, while liquid phase with large dihedral angles (>60°) does not (e.g., Watson, 1982; Nakamura and Watson, 2001).

Another model for the interfacial tension driven fluid migration was proposed by Riley and Kohlstedt (1991). They focused on the role of compaction/decompaction of the solid matrix, which was omitted in the model of Stevenson (1986). While the rate-controlling factors in the model of Stevenson (1986) are given only by melt transport properties (permeability and melt viscosity), the rate-controlling factors in the model of Riley and Kohlstedt (1991) are given not only by the melt transport properties but also by the viscosity of the solid matrix. Models similar to Riley and Kohlstedt (1991) were also derived by Bercovici and Ricard (2003) and Hier-Majumder et al. (2006). Riley and Kohlstedt (1991) conducted a melt infiltration experiment and reported that the best agreement between the melt migration profile and the simulation generated on the basis of their theory is obtained when permeability has a melt fraction exponent of 1. Because the melt fraction exponent expected for the melt existing as grain-edge tubules is about 2, there still remains a difficulty to understand the result of this analysis consistently with the microstructural observation of equilibrium melt geometry.

The purpose of this study is to extend the existing models of interfacial tension driven fluid migration by taking into account dissolution/precipitation and diffusive matter transport through the liquid phase. Previous studies on reactive, buoyancy-driven flow in porous media demonstrated that dissolution/precipitation causes channeling instability (e.g., Aharonov et al., 1995; Spiegelman et al.,

---

* Corresponding author.
*E-mail address:* ytakei@eri.u-tokyo.ac.jp (Y. Takei).

0012-821X/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.epsl.2009.09.016

2001). However, interfacial tension was not taken into account in these formulations. In the development of equilibrium pore geometry, in which mean curvature of the solid–liquid interface is constant, important role of dissolution/precipitation and matter diffusion through liquid caused by interfacial curvature dependent solubility has been recognized (e.g., Bulau et al., 1979; von Bargen and Waff, 1986). We incorporate this effect into the interfacial tension driven flow by using Thompson–Freundlich equation (e.g., Kingery et al., 1960).

The generalized formulation proposed in this study predicts two mechanisms of interfacial tension driven fluid migration; one of the two is similar to that derived by Riley and Kohlstedt (1991), Bercovici and Ricard (2003), and Hier-Majumder et al. (2006), and the other is newly proposed in this study. Importance of the new mechanism is discussed for the experimental data of melt infiltration experiment (Riley and Kohlstedt, 1991), shear-induced melt segregation into melt-rich bands (Holtzman et al., 2003; Holtzman and Kohlstedt, 2007), and rehomogenization of melt in the bands due to static annealing (Parsons et al., 2008).

## 2. Formulation

### 2.1. Governing equations

The interfacial tension driven fluid migration is formulated within the framework of continuum mechanics. Let $\rho_L$ and $\rho_S$ be liquid and solid densities, respectively, $\mathbf{v}^L$ and $\mathbf{v}^S$ be liquid and solid velocities, respectively, $P^L$ and $\sigma_{ij}^S$ be liquid pressure (compression positive) and solid stress (tension positive), respectively, and $\phi$ be liquid volume fraction. The solid phase is assumed to consist of component A, while the liquid phase is assumed to consist of components A and B. Let $c$ be the mass fraction of component A in the liquid phase. For simplicity, both solid and liquid phases are considered to be incompressible. Hence, $\rho_L$ and $\rho_S$ are constants. The variables introduced here represent the “macroscopic” quantities, which are defined by the phasic average in Representative Elementary Volume (REV). REV is larger than the grain size but is smaller than the scales of melt migration. In comparison, we use the term “microscopic” or “local” to represent smaller scale heterogeneities than the grain size.

Mass conservations of liquid and solid phases are written as

$$\frac{\partial(\rho_L\phi)}{\partial t} + \nabla\cdot(\rho_L\phi\mathbf{v}^L) = \Gamma \quad (1)$$

$$\frac{\partial\{\rho_S(1-\phi)\}}{\partial t} + \nabla\cdot\{\rho_S(1-\phi)\mathbf{v}^S\} = -\Gamma, \quad (2)$$

where Γ (kg/m$^3$/s) represents the mass exchange rate between solid and liquid due to dissolution/precipitation (dissolution positive). Mass conservation of component A in the liquid phase is written as

$$\frac{\partial(\rho_L c\phi)}{\partial t} + \nabla\cdot(\rho_L c\phi\mathbf{v}^L) = -\nabla\cdot(\phi\mathbf{J}) + \Gamma, \quad (3)$$

where density of diffusive flux of component A in the liquid phase, **J** (kg/m$^2$/s), is given by

$$\mathbf{J} = -\rho_L D_L \nabla c, \quad (4)$$

and the diffusivity of component A in the liquid is $D_L$ (m$^2$/s). Mass conservation equation of component B in the liquid phase can be obtained by subtracting Eq. (3) from Eq. (1). The definitions of $\mathbf{v}^L$ and **J** are identical to those used in the previous studies (e.g., Aharonov et al., 1995; Spiegelman et al., 2001).

In the presence of interfacial tension, $\gamma_{sl}$, microscopic pressure field is discontinuous at the solid–liquid interface. The discontinuity $p$ (Pa) is given by

$$p = \frac{2\gamma_{sl}}{r}, \quad (5)$$

where $r$ represents the mean curvature radius $[(r_1^{-1}+r_2^{-1})/2]^{-1}$ (positive when the center of the curvature lies on the solid side). We also assume textural equilibrium within the REV. By using simple 2D models given in Fig. 1, the mean curvature radius is calculated as

$$r = \begin{cases} \dfrac{2\phi^{\frac{1}{2}}d}{f(\theta)} & \phi<\phi_c,\ \text{grains in contact} \\ 2\beta(1-\phi)^{\frac{1}{2}}d & \phi\geq\phi_c,\ \text{no intergranular contact,} \end{cases} \quad (6)$$

where $\phi_c$ is the disaggregation melt fraction above which adjacent grains loose contact. The quantities $d$ and $\theta$ represent the grain size

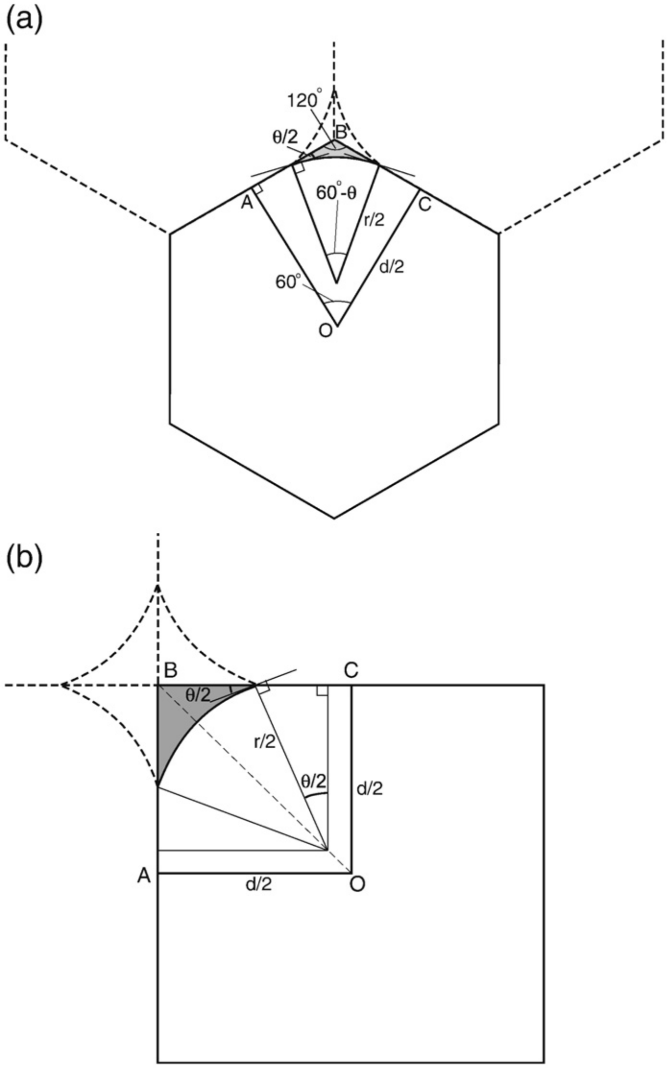

**Fig. 1.** 2D microstructural model with (a) hexagonal grains and (b) square grains. $r$ represents the mean curvature radius defined in the text. Liquid fraction $\phi$ is calculated as $\phi$ = shaded area/area of OABC.