RAPID IMBIBITION OF WATER IN FRACTURES WITHIN UNSATURATED SEDIMENTARY ROCK

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ABSTRACT

The spontaneous imbibition of water and other liquids into gas-filled fractures in variably-saturated porous media is important in a variety of engineering and geological contexts. However, surprisingly few studies have investigated this phenomenon. We present a theoretical framework for predicting the 1-dimensional movement of water into air-filled fractures within a porous medium based on early-time capillary dynamics and spreading over the rough surfaces of fracture faces. The theory permits estimation of sorptivity values for the matrix and fracture zone, as well as a dispersion parameter which quantifies the extent of spreading of the wetting front. Quantitative data on spontaneous imbibition of water in unsaturated Berea sandstone cores were acquired to evaluate the proposed model. The cores with different permeability classes ranging from 50 to 500 mD and were fractured using the Brazilian method. Spontaneous imbibition in the fractured cores was measured by dynamic neutron radiography at the Neutron Imaging Prototype Facility (beam line CG-1D, HFIR), Oak Ridge National Laboratory. Water uptake into both the matrix and the fracture zone exhibited square-root-of-time behavior. The matrix sorptivities ranged from 2.9 to 4.6 mm s^{-0.5}, and increased linearly as the permeability class increased. The sorptivities of the fracture zones ranged from 17.9 to 27.1 mm s^{-0.5}, and increased linearly with increasing fracture aperture width. The dispersion coefficients ranged from 23.7 to 66.7 mm² s⁻¹ and increased linearly with increasing fracture aperture width and damage zone width. Both theory and observations indicate that fractures can significantly increase spontaneous imbibition in unsaturated sedimentary rock by capillary action and surface spreading on rough fracture faces. Fractures also increase the dispersion of the wetting front. Further research is needed

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1 to investigate this phenomenon in other natural and engineered porous
2 media.

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4 **KEYWORDS**

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6 Neutron radiography; fractured porous media; unsaturated flow; capillarity;
7 fluid spreading; rough surfaces

1. INTRODUCTION

The movement of water and other liquids in fractures within unsaturated earth materials is important in a variety of contexts, including the storage of hazardous wastes within the deep vadose zone, enhanced oil recovery, shale gas extraction, and water damage to the foundations of buildings and other structures. Advances in neutron, nuclear magnetic resonance, and x-ray imaging technologies have significantly improved our experimental capabilities to visualize fracture flow in variably-saturated porous media. As a result, researchers are now able to routinely quantify the distribution and movement of water within a wide range of natural and engineered materials at the spatial and temporal resolutions relevant to fracture flow.

While a large body of literature exists documenting water entry into gas-filled fractures under positive water pressures (e.g., Chen and Kinzelbach, 2002; Rangel-German and Kovscek, 2002; Karpyn et al., 2009; Kanematsu et al., 2009; Polsky et al., 2013), surprisingly few studies have investigated water uptake into fractures when the gauge pressure of the water is zero or less. These studies have involved either fractured cementitious materials (concrete, engineered cementitious composites, and mortar) or rocks (granodiorite and sandstone).

The rapid penetration of water into individual air-filled fractures in steel-reinforced concrete was visualized using neutron imaging by Zhang et al. (2010a). Şahmaran and Li (2009) found through sorptivity testing that micro-cracks induced by mechanical loading increased the rate of water uptake in engineered cementitious composites. Zhang et al. (2010b; 2011) confirmed this finding using neutron radiography, while Snoeck et al. (2012) and Van Tittelboom et al. (2013) employed the same method to investigate the crack sealing efficiency of various water repellent chemicals on capillary uptake in fractured mortar mixtures.

1 An early neutron imaging study of water imbibition into a granodiorite
2 rock core containing a fault-gouge filled fracture was conducted by Lunati et
3 al. (2003). Due to the thickness of the core, however, a relatively long
4 exposure time (~40 s per image) was needed to obtain a reasonable spatial
5 resolution. Hall (2013) used neutron radiography to quantify differential
6 water uptake into an initially air-dry sandstone core that had been subjected
7 to triaxial compression until just after macroscopic failure. Local fluid flow
8 velocities, extracted from image analysis of the neutron radiographs,
9 indicated that water movement was fastest within compacted (low porosity)
10 shear bands. This behavior was attributed to higher capillary forces within
11 the shear bands, possibly due to increased micro-crack density associated
12 with localized damage.

13 Some of the above observations have been qualitatively interpreted in
14 terms of the dynamics of capillary uptake (Wittman et al., 2010). To date,
15 however, no theoretical framework has been proposed that allows for a
16 quantitative explanation of this phenomenon. The major objectives of this
17 research were to: (i) present a theoretical model for predicting the 1-
18 dimensional movement of water into a single air-filled fracture within a
19 porous medium, and (ii) acquire quantitative data on spontaneous imbibition
20 of water in unsaturated fractured Berea sandstone cores using dynamic
21 neutron radiography with which to evaluate the proposed model.

2. THEORETICAL FRAMEWORK

25 The dynamics of water displacing air within a single straight capillary tube
26 are well established (Washburn, 1921). Washburn's model was derived by
27 combining the Young-Laplace equation for the pressure drop across the
28 interface between two static fluids, with the Hagen-Poiseuille equation for
29 the pressure drop in a fluid flowing through a cylindrical tube. Cai et al.

(2010) modified this basic model to predict the dynamics of capillary rise in a single tortuous capillary tube.

Here we represent a porous medium as a bundle of non-interacting tortuous capillary tubes, replacing the single tube diameter, λ , in Cai et al. (2010) with an effective diameter, $\langle\lambda\rangle$, representing the central tendency of the tube diameters within the capillary bundle, i.e.

$$\frac{dL}{dt} = \frac{\langle\lambda\rangle\sigma\cos\theta}{8\mu\tau^2} \frac{1}{L} - \frac{\rho g\langle\lambda\rangle^2}{32\mu\tau^2} \quad [1]$$

where L is the height of the wetting front at time t , σ is the air-water surface tension, θ is the water-solid contact angle, μ is the viscosity of water, ρ is the density of water, τ is the tortuosity, and g is the gravitational acceleration. Integrating Eq. [1] from $L = 0$ at $t = 0$ to L at time t yields:

$$t = -\frac{A}{B^2} \ln\left(1 - \frac{B}{A}L\right) - \frac{L}{B} \quad [2]$$

where $A = \langle\lambda\rangle\sigma\cos\theta/(8\mu\tau^2)$ and $B = \rho g\langle\lambda\rangle^2/(32\mu\tau^2)$. At early times, the effects of gravity are insignificant so that the last term on the right-hand side of Eq. [1] can be neglected. Integrating the resulting expression produces a linear relationship between the wetting front elevation and the square root of time, i.e.

$$L = C\sqrt{t} \quad [3]$$

where $C = \sqrt{\frac{\langle\lambda\rangle\sigma\cos\theta}{4\mu\tau^2}}$ is a constant commonly referred to as the sorptivity (Philip, 1957; Hall, 1989; Reda Taha et al., 2001).

The brittle failure of a loaded porous material can be expected to result in an increase in the effective diameter, $\langle\lambda\rangle$, of the fracture zone as compared

to the unfractured matrix. The tortuosity, τ , of the fracture zone is also likely to be decreased relative to that in the unfractured matrix. The impact of such changes on rates of capillary rise within these two regions can be predicted from Eqs. [2] and [3] as illustrated in Fig. 1. Although increasing $\langle \lambda \rangle$ reduces the late-time equilibrium height of capillary rise in the fracture zone relative to that in the unfractured matrix, water imbibition is markedly increased at early times. This effect is magnified when there is a corresponding reduction in tortuosity of the fracture zone (Fig. 1). Thus, capillary dynamics can explain the more rapid uptake of water in a fracture than in the surrounding matrix during spontaneous imbibition.

In addition to capillary uptake within a fracture aperture, rapid wetting can also occur due to surface spreading of water on the side walls of the fracture. This phenomenon was first demonstrated by Tokunaga and Wan (2001), and several recent experimental studies (e.g., Ishino et al., 2007; Vorobyev and Guo, 2010; Liu et al., 2011) have documented the spreading of various liquids over surfaces with different roughness attributes. Hay and Dragila (2008) and Hay et al. (2008) have proposed a theoretical model to explain the physics of fluid spreading on rough surfaces.

We now derive an equation for the rapid early time uptake of water within an air-filled fracture that incorporates both capillarity and surface spreading. Assume L is the height of the wetting front at time t in a smooth-walled fracture. Then the total height of the wetting front, x , at time t due to the combination of capillary dynamics and surface spreading effects in a rough-walled fracture is given by (Hay et al., 2008):

$$x = D\sqrt{t} + L \quad [4]$$

1 where D is a constant of proportionality that is strongly dependent on the
2 roughness geometry (Hay et al. 2008). Substituting Eq. [3] into Eq. [4] and
3 simplifying the result gives:

$$x = D\sqrt{t} + C\sqrt{t} = E\sqrt{t} \quad [5]$$

7 where $E = (C + D)$ is a lumped sorptivity parameter that combines the
8 effects of both aperture capillarity and the wetting of rough side walls.

9 Lenormand (1990) studied the pore-scale interfacial geometries resulting
10 from immiscible displacement of one fluid by another in a porous medium.
11 Based on the relative dominance of viscous versus capillary effects the
12 displacement can be categorized as of one of the following types: viscous
13 fingering, capillary fingering, or stable displacement. The displacement of air
14 by water in a homogenous porous medium during spontaneous imbibition
15 results in a uniform wetting front at the macroscopic scale, so that flow can
16 be approximated as a 1-dimensional problem (Kang et al., 2013). The
17 presence of one or more fractures, however, introduces a second flow
18 domain, with water moving more rapidly in the fractures than in the
19 surrounding matrix. Over time the wetting front develops a spatially non-
20 uniform profile with many peaks and depressions (Zhang et al., 2010b). For
21 this reason, macroscopic models for predicting spontaneous imbibition in
22 heterogeneous porous media often include a dispersion parameter to
23 account for capillary dispersion (e.g., Langlo and Espedal, 1994; Zhang and
24 Tchelepi, 1999; Garmeh et al. 2009).

25 Here we derive a first approximation of the spatial dispersion of the
26 wetting front in a fractured porous medium in terms of the variance of the
27 distances travelled within the matrix and fracture zones, respectively. We
28 consider spontaneous imbibition only at early times (i.e., neglecting the
29 effect of gravity). It is assumed that flow in both the matrix and fracture

zones is 1-dimensional, with no interactions between the two flow domains. From Eq. [3], the distance travelled by the wetting front at time, t , in the matrix, L_m , can be written as:

$$L_m = C_m \sqrt{t} \quad [6]$$

where C_m is the matrix sorptivity. Likewise, from Eq. [5] the distance travelled by the wetting front at time, t , in the fracture zone, x_f , is given by:

$$x_f = E_f \sqrt{t} \quad [7]$$

where E_f is the fracture sorptivity. The dispersion coefficient, $\mathbb{D}$, representing the spatial spreading of the wetting front in a heterogeneous porous medium, can be calculated from the variance, σ^2 , of the travel distances using the following expression (Fried and Combarnous, 1971; Perfect and Sukop, 2001):

$$\mathbb{D} \approx \frac{\sigma^2}{2t} \quad [8]$$

For a dual domain porous medium comprised of an unfractured matrix and a fracture zone, the variance of the travel distances is given by:

$$\sigma^2 = \left(\frac{x_f - L_m}{2} \right)^2 \quad [9]$$

Substituting Eqs. [6], [7], and [9] into Eq. [8] and simplifying the result produces the following expression for the dispersion of the wetting front during early-time spontaneous imbibition in a fractured porous medium:

$$D \approx \frac{1}{8}(E_f - C_m)^2 \quad [10]$$

Equation [10] indicates that the dispersion coefficient is directly proportional to the difference in the sorptivities for the fractured and unfractured zones.

3. MATERIALS AND METHODS

3.1 Rock cores

Commercially available Berea sandstone cores with different hydraulic properties were investigated. They were purchased from Cleveland Quarries (Vermillion, OH). According to Cleveland Quarries the mineralogical composition of the Berea sandstone is as follows: 93.13% Silica (SiO_2), 3.86% Alumina (Al_2O_3), 0.11% Ferric Oxide (Fe_2O_3), 0.54% Ferrous Oxide (FeO), 0.25% Magnesium Oxide (MgO), and 0.10% Calcium Oxide (CaO). The individual cores were 2.54 cm dia. x 5.08 cm long. They were supplied in three different permeability classes: ~50 mD, ~200 mD, and ~500 mD. The porosities of the cores, determined from the solid volume measured with a temperature-controlled helium gas pycnometer (HumiPyc Model 1, InstruQuest Inc., Coconut Creek, FL) and the bulk volume measured with a NextEngine Desktop 3D Scanner (NextEngine Inc., Santa Monica, Ca, USA) using the method of Rossi et al. (2008) and Rossi and Graham (2010), were 0.19, 0.25, and 0.24 for the 50 mD, 200 mD, and 500 mD permeability classes, respectively (Table 1).

3.2 Core preparation and fracture induction

The cores were wrapped in Kapton[®] tape (DuPont, Wilmington, DE) to provide a no flow barrier along the sides. The wrapped cores were then fractured longitudinally by compression between parallel flat metal plates following the Brazilian method for determining indirect tensile strength (Li et

al., 2013). An example of a typical fracture zone produced by the Brazilian method is shown in Fig. 2.

3.3 Water imbibition setup

Prior to analysis the fractured cores were oven dried at 105°C for 24 hr. After cooling down they were suspended above a free water table in a high purity aluminum cell that was open to the atmosphere. The aluminum cell was placed in the CG-1D neutron beam line of the Neutron Imaging Prototype Facility at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory (ORNL). The cell was positioned immediately in front of the detector (see below), and was connected to a Mariotte bottle (McCarthy, 1934) located outside of the beam line using Tygon[®] tubing (Saint-Gobain S.A., La Défense, Courbevoie, France). The water table in the aluminum cell was controlled externally using the Mariotte bottle. The water level was carefully raised over time until it just touched the base of each core. The time of contact was taken as the baseline time, $t = 0$, for the imbibition measurements. The height of the water table was then held constant for the duration of each experiment.

3.4 Dynamic neutron radiography

Neutrons are ideally suited to the task of visualizing the distribution and movement of water within variably-saturated porous media (Perfect et al., 2014). This is because they are strongly attenuated by the hydrogen atoms in water molecules, but are relative insensitive to the gas phase in pores and minerals (such as silica and iron) which constitute the solid matrix. Digital images of the spatial pattern of neutron attenuation within a sample can be acquired in both 2- and 3-dimensions (2-d and 3-d, respectively). Neutron radiography is the preferred mode of data collection for dynamic processes

1 such as the rapid uptake of water within fractures since 2-d radiographs can
2 be acquired much more rapidly than 3-d tomograms.

3 Dynamic neutron radiography of the spontaneous imbibition of water
4 displacing air in fractured Berea sandstone cores was performed at CG-1D
5 Neutron Imaging Prototype Facility (HFIR, ORNL). CG-1D is a cold neutron
6 beam line with a curved guide, which eliminates gamma's and epithermal
7 neutrons from the spectrum. The neutron wavelengths vary between 0.8 and
8 6 Å, with peak intensity at 2.6 Å. Apertures with different diameters (δ) are
9 used at the entrance of the helium-filled flight path (pinhole geometry) to
10 allow for ℓ/δ variation from 400 to 800, where ℓ is the distance between the
11 aperture and the detector. With these ℓ/δ ratios, the beam has a very small
12 angular divergence (a fraction of a degree) and an intensity of
13 10^7 neutrons/cm²/s.

14 The detector consisted of stacked neutron sensitive microchannel plates
15 (MCP's) placed above a quad Timepix readout with a 28 x 28 mm² field of
16 view (FOV) (Tremsin, 2012). The cold neutron detection efficiency is 70%
17 (Tremsin et al., 2011). The native spatial resolution of the detector is 55 μm
18 determined by the pixel size of the Timepix readout, but sub-15 μm spatial
19 resolutions can be achieved through event centroiding (Tremsin et al. 2012).
20 The detector configuration with fast parallel electronics allows readout times
21 of ~290 μs (Tremsin, 2012; Tremsin et al. 2013) enabling > 1 kHz frame
22 rates. In our experiments, the dry rock cores were imaged at 25 μm spatial
23 resolution with the MCP detector in centroiding mode. For the dynamic
24 spontaneous imbibition experiments neutron radiographs were acquired at
25 frequencies of 10-20 frames per second (50-100ms, microsecond) (in order
26 to acquire sufficient number of neutrons per image) with the detector in
27 native spatial resolution mode.

3.5 Image analysis and parameter estimation

The raw neutron radiographs were imported into ImageJ (Schneider et al., 2012). The high spatial resolution images were normalized with respect to the open beam and despeckled to visualize the rock core and fracture damage zone (Fig. 2). The width of the fracture damage zone was measured at 15 equally-spaced locations along the entire length of the core, and mean values and standard errors were calculated from these measurements. Additionally, images of the top and bottom faces of the fractured cores were acquired with a digital camera, and these images were analyzed in ImageJ. The width of the fracture aperture on each core face was measured at 10 equally-spaced locations. The measurements from both fracture faces were then averaged to provide estimates of the mean fracture aperture width and its standard error.

The wetting images were normalized with respect to the initial image of the time sequence (when the core was completely dry) so that only the imbibing water was resolved (Fig. 3). The resulting normalized images were despeckled and thresholded so that the vertical distance between the water table and the tips of the wetting fronts in the matrix and fracture zone could be quantified over time (Fig. 3).

Matrix and fracture zone sorptivities, C_m and E_f , were estimated from the progression of the wetting front versus square root of time data using linear regression with no intercept. A total of 6 Berea sandstone cores were analyzed: 4 fractured cores (one from each permeability class plus a second core from the 500 mD permeability class to assess between core variability) and 2 unfractured cores (from the 200 and 500 mD permeability classes). Estimates of C_m (matrix sorptivity) for the 50 mD and second 500 mD permeability class cores were obtained by analyzing the unfractured matrix on either side of the water-filled fracture zone, well away from the lateral spread of the wetting plume. To assess within core variability, two

1 measurements of C_m were made within the unfractured matrix of each core.
2 Dispersion coefficients for the fractured cores were calculated from the
3 resulting estimates of C_m and E_f using Eq. [10].

4 5 **RESULTS AND DISCUSSION** 6

7 The matrix porosity and fracture characteristics of the Berea sandstone
8 cores are given in Table 1. The matrix porosity of the 50 mD permeability
9 core was substantially lower than the matrix porosities of the cores in the
10 other permeability classes (Table 1).

11 The Brazilian method created fractures with aperture widths (measured
12 on the top and bottom of the cores) ranging from 120 μm to 780 μm (Table
13 1). There was no obvious relationship between the width of the fractures
14 and the matrix porosity or permeability class of the cores. Instead, aperture
15 width and fracture morphology appeared to vary randomly, highlighting the
16 probabilistic nature of the tensile failure process. The 50 mD and second
17 500 mD permeability class cores produced simple planar fractures that were
18 uniformly narrow (Table 1), whereas the 200 mD and first 500 mD
19 permeability class cores produced complex planar fractures with
20 bifurcations that were much wider and more variable (Table 1).

21 The width of the fracture damage zone (estimated from the neutron
22 radiographs) varied between 2 and 10 times the width of the fracture
23 aperture (Table 1). This zone included damage, deformation, and micro-
24 cracking in addition to the fracture opening. The width of the fracture
25 damage zone generally increased in proportion to the width of the fracture
26 aperture (Table 1). No apparent relationship was found between the width
27 of damage zone and the matrix porosity, or the core permeability class.

28 Water uptake into all of the fractures was extremely rapid. Figure 3 is a
29 time sequence of neutron radiographs showing water moving into the air-
30 filled fracture damage zone of the 50 mD permeability core. The pore water

1 pressure was never positive as evidenced by the menisci visible where the
2 sides of the core contact the water table (Fig. 3). At the end of this
3 sequence of images the wetting front in the fracture zone had travelled
4 approximately an order of magnitude further than the wetting front in the
5 matrix (i.e., the dark gray zone at the base of the core).

6 The Bond number (Bo) is a dimensionless quantity that can be used to
7 demonstrate that gravity was negligible under the conditions of our
8 experiments. The equation for the Bond number can be written as:

$$Bo = \frac{(\rho_w - \rho_n)gk}{\sigma_{wn}} \quad [11]$$

12 where ρ_w and ρ_n are the densities of the wetting and non-wetting fluids,
13 respectively, k is the intrinsic permeability, σ_{wn} is the surface tension of the
14 interface between the wetting and non-wetting phases. If $Bo \ll 1$ then
15 gravity can be ignored. For water and air as the wetting and non-wetting
16 fluids, respectively ($\rho_w = 1000 \text{ kg/m}^3$, $\rho_n = 0 \text{ kg/m}^3$, and $\sigma_{wn} = 0.0728$
17 N/m) and $g = 9.81 \text{ m/s}^2$, the permeability values investigated in this study
18 ($k = 4.9 \times 10^{-15}$ to $4.9 \times 10^{-14} \text{ m}^2$) result in calculated Bo numbers of
19 between 6.6×10^{-9} and 6.6×10^{-8} which is much less than unity; so we can
20 safely assume the effect of gravity was negligible under the conditions of
21 our experiments.

22 Quantitative results from the image analyses of the neutron radiographs
23 are summarized in Figure 4 and Table 1. The spontaneous imbibition of
24 water into both the sandstone matrix and the fracture damage zone
25 exhibited square-root-of-time behavior. This is evidenced by the excellent
26 coefficient of determination (R^2) values (i.e. > 0.9) for Eqs. [6] and [7]
27 fitted to the experimental observations regardless of the location of the
28 uptake (Fig. 4). It can be seen that the first point of some of the data sets
29 (e.g., the fracture zone in Fig. 4a and the matrix in Fig. 4c) tends to fall

1 below the best fit line. These outliers indicate deviations from square-root-
2 of-time behavior at very early times. Possible causes of these deviations
3 are: (1) the base of the core may not have be perfectly aligned with the
4 water surface resulting in partial contact at very early times, and (2) the
5 first image was integrated over a relatively long period of time (5 to 10 ms)
6 relative to the rate of wetting at very early times.

7 Between core variability of the matrix and fracture zone water uptake
8 measurements can be assessed by comparing the results for the two 500
9 mD permeability class cores in Figs. 4(c) and 4(d). In general, both sets of
10 measurements were reasonably comparable; coefficients of variation (CV)
11 calculated from the slopes of the regression equations for the two cores
12 were 6% and 12% for the matrix and fracture zone data, respectively. In
13 some respects, one might have expected greater variability between the
14 fracture imbibition measurements for these two cores due to the different
15 aperture and damage zone widths (Table 1).

16 The replicate measurements of spontaneous imbibition of water into
17 the unfractured matrix of each core permit an assessment of the within core
18 variability for this measurement. The results in Fig. 4 indicate generally
19 good reproducibility; the 50 mD permeability class core (Fig. 4a) exhibited
20 the greatest variability with a CV of 17%, while the CV's for the other three
21 cores were all < 2.5%. Since there was only one fracture damage zone per
22 core it was not possible to assess within-core variability for this parameter.

23 The matrix sorptivities ranged from 2.90 to 4.55 mm s^{-0.5} (Table 1),
24 and increased linearly as the permeability class increased (Fig. 5a). The
25 matrix sorptivity coefficient changed by a factor of ~2 as the permeability
26 changed by a factor of ~4 which is consistent with the traditional assumed
27 square dependence of the permeability on the mean pore size (Table 1).

28 The average value for the 50 mD permeability class core was ~2.5 times
29 higher than the average matrix sorptivity for Berea sandstone determined

1 by Kang et al. (2013) using the same method. The cores analyzed by Kang
2 et al. (2013) were supplied by a different company and had a slightly lower
3 average permeability of ~42 mD. Moreover, due to differences in the
4 detector systems employed by the two studies, water uptake data were
5 collected over two completely different time frames. In the present study,
6 measurements were made over the range 0.1~2.0 s, whereas the Kang et
7 al. (2013) data were obtained over the range 5~50 s with a charge-coupled
8 device (CCD) detector at the same beam line. Thus, the lower matrix
9 sorptivities reported in Kang et al. (2013), as compared to those in Table 1
10 can be attributed to a combination of lower core permeability and the
11 increased effect of gravity at later times as described by Eq. [2].

12 The fracture damage zone sorptivities were between 4 and 7 times
13 greater than the corresponding matrix sorptivities (Table 1). The sorptivities
14 of the fracture damage zones ranged from 17.87 to 27.12 mm s^{-0.5} (Table
15 1), and increased linearly with increasing fracture aperture width (Fig. 5b).
16 The fracture sorptivity coefficients for the 50 md and 500 md (replicate #2)
17 permeability cores samples were similar, which is consistent with the similar
18 fracture aperture widths in these two samples (Table 1).

19 The sorptivity results indicate that fractures can significantly increase
20 spontaneous imbibition in unsaturated sedimentary rock by capillary action
21 and surface spreading on rough fracture faces. Our results are consistent
22 with the studies by Şahmaran and Li (2009) and Zhang et al. (2010a,b),
23 which showed enhanced sorption in fractured cementitious materials. The
24 rates of water uptake observed in the present study were surprisingly rapid.
25 The fracture sorptivities in Table 1 indicate that wetting fronts can travel
26 between 2 and 3 cm in 1 second in unsaturated damage zones.

27 We ran a simple qualitative experiment to isolate the phenomenon of
28 water spreading over a rough fracture surface, Eq. [4] from the dynamics of
29 capillary rise within a narrow fracture aperture, Eq. [3]. An oven-dry 500

1 mD permeability class Berea sandstone core was fractured as described previously. The two halves of the fractured rock core were carefully separated and one half was rotated 180° relative to the other half. The two halves were then placed side-by-side, at the same time, in a shallow pan of water so that water uptake could be monitored simultaneously on the interior (fracture face) and exterior surfaces of the core.

Figure 6 shows the results of spontaneous imbibition of water into the two halves of the fractured core after approximately ~1-2 s of wetting. It can be clearly seen that the wetting front has moved approximately twice as far on the rough fracture surface as compared to the wetting front on the smooth exterior of the core. This movement occurred in the absence of the second fracture face, indicating that water spread over the rough surface of the exposed fracture face, similar to the observations of Tokunaga and Wan (2001) and Vorobyev and Guo (2010). The wetting front on the other half of the core, with its smooth exterior exposed, can be attributed to capillary rise within the unfractured matrix. It is interesting to note that the wetting front on the exterior of the core was curvilinear. This was likely due to surface spreading on the (unseen) fracture face on the other side of this half of the core which has induced water to move more rapidly along the edges. Overall, this simple experiment demonstrates that surface spreading contributes significantly to the rapid imbibition of water in fractured sedimentary rocks.

The dispersion coefficients, calculated from the matrix and fracture sorptivities, ranged from 23.7 to 66.7 mm² s⁻¹ (Table 1). Figures 5(c) and 5(d) indicate that dispersion increased linearly with increasing width of both the fracture aperture and its associated damage zone. The strongest relationship was with the fracture aperture width. The larger the dispersion parameter, the greater the spreading of the wetting front within the core.

4. CONCLUDING REMARKS

Neutron radiography was able to capture the early-time dynamics of spontaneous imbibition of water in fractured, unsaturated sedimentary rock cores. Rates of water uptake into the fracture damage zones were surprisingly rapid, with wetting fronts travelling between 2 and 3 cm per second. A theoretical model was derived to explain these results. The rapid water uptake was attributed to a combination of early time capillary rise within the fracture aperture and spreading over the rough surfaces of the opposing fracture faces. The theory also permits estimation of sorptivity values for the matrix and fracture zones, as well as a dispersion parameter, which quantifies the extent of spreading of the wetting front. The sorptivity of the fracture zone and the dispersion coefficient both increased linearly with increasing fracture aperture width.

The current theoretical framework represents only a 1-dimensional approximation of spontaneous imbibition within a fractured porous medium. To fully describe this phenomenon the derivation of a 2-dimensional model, that permits interaction between the fracture and matrix, will be needed. The development of such a model represents an exciting opportunity for future research.

The present results for fractured Berea sandstone are consistent with observations of rapid water uptake in fractured cementitious materials. Additional research is needed to document this phenomenon in other natural and engineered porous media. The phenomenon is likely to be particularly important in the case of porous materials such as bricks and building stone used in engineering applications. Given the current high level of interest in unconventional gas from black shale, similar experiments performed on fractured shale cores may also be valuable. The spontaneous imbibition of water-based fracking fluids into damage zones created by

hydraulic fracturing operations could potentially result in significant losses of water ("leakoff") with concomitant blockages of the pathways available for efficient gas extraction.

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Table 1. Mean values and associated standard errors (in parentheses) of measured hydraulic parameters for the fractured Berea sandstone cores.

Permeability class (mD)	Matrix porosity (m ³ m ⁻³)	Matrix sorptivity ^a (mm s ^{-0.5})	Fracture aperture width (μm)	Fracture damage zone width (μm)	Fracture zone sorptivity (mm s ^{-0.5})	Dispersion coefficient (mm ² s ⁻¹)
50	0.19 (0.01)	2.90 (0.21)	140 (20)	620 (80)	17.87 (0.78)	28.01 (3.71)
200	0.25 (0.01)	4.05 (0.19)	780 (120)	1,610 (100)	27.12 (0.96)	66.53 (6.63)
500 (replicate 1)	0.24 (<0.01)	3.77 (0.24)	350 (50)	1,310 (80)	19.86 (0.85)	32.36 (4.38)
500 (replicate 2)	0.24 (<0.01)	4.55 (0.25)	120 (10)	1,170 (130)	18.32 (0.54)	23.70 (2.72)

^a Average of the two replicate measurements reported in Fig. 4

FIGURE CAPTIONS

Figure 1. Wetting front elevation versus time predicted using Eqs. [2] and [3] for a two-region porous medium with $\langle \lambda \rangle = 50 \mu\text{m}$ and $\tau = 1.8$ in the unfractured matrix and $\langle \lambda \rangle = 150 \mu\text{m}$ and $\tau = 1.2$ in the fracture zone. All of the other parameters in Eqs. [2] and [3] were the same for the two regions, i.e. $\sigma = 0.0728 \text{ N/m}$, $\mu = 0.001 \text{ Ns/m}^2$, $\cos(\theta) = 1$, $\rho = 1000 \text{ kg/m}^3$, and $g = 9.81 \text{ m/s}^2$.

Figure 2. Example of a longitudinal air-filled fracture zone (dark gray) in an oven dry 50 mD permeability class Berea sandstone core (light gray) imaged using neutron radiography at the HFIR's CG-1D beam line, ORNL (see Kang et al., 2013 for technical details). For scale, the core is 2.54 cm dia. x 5.08 cm long. The aluminum container and basal reservoir can also be seen in outline. The core was wrapped in Kapton tape to provide a no flow barrier along the sides. The top of the core was wrapped more thickly than the bottom resulting in the contrasting lighter and darker sections. The $28 \times 28 \text{ mm}^2$ FOV for the fracture imbibition images in Fig. 3 is shown as a white box.

Figure 3. Time sequence of neutron radiographs running from left to right and top to bottom showing the rapid uptake of water into a longitudinal, air-filled fracture zone in Berea sandstone ($\sim 50 \text{ mD}$). The images have been normalized with respect to the initial dry image, so that only water (black) is visible. The rock core, similar to the one in Figure 2, was positioned above a water table which contacted its base immediately after the first image. The height of the water table was then held constant. The annotations on the 1.4 sec image indicate where the distances traveled by the wetting front in

the matrix, $L_{m,}$ (2 replicate locations) and fracture zone, x_f , were measured. For scale the width of each image is 26 mm.

Figure 4. Height of the wetting front versus the square root (Sqrt) of time in the matrix (2 replicate measurements per core) and fracture zones of Berea sandstone measured using dynamic neutron radiography. Each graph corresponds to a different permeability class: (a) 50mD, (b) 200mD, (c) 500mD (core #1), and (d) 500mD (core #2). The replicate matrix measurements in (a) and (c) were measured on the sides of the fractured cores, while those in (b) and (d) were measured on separate unfractured cores. The slopes of the fitted linear regression models represent the sorptivity estimates.

Figure 5. Linear regression relationships between the sorptivity and dispersion estimates and core permeability class and fracture parameters: (a) matrix sorptivity versus permeability class, (b) fracture zone sorptivity versus fracture aperture width, (c) dispersion coefficient versus fracture aperture width, and (d) dispersion coefficient versus fracture zone width. The horizontal and vertical bars around the individual data points represent plus and minus standard errors.

Figure 6. Spontaneous imbibition of water into a fractured, oven dry 500 mD permeability class Berea sandstone core showing the differential extent of wetting on the rough interior fracture surface as compared to the smooth exterior surface.

Fig. 1

Fig. 2

Fig. 3

(a)

(b)

(c)

(d)

Fig. 4

Fig. 5

Fig. 6