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A unified model for the permeability, electrical conductivity and streaming potential coupling coefficient in variably saturated fractured media

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Luong Duy Thanh¹, Nguyen Van Nghia*¹, Phan Van Do¹, Pham Tien Du¹, Damien Jougnot² ¹ Thuyloi University, 175 Tay Son, Dong Da, Hanoi, Vietnam Sorbonne Université, CNRS, EPHE, UMR 7619 Metis, F-75005, Paris, France *** Email: nghia_nvl@tlu.edu.vn (October 13, 2022)

ABSTRACT

We present a new unified model for the permeability, electrical conductivity, and streaming 8 potential coupling coefficient in variably saturated fractured media. For those, we con-9 ceptualize the fractured medium as a partially saturated bundle of parallel capillary slits 10 with varying sizes. We assume that the fracture size distribution of the corresponding 11 medium follows a fractal scaling law, which allows us establish a pressure head-saturation 12 relationship based on the Laplace equation. We first describe the flow rate, the conduc-13 tion current, and the electrokinetic streaming current within a single fracture. Then, we 14 upscale these properties at the scale of an equivalent fractured media partially saturated 15 in order to obtain the relative permeability, the electrical conductivity and the streaming 16 potential coupling coefficient. The newly proposed model explicitly depends on pore water 17 chemistry, interface properties, microstructural parameters of fractured media, and water 18 saturation. Model predictions are in good agreement with both experimental and simulated 19 data and with another model from the literature. The results of this work constitute a useful 20 framework to estimate hydraulic properties and monitor water flow in fractured media. 21

Keywords: Fractured media; Streaming potential; Electrical conductivity; Permeability;
 Fractal

INTRODUCTION

The streaming potential (SP) is a contribution to the self-potential signal that is generated 24 by water flow in porous media. Due to the sensitivity of the SP method to subsurface water 25 flow, the SP technique has drawn an increasing attention to find or track underground 26 water in aquifers or reservoirs (e.g., Revil et al., 2012; Parsekian et al., 2015; Binley et al., 27 2015). This technique has been used for identifying and monitoring subsurface water flow 28 (e.g., Jouniaux et al., 1999; Fagerlund and Heinson, 2003; Titov et al., 2005; Aizawa et al., 29 2009), monitoring geothermal and volcanic areas (e.g., Corwin and Hoover, 1979; Finizola 30 et al., 2004; Mauri et al., 2010; Soueid Ahmed et al., 2018; Grobbe and Barde-Cabusson, 31 2019), mapping areas influenced by a contaminant plume (e.g., Martinez-Pagan et al., 2010; 32 Naudet et al., 2003; Roy, 2022), monitoring water flow in the vadose zone (e.g., Doussan 33 et al., 2002; Jougnot et al., 2015; Hu et al., 2020) or eco-hydrology (e.g., Voytek et al., 34 2019). The SP technique can be applied to estimate hydrogeological parameters of the 35 aquifer (e.g., Jardani et al., 2007; Straface et al., 2010; Revil and Jardani, 2013). 36

Fractured rocks are ubiquitous in the environment and they play a major role in a wide 37 range of geoscience issues, such as groundwater flow and contaminant transport (e.g., Neu-38 man, 2005; Medici et al., 2019), hydraulic fracturing (e.g., Osiptsov, 2017; Peshcherenko 39 et al., 2022), storage of CO₂ and nuclear waste (e.g., Bodvarsson et al., 1999; Wang and 40 Hudson, 2015; Ren et al., 2017), geothermal production (e.g., Murphy et al., 1981; Patter-41 son et al., 2020). Geophysical methods offer a variety of tools to obtain information on 42 subsurface structure and physical properties of fractured rocks. Examples of those methods 43 include the electrical conductivity imaging (e.g., Stesky, 1986; Shen et al., 2009; Roubinet 44 and Irving, 2014), seismic technique (e.g., Herwanger et al., 2004; Li, 1997; Clair et al., 45 2015), or the self potential technique (e.g., Fagerlund and Heinson, 2003; Wishart et al., 46 2006; Maineult et al., 2013). It is shown that numerical approaches are effective to char-47 acterize fractured media (e.g., Roubinet and Irving, 2014; Roubinet et al., 2016; Demirel 48

et al., 2018; Haas et al., 2013; DesRoches et al., 2018; Jougnot et al., 2020). However, to the best of our knowledge, there are only few analytical models for fractured rocks in the literature. For example, Thanh et al. (2021) proposed a model for the electrical conductivity and streaming potential coupling coefficient in fractured media under saturated conditions using a capillary bundle model following the fractal scaling law. Guarracino and Jougnot (2022) presented a model to predict the effective excess charge density for fully and partially water saturated fractured media that are described by the fractal Sierpinski carpet.

The aim of this study is to develop a unified model for the permeability, electrical 56 conductivity, and streaming potential coupling coefficient in fractured media by extend-57 ing the work proposed by Thanh et al. (2021) to partially saturated conditions. For this 58 purpose, we conceptualize a fractured medium as a partially saturated bundle of parallel 59 capillary slits following the fractal scaling law. This conceptualization allows us to de-60 termine the capillary pressure-saturation relationship and later deduce expressions for the 61 electrical conductivity and permeability of fractured media under partially saturated con-62 ditions. From the electrokinetic streaming current and conduction current within a single 63 slit, we obtain an upscaled expression for the streaming potential coupling coefficient. The 64 new obtained model explicitly depends on properties of fracture water, interface properties, 65 microstructural parameters of fractured media and water saturation. Model predictions are 66 then compared with experimental data, simulated data as well as another previous model 67 in the literature. 68

THEORETICAL BACKGROUND OF STREAMING POTENTIAL

The streaming current is caused by electrokinetic coupling, that is the drag of electrical charge by water flow in porous media conceptualized as a bundle of cylindrical capillary tubes or a bundle of capillary slits. This phenomenon is directly related to the presence of an electric double layer (EDL) that exists at the solid-water interface of the tubes or fractures (e.g., Overbeek, 1952; Hunter, 1981). This EDL contains an excess of charge in water to compensate the charge deficit of the capillary inner surface. The EDL is composed of the Stern and diffuse layers. The Stern layer only contains counter-ions, i.e., the ions

with opposite sign to the charged surface. The ions in the Stern layer can be considered 76 as immobile due to strong electrostatic attraction. The diffuse layer contains both the 77 counter-ions and co-ions, i.e., the ions with same sign as the charged surface. The ions in 78 the diffuse layer are free to move but with a net excess of charge (e.g., Hunter, 1981; Jougnot 79 et al., 2020). The interface between the Stern layer and the diffuse layer corresponds to the 80 shear plane or slipping plane that separates the stationary fluid and the moving fluid. The 81 electrical potential at this plane is called the zeta potential ζ (V) that mostly depends on 82 mineral composition of porous media, ionic strength, temperature and pH of water (e.g., 83 Hunter, 1981: Vinogradov et al., 2022b). The generated streaming current is, in turn, 84 balanced out by an electrical conduction current in the opposite direction, leading to a so-85 called streaming potential. At the steady state condition, the streaming potential coupling 86 cofficient (SPCC) is defined as (e.g., Smoluchowski, 1903; Morgan et al., 1989): 87

$$C_S = \frac{\Delta V}{\Delta P},\tag{1}$$

where ΔV (V) and ΔP (Pa) are the measured streaming potential and the imposed pressure difference across a probed medium, respectively. There have been two approaches to determine the SPCC at saturated conditions in the literature. For the first approach, the classical one, the SPCC that is expressed in terms of the zeta potential ζ is given by (e.g., Smoluchowski, 1903)

$$C_S = \frac{\epsilon_r \epsilon_0 \zeta}{\eta \sigma_w},\tag{2}$$

⁹³ where ϵ_r (no units) is the relative permittivity, ϵ_0 (F/m) is the dielectric permittivity in ⁹⁴ vacuum, η (Pa s) is the dynamic viscosity and σ_w (S/m) is the electrical conductivity of ⁹⁵ water. Eq. (2) is called the Helmholtz-Smoluchoski (HS) equation. Note that the surface ⁹⁶ electrical conductivity σ_s is not considered in Eq. (2). If σ_s is taken into consideration, ⁹⁷ Eq. (2) can be replaced by the following equation (e.g., Hunter, 1981; Ishido and Mizutani, ⁹⁸ 1981)

$$C_S = \frac{\epsilon_r \epsilon_0 \zeta}{\eta(\sigma_w + 2\frac{\Sigma_s}{\Lambda})},\tag{3}$$

⁹⁹ where Σ_s (S) is the specific surface conductance and Λ (m) is a characteristic length scale ¹⁰⁰ of porous media (Johnson et al., 1986). For the second approach, the SPCC is expressed in ¹⁰¹ terms of the effective excess charge density \hat{Q}_v (C/m³) dragged by water (e.g., Kormiltsev ¹⁰² et al., 1998; Revil and Leroy, 2004; Jougnot et al., 2020)

$$C_S = -\frac{k\widehat{Q}_v}{\eta\sigma},\tag{4}$$

where k (m²) and σ (S/m) are the permeability and electrical conductivity of fully saturated porous media, respectively.

One can note that Thanh et al. (2021) developed a model for the SPCC using the zeta potential ζ for fully saturated fractured media, whereas Guarracino and Jougnot (2022) proposed a model to predict the \hat{Q}_v , for fully and partially water saturated fractured media, that also permits to determine the SPCC.

THEORETICAL DEVELOPMENT

¹⁰⁹ Description of fractured media

Fractures in geological media exist over a wide range of scales, from microns to thousands of 110 kilometers, and fractal patterns for fractured rocks have been reported in published works 111 (e.g., Okubo and Aki, 1987; Bonnet et al., 2001; Kruhl, 2013). To derive the SPCC in 112 fractured media, we regard the geometrical description reported in the literature for frac-113 tured media which are assumed to be made up of the fractures and the surrounding matrix 114 (e.g., Tyler and Wheatcraft, 1990; Miao et al., 2015; Roubinet et al., 2016; Guarracino and 115 Jougnot, 2022). The matrix permeability is usually much smaller than that of the fractures 116 and thus the matrix can be considered as impermeable and no fluid exchange through the 117 fracture walls. Note that, for consideration of fluid transfer from the matrix to the fractures, 118 we refer readers to the work reported by Miao et al. (2019), for example. The representative 119 elementary volume (REV) is assumed to be a cuboid of length of L_o (m) and cross-section 120 area $A (m^2)$ as shown in Fig. 1. We conceptualize the fractures of the REV as a bunch 121

of parallel tortuous slits of varying aperture a (m) and width w (m) following the fractal scaling law (e.g., Tyler and Wheatcraft, 1990; Miao et al., 2015, 2019):

$$f(w) = D_f w_{\max}^{D_f} w^{-D_f - 1}, \ w_{\min} \le w \le w_{\max},$$
(5)

where D_f (no units) is the fractal dimension that is between 1 and 2 in two-dimensional spaces and it can be determined by a box-counting method (e.g., Miao et al., 2015, 2019), w_{\min} (m) and w_{\max} (m) are the smallest and largest fracture widths in the REV, respectively, representing the lower and upper bounds of the fractal distribution. Therefore, the number of fractures whose widths in the range from w to w+dw is given by f(w)dw (e.g., Majumdar and Bhushan, 1990; Miao et al., 2015). The total number of fractures, from w_{\min} to w_{\max} , is given by

$$N_t = \int_{w_{\min}}^{w_{\max}} f(w) \mathrm{d}w \approx \left(\frac{w_{\max}}{w_{\min}}\right)^{D_f}.$$
 (6)

Dividing Eq. (5) by Eq. (6), one is able to obtain the probability density function $f_r(w)$

$$f_r(w) = D_f w_{\min}^{D_f} w^{-D_f - 1}.$$
 (7)

It is shown that the aperture a is normally related to the width w by a linear scaling law (e.g., Torabi and Berg, 2011; Miao et al., 2015):

$$a = \beta w, \tag{8}$$

where β (unitless) is the proportionality coefficient called the fracture aspect ratio.

135 Hydraulic properties

¹³⁶ The porosity of the REV is defined as

$$\phi = \frac{V_p}{V_t} = \frac{\int_{w_{\min}}^{w_{\max}} (aw)(L_{\tau})f(w)dw}{L_o A} = \frac{\beta\tau D_f w_{\max}^{D_f}}{A} \int_{w_{\min}}^{w_{\max}} w^{1-D_f}dw$$

$$= \frac{\beta\tau D_f w_{\max}^2}{A(2-D_f)} (1-\alpha^{2-D_f}),$$
(9)

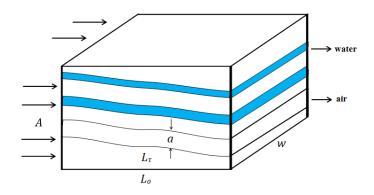


Figure 1: Schematic view of a fractured medium conceptualized as of a bunch of parallel fractures.

where V_p and V_t are the pore volume and total volume of the REV, respectively, L_o is the length of the REV, L_{τ} is the real length of the fracture, $\tau = L_{\tau}/L_o$ is the dimensionless hydraulic tortuosity of the fracture and $\alpha = w_{\min}/w_{\max}$. For the purpose of simplification, the fracture length is assumed to be unchanging with its width, hence τ is considered to be constant over the REV and therefore independent from the water saturation.

We suppose that the REV is initially filled by water and dewatered by the application of a pressure head h (m). For a capillary slit, the relationship between the fracture width w_h and the pressure head h is approximately given by (e.g., Bullard and Garboczi, 2009)

$$h = \frac{2T_s cos\theta}{\rho_w g \beta w_h},\tag{10}$$

where T_s (N/m) is the surface tension of water, θ (°) is the contact angle, ρ_w (kg/m³) is the 145 water density and $g \,(m/s^2)$ is the gravitational acceleration. A fracture is fully desaturated 146 when its width w is greater than value w_h given by Eq. (10). We assume that each fracture 147 is filled by either water or air. Therefore, in water-wet systems, water fills the fractures of 148 the smallest widths, while air occupies fractures of the largest widths when all the fractures 149 are simultaneously accessible. In other words, fractures with widths w comprised between 150 w_{\min} and w_h will be occupied by water while those with widths comprised between w_h and 151 $w_{\rm max}$ will be filled by air under application of the pressure head h. The contribution of 152

water in the REV depends on the effective water saturation S_e (unitless), that is defined as

$$S_e = \frac{S_w - S_{wr}}{1 - S_{wr}},\tag{11}$$

where S_w (unitless) is the water saturation and S_{wr} (unitless) is the residual water saturation that represents the water held as films on the fracture walls which can not be drained by the pressure head h or in non-connected fractures which cannot be taken into account in the present conceptual model.

Following a similar approach to what is reported in the literature (e.g., Guarracino, 2006; Thanh et al., 2020; Guarracino and Jougnot, 2022), S_e is expressed in terms of w_h as follows:

$$S_e = \frac{\int_{w_{\min}}^{w_h} (aw) L_{\tau} f(w) dw}{\int_{w_{\min}}^{w_{\max}} (aw) L_{\tau} f(w) dw} = \frac{w_h^{2-D_f} - w_{\min}^{2-D_f}}{w_{\max}^{2-D_f} - w_{\min}^{2-D_f}}.$$
(12)

¹⁶¹ Combining Eq. (10) and Eq. (12), the capillary pressure curve for fractured media can be ¹⁶² obtained as

$$S_e = \frac{h^{D_f - 2} - h_{\max}^{D_f - 2}}{h_{\min}^{D_f - 2} - h_{\max}^{D_f - 2}}, \ h_{\min} \le h \le h_{\max},$$
(13)

where $h_{\min} = \frac{2T_s cos\theta}{\rho_w g \beta w_{\max}}$ and $h_{\max} = \frac{2T_s cos\theta}{\rho_w g \beta w_{\min}}$. Under laminar flow conditions, the average velocity in a single fracture of aperture *a* is given by (e.g., Chung, 2010)

$$\overline{v} = \frac{\rho_w g a^2}{12\eta\tau} \frac{\Delta h}{L_o},\tag{14}$$

where Δh is the pressure head drop across the REV.

The flow rate in a single fracture follows the well-known cubic law as (e.g., Neuzil and Tracy,
1981; Klimczak et al., 2010)

$$q = \overline{v}.(aw) = \frac{\rho_w g a^3 w}{12\eta\tau} \frac{\Delta h}{L_o}.$$
(15)

¹⁶⁹ The total volumetric flow through the REV under unsaturated conditions is given by

$$q^{\text{REV}} = \int_{w_{\min}}^{w_h} qf(w) \mathrm{d}w.$$
(16)

170 Combining Eq. (5), Eq. (8), Eq. (15) and Eq. (16), one obtains

$$q^{\text{REV}} = \int_{w_{\min}}^{w_h} \frac{\rho_w g a^3 w}{12\eta \tau} \frac{\Delta h}{L_o} [D_f w_{\max}^{D_f} w^{-D_f - 1} dw] = \frac{\rho_w g \beta^3 D_f}{12\eta \tau} w_{\max}^{D_f} \frac{w_h^{4-D_f} - w_{\min}^{4-D_f}}{4 - D_f} \frac{\Delta h}{L_o}.$$
 (17)

¹⁷¹ Combining Eq. (12) and Eq. (17), one can express q^{REV} in terms of S_e as

$$q^{\text{REV}} = \frac{\rho_w g \beta^3 D_f}{12\eta \tau} w_{\text{max}}^4 \frac{\left[S_e(1 - \alpha^{2-D_f}) + \alpha^{2-D_f}\right]^{\frac{4-D_f}{2-D_f}} - \alpha^{4-D_f}}{4 - D_f} \frac{\Delta h}{L_o}.$$
 (18)

172 Following Darcy's law for Newtonian fluid flow in fractured media, q^{REV} is given by

$$q^{\rm REV} = \frac{kA}{\eta} \frac{\rho_w g \Delta h}{L_o},\tag{19}$$

where k and A are the permeability and the cross sectional area of the REV.

Combining Eq. (18) and Eq. (19), we obtain an expression for k under unsaturated conditions as

$$k(S_e) = \frac{\beta^3 D_f}{12\tau A} w_{\max}^4 \frac{\left[S_e(1 - \alpha^{2-D_f}) + \alpha^{2-D_f}\right]^{\frac{4-D_f}{2-D_f}} - \alpha^{4-D_f}}{4 - D_f}.$$
 (20)

Hence, the permeability under fully saturated conditions $(S_e = 1)$ is given by

$$k_s = \frac{\beta^3 D_f}{12\tau A} w_{\max}^4 \frac{1 - \alpha^{4 - D_f}}{4 - D_f}.$$
(21)

177 The relative permeability of fractured media, that is defined as $k_r = k(S_e)/k_s$, is given by

$$k_r(S_e) = \frac{\left[S_e(1 - \alpha^{2-D_f}) + \alpha^{2-D_f}\right]^{\frac{4-D_f}{2-D_f}} - \alpha^{4-D_f}}{1 - \alpha^{4-D_f}}.$$
(22)

It is remarked that using a classical fractal object known as the Sierpinski carpet for the fracture network in combination with the Burdine model, Guarracino (2006) obtained an expression for k_r as a function of S_e as follows:

$$k_r(S_e) = S_e^2 \frac{\left[S_e(1 - \alpha^{2-D_f}) + \alpha^{2-D_f}\right]^{\frac{4-D_f}{2-D_f}} - \alpha^{4-D_f}}{1 - \alpha^{4-D_f}}.$$
(23)

The only difference between our proposed model given by Eq. (22) and the one proposed by Guarracino (2006) given by Eq. (23) for k_r is a prefactor S_e^2 .

183 If one invokes Eq. (9), k_s can be expressed as

$$k_s = \frac{\beta^2 w_{\max}^2 \phi}{12\tau^2} \frac{1 - \alpha^{4-D_f}}{1 - \alpha^{2-D_f}} \frac{2 - D_f}{4 - D_f} = \frac{a_{\max}^2 \phi}{12\tau^2} \frac{1 - \alpha^{4-D_f}}{1 - \alpha^{2-D_f}} \frac{2 - D_f}{4 - D_f}.$$
 (24)

For $w_{max} >> w_{min}$ ($\alpha \to 0$) that is normally reported to be satisfied for the fractal fractured media (e.g., Guarracino, 2006; Miao et al., 2015, 2019; Thanh et al., 2021), Eq. (24) reduces to

$$k_s = \frac{\beta^2 w_{\max}^2 \phi}{12\tau^2} \frac{2 - D_f}{4 - D_f} = \frac{a_{\max}^2 \phi}{12\tau^2} \frac{2 - D_f}{4 - D_f}.$$
(25)

187 Electrical conductivity

Following Thanh et al. (2021), the total electrical conductivity in a single fracture with consideration of the surface conductivity is given by

$$\sigma_f(w) = \sigma_w \frac{\beta w^2}{A\tau} + \sum_s \frac{2(1+\beta)w}{A\tau}.$$
(26)

Recall that σ_w (S/m) and Σ_s (S) are the electrical conductivity of water and specific surface conductance at the solid–water interface as previously mentioned.

The total electrical conductivity of considered porous media under unsaturated conditions
is therefore obtained by

$$\sigma = \int_{w_{\min}}^{w_h} \sigma_f(w) f(w) \mathrm{d}w.$$
(27)

¹⁹⁴ Combining Eq. (5), Eq. (26) and Eq. (27) yields the following:

$$\sigma = \frac{1}{A\tau} \left\{ \sigma_w \beta D_f w_{\max}^{D_f} \frac{w_h^{2-D_f} - w_{\min}^{2-D_f}}{2 - D_f} + 2\Sigma_s (1+\beta) D_f w_{\max}^{D_f} \frac{w_h^{1-D_f} - w_{\min}^{1-D_f}}{1 - D_f} \right\}.$$

$$= \frac{\beta D_f w_{\max}^{D_f} (w_h^{2-D_f} - w_{\min}^{2-D_f})}{A(2 - D_f)\tau} \left\{ \sigma_w + \frac{2(1+\beta)\Sigma_s}{\beta} \frac{2 - D_f}{1 - D_f} \frac{w_h^{1-D_f} - w_{\min}^{1-D_f}}{w_h^{2-D_f} - w_{\min}^{2-D_f}} \right\}.$$
(28)

Invoking Eq. (12), the σ under unsaturated conditions is expressed in terms of S_e as

$$\sigma = \frac{\beta D_f w_{\max}^2 S_e(1 - \alpha^{2 - D_f})}{A(2 - D_f)\tau} \left\{ \sigma_w + \frac{2(1 + \beta)\Sigma_s}{\beta w_{\max}} \frac{2 - D_f}{1 - D_f} \frac{\left[S_e(1 - \alpha^{2 - D_f}) + \alpha^{2 - D_f}\right]^{\frac{1 - D_f}{2 - D_f}} - \alpha^{1 - D_f}}{S_e(1 - \alpha^{2 - D_f})} \right\}$$
(29)

¹⁹⁶ Substituting A from Eq. (9) into Eq. (28), the σ is obtained as

$$\sigma = \frac{\phi S_e}{\tau^2} \left\{ \sigma_w + \frac{2(1+\beta)\Sigma_s}{\beta w_{\max}} \frac{2-D_f}{1-D_f} \frac{\left[S_e(1-\alpha^{2-D_f})+\alpha^{2-D_f}\right]^{\frac{1-D_f}{2-D_f}}-\alpha^{1-D_f}}{S_e(1-\alpha^{2-D_f})} \right\}.$$
 (30)

Eq. (30) shows the dependence of σ under unsaturated conditions on microstructural parameters of the fractured media $(D_f, \phi, \alpha, \beta, w_{max}, \tau)$, water electrical conductivity σ_w , specific surface conductance Σ_s , and effective water saturation S_e .

200 Streaming potential coupling coefficient

201 Streaming current through the REV

²⁰² Under the thin EDL assumption in which the Debye length is small compared to fracture ²⁰³ widths and the Debye-Hückel approximation that is applicable for small values of ζ (i.e., 50 ²⁰⁴ mV) for a binary symmetric 1:1 electrolyte, for example (e.g., Rice and Whitehead, 1965; ²⁰⁵ Pride, 1994), the electrokinetic streaming current in a single fracture due to transport of ²⁰⁶ excess charge in the EDL by water flow is given by (e.g., Thanh et al., 2021)

$$i_s(w) = -\frac{\epsilon_r \epsilon_o \zeta}{\eta} \frac{wa}{\tau} \frac{\rho_w g \Delta h}{L_o} = -\frac{\epsilon_r \epsilon_o \zeta}{\eta} \frac{\beta w^2}{\tau} \frac{\rho_w g \Delta h}{L_o}.$$
(31)

²⁰⁷ The total streaming current through the REV under unsaturated conditions is determined²⁰⁸ by

$$I_s = \int_{w_{\min}}^{w_h} i_s(w) f(w) \mathrm{d}w \tag{32}$$

²⁰⁹ From Eq. (5), Eq. (31) and Eq. (32), the following is obtained

$$I_{s} = -\frac{\epsilon_{r}\epsilon_{o}\zeta}{\eta\tau}\frac{\rho_{w}g\Delta h}{L_{o}}(\beta D_{f}w_{\max}^{D_{f}})\int_{w_{\min}}^{w_{h}}w^{1-D_{f}}dw$$

$$= -\frac{\epsilon_{r}\epsilon_{o}\zeta\beta D_{f}}{\eta\tau}\frac{w_{\max}^{D_{f}}}{(2-D_{f})}(w_{h}^{2-D_{f}}-w_{\min}^{2-D_{f}})\frac{\rho_{w}g\Delta h}{L_{o}}.$$
(33)

²¹⁰ Conduction current through the REV

The streaming current is accounted for the streaming potential ΔV that is built up across the REV due to the water flow. In turn, an electric conduction current is generated in the REV due to the electrical potential difference ΔV . Namely, the conduction current in a single fracture is given by Thanh et al. (2021)

$$i_c(w) = \left[\sigma_w \frac{\beta w^2}{L_o \tau} + \Sigma_s \frac{2(1+\beta)w}{L_o \tau}\right] \Delta V.$$
(34)

The total electric conduction current through the REV under unsaturated conditions is given by

$$I_{c} = \int_{w_{\min}}^{w_{h}} i_{c}(w)f(w)dw$$

= $\frac{\beta D_{f}w_{\max}^{D_{f}}(w_{h}^{2-D_{f}} - w_{\min}^{2-D_{f}})}{(2-D_{f})\tau} \left[\sigma_{w} + \frac{2(1+\beta)\Sigma_{s}}{\beta}\frac{2-D_{f}}{1-D_{f}}\frac{w_{h}^{1-D_{f}} - w_{\min}^{1-D_{f}}}{w_{h}^{2-D_{f}} - w_{\min}^{2-D_{f}}}\right] \frac{\Delta V}{L_{o}}.$ (35)

217 Streaming potential coupling coefficient

218 Considering thermodynamic equilibrium, the following condition is satisfied

$$I_s + I_c = 0. ag{36}$$

²¹⁹ Consequently, the SPCC is given by

$$C_S = \frac{\Delta V}{\Delta P} = \frac{\Delta V}{\rho_w g \Delta h} = \frac{\epsilon_r \epsilon_0 \zeta}{\eta \left[\sigma_w + \frac{2(1+\beta)\Sigma_s}{\beta} \frac{2-D_f}{1-D_f} \frac{w_h^{1-D_f} - w_{\min}^{1-D_f}}{w_h^{2-D_f} - w_{\min}^{2-D_f}} \right]}.$$
(37)

²²⁰ Invoking Eq. (12), the SPCC can be expressed in terms of S_e as

$$C_{S} = \frac{\epsilon_{r}\epsilon_{0}\zeta}{\eta \left[\sigma_{w} + \frac{2(1+\beta)\Sigma_{s}}{\beta w_{\max}} \frac{2-D_{f}}{1-D_{f}} \frac{\left[S_{e}(1-\alpha^{2-D_{f}})+\alpha^{2-D_{f}}\right]^{\frac{1-D_{f}}{2-D_{f}}}}{S_{e}(1-\alpha^{2-D_{f}})}\right]}.$$
(38)

Eq. (38) is an expression for the SPCC of fractured media under unsaturated conditions. It predicts that the SPCC is dependent of water properties (σ_w , ϵ_r and η), physico-chemical properties of the solid-water interface (Σ_s and ζ), microstructural parameters of fractured media (D_f , ϕ , α , β , w_{max}) and saturation state (S_e). When $\Sigma_s = 0$, Eq. (38) reduces to the HS equation given by Eq. (2) that has been proposed for porous media rather than fractured media regardless of S_e . Under saturated conditions $S_e = 1$, Eq. (38) simplifies to that proposed by Thanh et al. (2021):

$$C_S = \frac{\epsilon_r \epsilon_0 \zeta}{\eta \left[\sigma_w + \frac{2(1+\beta)\Sigma_s}{\beta w_{\max}} \frac{2-D_f}{1-D_f} \frac{1-\alpha^{1-D_f}}{1-\alpha^{2-D_f}} \right]}.$$
(39)

Combining Eq. (4), Eq. (30) and Eq. (38), one can infer an expression for the effective excess charge density as following

$$\widehat{Q}_v = -\epsilon_r \epsilon_0 \zeta \frac{\phi S_e}{k\tau^2}.$$
(40)

We remark that Guarracino and Jougnot (2022) proposed a model for \widehat{Q}_v , that is deduced from their Eq. (14) and Eq. (23), as below

$$\widehat{Q}_v = -\epsilon_r \epsilon_0 \zeta \left[1 + \frac{1}{54} \left(\frac{e\zeta}{k_B T} \right)^2 \right] \frac{\phi S_e}{k},\tag{41}$$

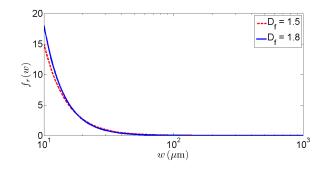


Figure 2: Probability density function associated with fracture size distribution in this work in the width range from $w_{\min} = 10 \ \mu \text{m}$ to $w_{\max} = 1000 \ \mu \text{m}$ ($\alpha = 0.01$) for two values of D_f (1.5 and 1.8).

where e (C) the elementary charge, k_B (J/K) the Boltzman constant and T (K) is the absolute temperature.

²³⁴ Under the Debye-Hückel approximation in which $\left(\frac{e\zeta}{2k_BT}\right)^2 << 1$ (e.g., Pride, 1994), Eq. ²³⁵ (41) reduces to

$$\widehat{Q}_v = -\epsilon_r \epsilon_0 \zeta \frac{\phi S_e}{k}.$$
(42)

Obviously, our finding given by Eq. (40) is the same as that proposed by Guarracino and Jougnot (2022) under the Debye-Hückel approximation given by Eq. (42). It is noted that Guarracino and Jougnot (2022) did not consider τ in their model (i.e., $\tau = 1$).

RESULTS AND DISCUSSION

239 Sensitivity of the model

Figure 2 shows the representative probability density function of fractures predicted from Eq. (7) in the width range from $w_{\min} = 10 \ \mu m$ to $w_{\max} = 1000 \ \mu m$ ($\alpha = 0.01$) for two different values of D_f (1.5 and 1.8). It is seen that: (i) the frequency distribution of fractures becomes skewed toward smaller fracture width and (ii) there is a larger number of small fractures for larger value of D_f .

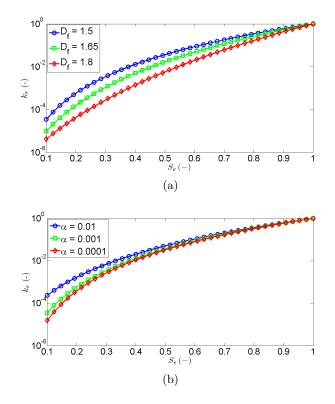


Figure 3: Variation of the k_r with S_e predicted from Eq. (22): (a) for three representative values of D_f (1.5, 1.65 and 1.8) at a given representative value of $\alpha=0.001$, (b) for three representative values of α (0.01, 0.001 and 0.0001) at a given representative value of $D_f = 1.5$.

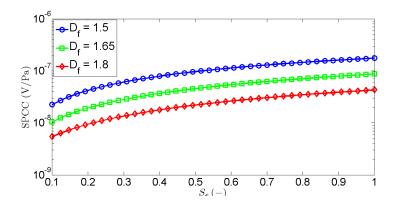


Figure 4: Dependence of the SPCC in magnitude on the effective water saturation for different values of D_f (1.6, 1.65 and 1.8) predicted from Eq. (38)

From Eq. (22), we can predict the variation of k_r with S_e for fractured media. For 245 example, Fig. 3 shows the S_e - k_r relationship for: (a) three representative values of D_f (1.5, 246 1.65 and 1.8) at a given representative value of $\alpha = 0.001$, (b) three representative values of 247 α (0.01, 0.001 and 0.0001) at a given representative value of $D_f = 1.5$. It is seen that k_r is 248 sensitive to parameters of S_e , D_f and α . At given values of S_e and α , k_r decreases with an 249 increase of D_f . The reason is that when D_f increases, the number of fractures in the REV 250 with small widths increases as indicated in Fig. 2. Therefore, at the same water saturation, 251 w_h decreases and the total flow rate through the REV becomes smaller. Consequently, 252 k_r decreases with increasing D_f . It is also predicted that at given values of S_e and D_f , 253 k_r decreases with a decrease of α . The reason is that when α decreases, there is a larger 254 fraction of fractures with smaller widths due to the property of the fractal distribution. As 255 a result, k_r decreases. 256

Figure 4 shows the variation of the SPCC in magnitude with S_e for different values of D_f (1.6, 1.65 and 1.8) predicted from Eq. (38). Representative parameters used in Eq. (38) are: $\sigma_w = 0.02$ S/m, $\Sigma_s = 10^{-9}$ S, $\zeta = -0.030$ V, $w_{\text{max}} = 200.10^{-6}$ m and $\beta = 0.01$. It is shown that the SPCC in magnitude increases with increasing S_e . This prediction is in good agreement with those observed in published work but for porous media such as sand columns, dolomite cores or limestone cores (e.g., Guichet et al., 2003; Revil and Cerepi,

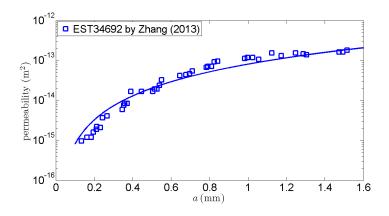


Figure 5: Variation of the k_s of a single fracture as a function of aperture a obtained from Zhang (2013) for the sample EST34692 and prediction from Eq. (25) for a fractured medium with $D_f = 1.8$, $\tau = 1.2$, $\phi = 0.025$ and $a_{\text{max}} = a/40$.

263 2004; Vinogradov and Jackson, 2011). Additionally, it is also seen that the SPCC decreases 264 with increasing D_f for the same value of S_e . The reason is that when D_f increases, the 265 number of fractures characterized by relatively small widths increases as shown by Fig. 2. 266 Hence, the surface conductivity of fractured media increases and the SPCC in magnitude 267 decreases. It is remarked that the surface conductivity of fractured media is dominated by 268 the contribution from the smaller width fractures for given values of σ_w and Σ_s .

²⁶⁹ Comparison with published data

There are not many published experimental data of single fracture permeability measure-270 ment in the literature. Nevertheless, Zhang (2013) presents gas permeability measurement 271 on a Callovo-Oxfordian (COx) clayrock sample which exhibits a single fracture. Non frac-272 tured COx clay rocks are known for their very low permeability (typically 10 nd at saturation 273 as indicated by Jougnot et al. (2010)). Hence, the measured permeability of a fractured 274 sample is largely due to the fracture itself. Fig. 5 shows the evolution of the sample per-275 meability as a function of the aperture a of the fracture. The variation of the fractured 276 medium k_s under saturated conditions with a_{max} is predicted from Eq. (25) as shown by 277 the solid line in Fig. 5. The model can describe the behavior of the permeability well with 278

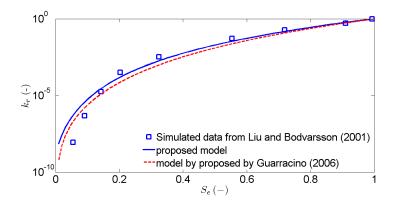


Figure 6: Variation of k_r with S_e simulated by Liu and Bodvarsson (2001) for two dimensional fracture networks (symbols) and corresponding predictions from Eq. (22) and Eq. (23.)

 $D_f = 1.8, \tau = 1.2, \phi = 0.025, a_{\text{max}} = a/40$. It should be remarked that the fitting parameters (D_f, τ, ϕ) are optimized using a function "fmincon" in Matlab to seek a minimum root-mean-square error between the experimental data and predicted values.

Figure 6 shows the variation of k_r with S_e simulated by Liu and Bodvarsson (2001) for 282 two dimensional fracture networks (symbols). This observation can be predicted by Eq. 283 (22) with optimized parameters $D_f = 1.6$ and $\alpha = 2.5 \times 10^{-4}$ (solid line). As pointed out 284 by Guarracino (2006), the simulated data in Fig. 6 can also be reproduced by Eq. (23)285 with $D_f = 1.5$ and $\alpha = 0.01$ (dashed line). It is seen that the proposed model is in very 286 good agreement with simulated data and the model proposed by Guarracino (2006) (The 287 root mean square deviation (RMSD) of the proposed model and that of Guarracino (2006) 288 are calculated to be 0.0241 and 0.0254, respectively). 289

Similarly, Fig. 7 shows the variation of k_r with S_e for the fractured wellbore cement measured by Rod et al. (2019) (symbols) and corresponding predictions from our model and Guarracino (2006). The fitting parameters are optimized in the same way as previously mentioned are $D_f = 1.2$, $\alpha = 0.001$ and $D_f = 1.1$, $\alpha = 0.01$ for Eq. (22) and Eq. (23), respectively. One can see that our proposed model (RMSD = 0.0985) can provide a better fit than Guarracino (2006) (RMSD = 0.1469). The main reason may be come from the

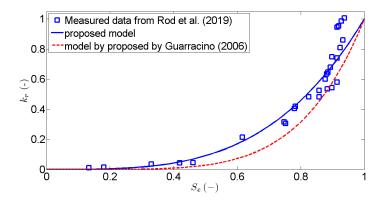


Figure 7: Variation of k_r with S_e for the fractured welbore cement measured by Rod et al. (2019) (symbols) and corresponding predictions from Eq. (22) and Eq. (23.

difference in the prefactor S_e^2 in Eq. (23) compared to Eq. (22). Therefore, k_r predicted from Guarracino (2006) is lower than our model, especially at low values of S_e . Note that S_e is always less than 1.

To the best of our knowledge, there have been only few publications on SPCC mea-299 surements of fractured media under saturated conditions (e.g., Moore and Glaser, 2007; 300 Vinogradov et al., 2022a) and no publications under unsaturated conditions. For example, 301 Fagerlund and Heinson (2003) measured the zeta potential of fractured rocks by crushing 302 rocks and packing obtained crushed material into a tube. Hence, the experimental data for 303 crushed material reported by Fagerlund and Heinson (2003) is not applicable for our model. 304 Vinogradov et al. (2022a) measured the SPCC for a fractured Lewisian gneiss sample which 305 was assumed to have a single fracture at different values of confining pressure and ionic 306 strengths. For example, the measured values for the SPCC at two different values of confin-307 ing pressures (4 MPa and 7 MPa) for the ionic strength of NaCl of 0.7 M, that is denoted 308 by C_f were reported to be -1.21 mV/MPa and -1.23 mV/MPa, respectively. The pore elec-309 trical conductivity σ_w can be estimated by the relationship $\sigma_w = 10C_f = 7$ S/m (e.g., Sen 310 and Goode, 1992). For this high value of σ_w , the contribution of the surface conductivity 311 to the total effective conductivity can be neglected (e.g., Alkafeef and Alajmi, 2006; Thanh 312

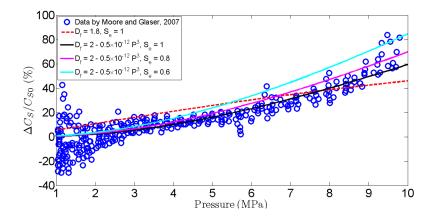


Figure 8: Variation of the relative SPCC difference with injection pressure $\Delta C_S(P)/C_{S0}$. The symbols are experimental data reported by Moore and Glaser (2007). The best fit of the proposed model to experimental data are displayed by lines.

and Sprik, 2016). Therefore, we can apply Eq.(39) without consideration of the term of $\frac{2(1+\beta)\Sigma_s}{\beta w_{\max}} \frac{2-D_f}{1-D_f} \frac{1-\alpha^{1-D_f}}{1-\alpha^{2-D_f}}$ to determine the ζ . The obtained values of the ζ are -11.8 mV and -12.0 mV for the confining pressures of 4 MPa and 7 MPa, respectively. This finding is in good agreement with the result of Vinogradov et al. (2022a) where ζ was reported to be -10.52 mV and -10.69 mV, respectively.

Additionally, Moore and Glaser (2007) measured the relative SPCC difference as a 318 function of injection pressures for microcracked Sierra granite samples during hydraulic 319 fracturing in the laboratory as shown in Fig. 8 (symbols). It is remarked that the relative 320 SPCC difference is defined as $\Delta C_S(P)/C_{S0}$, where C_{S0} is the SPCC at zero pressure drop. 321 It is seen that there is a tendency of increase of the SPCC with pressure. The reason is 322 related to an increase of dilatancy of microcracks with increasing pressures, which causes an 323 increase in permeability and therefore in apertures. The increase of apertures of fractured 324 media results in a decrease of the surface electrical conductivity and hence the SPCC. 325 Moore and Glaser (2007) showed the variation of the permeability k_s with pressure drop P 326 as follows: 327

$$k_s = 10^{-18} e^{2.5 \times 10^{-4} P},\tag{43}$$

³²⁸ where k_s is in m² and P is in kPa.

Combining Eq. (24) and Eq. (43), one can obtain w_{max} as a function of P as following:

$$w_{\rm max} = \chi \sqrt{k_s} = \chi 10^{-9} e^{1.25 \times 10^{-4} P},\tag{44}$$

330 where
$$\chi = \frac{\tau}{\beta} \sqrt{\frac{12(1-\alpha^{2-D_f})(4-D_f)}{\phi(1-\alpha^{4-D_f})(2-D_f)}}$$
.

From Eq. (39) and Eq. (44), one can predict the $\Delta C_S/C_{S0}$ as a function of P for 331 the microcracked samples reported by Moore and Glaser (2007) as shown by the solid line 332 in Fig. 8. Input parameters for the prediction are shown in Table 1 where superscripts * 333 stands for values measured by Moore and Glaser (2007) and superscripts ⁺ stands for fitting 334 parameters. Namely, σ_w , ϕ , and ζ are reported to be 0.015 S/m, 0.009 (unitless) and -34 335 mV, respectively. Due to constraints associated with a large number of model parameters 336 for $\Delta C_S/C_{S0}$, we search for those parameters that provide a relatively good fit by a trial-337 and -error method. The obtained values for τ , Σ_s , D_f , β , and α are 2 (unitless), 0.17×10^{-10} 338 S. 1.8 (unitless), 0.01 (unitless), and 0.001 (unitless), respectively, it is noted that the found 339 fitting parameters are in the ranges normally reported in literature for fractured rocks. For 340 example, τ was reported between 1.1 and 30 (e.g., Wang et al., 2022; Violay et al., 2010; 341 Roubinet et al., 2018). Using the box-counting technique, D_f was found between 1.2 and 342 1.85 (e.g., Walsh and Watterson, 1993; Roy et al., 2007). The surface conductance Σ_s was 343 reported in the range from 0.1×10^{-9} S to 4×10^{-9} S for silica surface in contact with NaCl 344 electrolytes (e.g., Thanh et al., 2019; Revil and Glover, 1998). Ghanbarian et al. (2019) 345 found β in the range from 0.001 to 0.1 for tensile fractures in the Krafla fissure swarm of 346 northeast Iceland. Additionally, Miao et al. (2015) also used $\beta = 0.002$ for fitting their 347 model with simulated data. Values of α were inferred between 0.0001 and 0.01 for fractured 348 carbonate core rock samples (e.g., Erol et al., 2017). Fig. 8 shows that the proposed model 349 can produce the key behavior of experimental data. We remark that the model can provide 350 a better fit if one takes into account the variation of D_f with P (Guarracino and Jougnot, 351 2022). For example, the relationship $D(P) = 2 - 0.5 \times 10^{-12} P^3$ provides a better fit to the 352 experimental data as shown in Fig. 8. We also illustrate variations of $\Delta C_S/C_{S0}$ with P 353

Table 1: Input parameters for the microcracked samples reported by Moore and Glaser (2007). Note that superscripts * stands for values measured by Moore and Glaser (2007) and superscripts + stands for fitting parameters

Parameter	Value	\mathbf{Units}
$\sigma^*_w \ \phi^*$	0.015	S/m
ϕ^*	0.009	unitless
$ au^+$	2	unitless
ζ^*	-34	mV
Σ_s^+	0.17×10^{-10}	\mathbf{S}
D_f^+	1.8	unitless
β^{+}	0.01	unitless
α^+	0.001	unitless

predicted from the proposed model for other effective saturations S_e (0.8 and 0.6) as shown by the colored solid lines in Fig. 8.

CONCLUSIONS

We present a new unified model for the permeability, electrical conductivity, and streaming 356 potential coupling coefficient in variably saturated fractured media. For those, we con-357 ceptualize the fractured medium as a bundle of parallel capillary fractures or slits with 358 varying sizes that is partially saturated. We assume that the fracture size distribution of 359 the corresponding medium follows the fractal scaling law, therefore allowing us to determine 360 the pressure head-water saturation relationship. From the flow rate, conduction current, 361 and electrokinetic streaming current within a single saturated fracture, we can upscale ex-362 pressions for the permeability, relative permeability, electrical conductivity, and streaming 363 potential coupling coefficient for fractured media under partially saturated conditions at 364 the REV scale. This new unified model explicitly depends on properties of fracture water 365 $(\sigma_w, \epsilon_r \text{ and } \eta)$, interface properties $(\Sigma_s \text{ and } \zeta)$, microstructural parameters of fractured 366 media $(D_f, \phi, \alpha, \beta, w_{\text{max}})$ and saturation state (S_e) . Model predictions are in good with 367 experimental data, simulated data as well as another previous model in the literature. This 368 newly proposed model constitutes a practical framework to estimate hydraulic properties 369 and monitor water flow in fractured media based on self potential measurements and pos-370

³⁷¹ sibly monitor fracking processes.

DATA AVAILABILITY STATEMENT

- ³⁷² The data underlying this article will be shared on reasonable request to the corresponding
- 373 author.

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