A fractal model to describe the evolution of multiphase flow properties during mineral dissolution

Luis Guarracino [a], Tobias Rötting [b,c,*], Jesus Carrera [c]

[a] CONICET, Facultad de Ciencias Astronómicas y Geofísicas, Universidad Nacional de La Plata, Paseo del Bosque s/n, 1900 La Plata, Argentina
[b] Department of Geotechnical Engineering and Geo-Sciences, Technical University of Catalonia (UPC Barcelona Tech), c/ Jordi Girona 1-3, 08034 Barcelona, Spain
[c] Department of Geosciences, Institute of Environmental Assessment and Water Research (IDAEA), Spanish National Research Council (CSIC), c/ Jordi Girona 18, 08034 Barcelona, Spain

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ABSTRACT

Understanding the changes in multiphase flow parameters caused by mineral dissolution-precipitation is required for multiple applications ranging from geological storage of CO_2, enhanced geothermal energy production or ground water pollution. We present a physically-based theoretical model for describing the temporal evolution of porosity, saturated and relative permeabilities, retention curve and diffusion coefficient during rock dissolution by reactive fluids. The derivation of the model is based on the assumption that the pore structure of the rock can be represented by an ensemble of capillary tubes with fractal tortuosity and cumulative pore size distribution. Therefore, the model depends only on the minimum and maximum pore radii, the size of the representative elementary volume and the fractal dimensions of pore size and tortuosity, but do not need any other fitting parameters. Using this fractal description and known physical properties, we obtain analytical expressions for the hydrodynamic properties required by continuum (i.e., Darcy scale) multiphase flow models. Further, assuming periodic fluctuations in the radius of the pores, it is also possible to represent constrictivity and hysteresis. Finally, assuming a constant rate dissolution reaction it is possible to derive closed-form analytical expressions for the time evolution of porosity, retention curve, saturated and relative permeabilities and diffusion coefficient.

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1. Introduction

Mineral dissolution or precipitation may play an important role in many vadose zone and multiphase flow processes. These include the geological storage of carbon dioxide, enhanced oil or gas recovery, flow and transport in unsaturated soil, geothermal systems, etc. (e.g., [1], and citations therein). Modelling these phenomena is most frequently achieved at the continuum (Darcy) scale, which requires specifying macroscale parameters, such as porosity, saturated and relative permeabilities, retention curve and molecular diffusion coefficient. Mineral precipitation or dissolution caused by the flow of a reactive fluid produces volume and surface variations at the pore level that are translated into variations of the multiphase flow and transport properties at the macroscale (e.g., [2]). The characterization of these parameters and constitutive relationships is therefore essential for adequate understanding and modelling of the transport of reactive substances.

Most published studies on dissolution processes in porous media concentrate on the changes of porosity and saturated liquid permeability (e.g., [3]), while relationships with or between the other multiphase flow parameters have been investigated less intensively. The most widely used approach relates saturated permeability to porosity via a power law $K \propto \phi^n$. It can be derived from the equations of Kozeny [4] and Carman [5], which leads to an exponent n equal to 3. Later works, e.g., [6–9], showed that this exponent can actually vary considerably depending on the type of porous medium and the flow and transport conditions. In some cases, exponents greater than 10 [6] or even 100 [9] have been found experimentally, particularly when preferential flow paths or "wormholes" were developing in previously homogeneous and isotropic media. These relationships have been applied extensively (see [10] for a review), but they have been derived for saturated conditions and are not linked to the other multiphase flow parameters.

Two families of models can be identified that have the potential to simulate the impact of dissolution on multiphase flow parameters: Pore scale models and bundles of tubes.

Pore scale models are based on simulating flow, transport and chemical reactions at the microscale (e.g., [11–14]). In a recent

review, Meakin and Tartakovsky [14] stated that well-developed methods are available to represent saturated subsurface flow systems at the pore-scale. In practice, this is difficult, even for single phase, because it requires a detailed description of the porous structure, but it is feasible. However, the simulation of multiphase systems is much more challenging due to potentially large density or viscosity ratios of contacting fluids, and the complex behaviour of fluid-fluid-solid contact lines and their impact on dynamic contact angles. Pore-scale models can represent dissolution or precipitation and their impact on fluid flow (e.g., [15–18]). Moreover, hybrid models and other upscaling approaches have been developed for combining micro- and macroscale descriptions in order to study larger (Darcy or field) scale problems (e.g., [19–22]). However, by requiring an image of the pore space and the link between pore network and continuum scale equations, these models appear to be too complex for a widespread application in the near future.

The porous medium can also be represented by bundles of tubes to derive macroscale continuum properties. Contrary to pore network models, bundles of tubes imply perfect pore connectivity. As such, they cannot yield multicontinuum descriptions of the medium, which can be achieved with pore network models (e.g., [23–25]). The motivation of bundles of tubes lies on their simplicity. In fact, many empirical models have been developed and widely used for predicting relative permeability and retention curve assuming different pore size distributions (e.g., [26–29]). More recently, fractal distribution has often been adopted to describe the distribution of pore or particle sizes. They are based on the assumption that the soil solid phase, the soil void space or both display self-similarity (e.g., [30–33]). Tyler and Wheatcraft [33] described the soil pore size distribution using the Sierpinski Carpet [34] to develop a power-law form for the retention curve, equivalent to the functions of Brooks and Corey [26] and Campbell [35]. The Sierpinski Carpet has been also used to derive constitutive models for unsaturated flow in fractured rocks [36,37].

Fractal models of soil mass, pore volume and surface, fragmentation, soil-water retention and unsaturated hydraulic conductivity were reviewed and compared by Gimenez et al. [38] and Ghanbarian-Alavijeh et al. [39], but none of these approaches are capable of representing dissolution processes, hysteresis or diffusion.

Actual changes in hydrologic properties of aquifer media caused by chemical reactions are reviewed by Saripalli et al. [2]. They cite many different approaches to represent these changes in saturated media, i.e. geochemical equilibrium and kinetic models, chemical divide pathway models, flow and transport models, precipitation/dissolution wave theory, network models, porosity and permeability reduction models. However, they did not find any fractal approaches to model these changes, nor methods for unsaturated media or multiphase flow problems. More recently, several approaches have been proposed to model changes in porosity and permeability due to chemical reactions. Bartels et al. [40] combined the fractal porosity-permeability relationship of Pape et al. [6] with a numerical reactive transport code to calculate permeability changes in hydrogeothermal problems under fully saturated conditions from the change of bulk porosity. Freedman et al. [41] developed a film depositional model of permeability for mineral reactions in unsaturated media. They assumed that, even at partial saturation, reactions and volumetric changes occur throughout the entire pore-size distribution, and that the shape of the pore-size distribution would remain the same. Wissmeier and Barry [42] developed a selective radius shift model to describe the effect of mineral reactions on the hydraulic properties of unsaturated soils. In their model, reactions occur and pore radii change only in the smaller, water filled pores.

Hysteresis in the capillary pressure curve, i.e. the differences in water content at the same matric potential during drying and wetting, occurs due to contact angle effects, entrapped air, swelling and shrinking, and inkbottle effects, i.e. pores with narrow necks and wider bodies (e.g., [43]). Soil water hysteresis has been described using several approaches [44]: modelling by empirical and semi-empirical synthetic formulae, percolation theory, thermodynamic theory, fractal approach and domain theory. Details on many of the non-fractal approaches can be found in recent reviews [45–47]. Ojeda et al. [48] developed an early fractal model to describe hysteresis by defining a piecewise water-retention curve. Perfect [49] proposed a closed-form expression to describe the primary drainage curve of prefractal porous media. Russell and Buzzi [47] described the pores as a piecewise succession of bodies and throats and defined fractal distributions of body or throat sizes, which allowed them to describe the hysteretic loop observed during a drying–wetting–drying cycle.

Diffusion coefficients in porous media have been predicted from porosity (e.g., [50,51]) analogous to Archie's law [52]. Similar exponential relationships have also been derived experimentally [53] or using fractal approaches [54]. Analytical solutions for diffusion problems with precipitation-dissolution reactions were derived by Hayek et al. [55]. A method to predict diffusion coefficients from interfacial areas in unsaturated media represented by tortuous pore bundles was developed by Saripalli et al. [56].

From the above, it is clear that considerable advances have been achieved in the modelling of each of the processes involved in multiphase flow and reactive transport. However, no model is available that links all the involved parameters, which is a pre-requisite for modelling how these parameters change in response to changes in the pore structure caused by chemical reactions.

The objective of this work is to derive a physically-based model that describes saturated and relative permeabilities, porosity, retention curve and molecular diffusivity of a porous medium and how they change in response to dissolution processes. To this end, all these properties are obtained in terms of a fractal pore-size and tortuosity distribution, maximum and minimum radii of pores, and the size of the representative elementary volume (REV). We then present a simple theoretical model to predict the temporal evolution of these properties in response to dissolution of the solid matrix. In this model the rock is described as an impermeable matrix with cylindrical circular pores whose sizes follow a fractal distribution law. By assuming periodic fluctuations in the radius of the pores, it is possible to represent hysteresis in the retention curve and in the relative permeability function. The porous medium is then assumed to be dissolved by a fluid that reacts uniformly with the surface of the pores and that this process is far from local chemical equilibrium. We apply a function that describes the variation of the pore radii caused by dissolution or precipitation to derive continuous closed-form analytical expressions for the temporal evolution of the parameters and constitutive relationships that can be easily implemented into continuum-scale reactive transport codes.

2. Constitutive model

The rock matrix is conceptualized as an equivalent bundle of tortuous capillary tubes with a fractal law distribution of pore sizes [33,57,58]. The REV is assumed to be a straight circular cylinder of radius R and length L.

The pore structure of the REV is represented by an ensemble of tubes with periodically varying aperture, as illustrated in Fig. 1. The pore radius is described by the following sinusoidal function (similar to Czachor [59] and references therein):

$$r(x) = \bar{r} + r' \sin(2\pi x/\lambda) = \bar{r}(1 + 2a\sin(2\pi x/\lambda)) \tag{1}$$

where $\bar{r}$ is the average radius, r' the amplitude of the fluctuation, λ the wavelength, and a the fluctuation ratio defined by $a = r'/2\bar{r}$ with