Revealing soil-borne hydrogel effects on soil hydraulic properties using a roughness-triangular pore space model

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Abstract
Understanding the influence of soil-borne hydrogels (e.g., bacterial extracellular polymeric substances [EPS] and biofilm or root-derived mucilage) on soil hydraulic properties is crucial for accurate modeling of flow and transport in porous media. The underlying mechanisms of how hydrogels modify soil physical and hydraulic properties, however, remain unclear. In this study, we applied a roughness-triangular pore space model (R-TPSM) to simulate water retention curve and hydraulic conductivity of hydrogel-affected soils. The model considers the effective soil pore size distribution and physicochemical properties (e.g., surface tension and surface roughness) of soil and soil solution. The modeling results are in good agreement with measured water retention curves and hydraulic conductivity for different textured soils. Comparison of the fitted model parameters between control and hydrogel-treated soil samples indicates that hydrogels reduce surface tension and shift the effective pore size distribution to a narrower size range. Hydrogels generally increase water retention in the form of adsorptive films and reduce film flow under relatively dry conditions. This study highlights the importance of considering reduced surface tension and mean pore size by hydrogels in changing soil hydraulic properties, which has important implications for accurate modeling of water distribution and flow in the rhizosphere.

1 INTRODUCTION

Soil-borne hydrogels, including microbial exudates such as bacterial biofilm or extracellular polymeric substances (EPS) (Flemming & Wingender, 2010), and root-derived mucilage (Czarnes, Hallett, Bengough, & Young, 2000), exist widely in natural environments. Those soil-borne hydrogels have common properties. For example, they have unique three-dimensional (3D) structures, high water-adsorbing capacity, and higher viscosity than water and have been found to greatly affect water retention and soil hydraulic properties due to their hygroscopic nature and through changing soil structures (Carminati, Benard, Ahmed, & Zarebanadkouki, 2017; Or, Phutane, & Dechesne, 2007). In addition, these hydrogels contain surfactant compounds that reduce surface tension at the gas–liquid interface, which further accentuates their effects on water retention and flow behavior in hydrogel-affected soils (Read, Gregory, & Bell, 1999;...
Naveed et al., 2019). Water status in agricultural soils controls the availability of water and nutrients for plant production. Therefore, understanding the influence of hydrogels on soil water retention and hydraulic properties is critical to water management, as well as to developing potential strategies of applying hydrogels to remediate soils with poor quality (Abedi-Koupai, Sohrab, & Swarbrick, 2008; Brax, Buchmann, & Schaumann, 2017).

Experimental studies have led to several proposed mechanisms of how hydrogels modify soil physical and hydraulic properties. Specifically, a hydrogel, having a 3D network structure, may exist as a separate phase and exhibit distinct water adsorption properties. It has been reported that a hydrogel behaves like a sponge and is capable of adsorbing water at an amount of tens (xanthan or bacterial EPS; Chenu & Roberson, 1996; Roberston & Firestone, 1992) to hundreds times (root mucilage of maize [Zea mays L.]; Ahmed, Holz, Woche, Bachmann, & Carminati, 2015; Guinel & McCully, 1986) of its dry weight. Water adsorption can result in swelling of the EPS matrix, leading to progressive increase in spaces between their biopolymeric strands (Shaw et al., 2003). The observed increase in saturated water content of biofilm-amended soils (Rosenzweig, Shavit, & Furman, 2012; Volk, Iden, Furman, Durner, & Rosenzweig, 2016; Zheng et al., 2018) is attributed to such hygroscopic effect.

In addition to large water-holding capacity, recent findings revealed additional mechanisms including hydrogels’ ability to modify the distribution of the liquid phase under partially saturated conditions. Direct visualization depicted the heterogeneous structure of hydrogels. For example, root mucilage and bacteria biofilm have been shown to clog small pores or separate large pores into smaller spaces. Such impacts indicate that hydrogels can reduce flow conductance and cause additional capillary effects (Benard et al., 2019; Carminati et al., 2017; Kroener, Holz, Zarebanadkouki, Ahmed, & Carminati, 2018; Zheng et al., 2018).

Hydrogels also modify the surface tension and viscosity of soil solution. Root mucilage of maize and barley (Hordeum vulgare L.) was shown to behave like surfactants and reduced surface tension (Read et al., 1999; Naveed et al., 2019). Read et al. (1999) reported that surface tension decreased from 72 to 45–50 mN m⁻¹ at the concentration of 1 mg ml⁻¹ mucilage from maize and lupine seedlings. Some microbes (bacteria or fungi) produce biosurfactants (Santos, Rufino, Luna, Santos, & Sarubbo, 2016), such as rhamnolipids from Pseudomonas aeruginosa, and surfactin from Bacillus subtilis. Deziel, Paquette, Villemur, Lepine, and Bisaillon (1996) showed that a Pseudomonas aeruginosa 19SJ strain growing on polycyclic aromatic hydrocarbon reduced surface tension of the culture supernatants (cell concentration was not reported) from 74 to 33 mN m⁻¹. Reduced surface tension of soil solution generally facilitates drainage of large soil pores. However, its role in soil water dynamics, particularly in rhizosphere hydrology, remains open (Carminati et al., 2017). Soil-borne hydrogels have higher viscosity values than water and thus can increase flow resistance. Angelini, Roper, Kolter, Weitz, and Brenner (2009) reported that the viscosity of biofilm produced by Strain 3610 B. subtilis cells measured at an air–liquid interface was 200 times higher than that of pure water. The viscosity of root exudates increased with an increasing mucilage concentration (Naveed et al., 2019). The highly viscous nature of hydrogels and their ability to form filaments and two-dimensional interconnected structures that span across multiple pores enhance water retention, maintain the connectivity of the liquid phase in drying soils, and decrease local water fluxes (Benard et al., 2019). These findings clearly demonstrate that multiple factors and complex interactions are involved in affecting the hydraulic properties of hydrogel-mediated porous media.

The abovementioned experimental studies have provided insights on how hydrogels may modify soil water retention and hydraulic properties. Modeling efforts have also contributed to the improved understanding of the involved mechanisms or processes. The so-called composite media models (Kroener et al., 2018; Rockhold, Yarwood, Niemet, Bottomley, & Selker, 2002; Rosenzweig et al., 2012) belong to a modeling approach where soil and hydrogel are treated as separate phases and water retention characteristics are calculated in a linear superposition manner by applying a predefined model (such as Brooks & Corey, 1966; van Genuchten, 1980) to each phase. Rosenzweig et al. (2012) applied this approach to compare model calculations with experimentally measured water retention data and concluded that the dominant mechanism for increased water retention was the distinct water holding-capacity of EPS. Although the composite media model can provide simulations that match experimental data, it does not explicitly consider changes in soil structure, pore size distribution, and the

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**Core Ideas**

- A R-TPSM was applied to model hydraulic properties of hydrogel-affected soils.
- Hydrogels reduce surface tension and shift pore size distribution to a narrower size range.
- Hydraulic property models of hydrogel-affected soils should consider reduced surface tension.
The process of hydrogel effects on soil hydraulic properties, such as water retention curve and hydraulic conductivity, is complex due to the interplay of physical and chemical factors. Volk et al. (2016) applied the Peters–Durner–Iden model (PDI), which is based on a binary-pore system assumption, and obtained excellent fits to experimental data for both water retention curve and hydraulic conductivity of biofilm-affected soil samples. The PDI model highlights the importance of water film and film conductivity; however, no physical interpretation of the model parameters was presented in Volk et al. (2016).

An improved approach to model hydraulic properties for hydrogel-affected soils is by taking hydrogel spatial distribution into consideration. Rosenzweig, Furman, and Shavit (2013) and Rosenzweig, Furman, Dosoretz, and Shavit (2014) developed a pore network model considering three hydrogel clogging scenarios and demonstrated that hydrogel spatial distribution had significant effects on water retention curve and hydraulic conductivity. There is yet another modeling approach, which is based on the framework of bundles of cylindrical capillary (BCC). For example, Carles Brangari et al. (2017) assumed that hydrogel created new channel geometries (i.e., one capillary tube was split by hydrogel into several smaller tubes inside the original one) and showed that even very small amounts of hydrogel could lead to significant changes in hydraulic properties.

The above models, to some extent, demonstrated the significance of pore structural change in reshaping water retention curve and hydraulic conductivity, but none took into account potential changes in soil hydraulic properties mediated by hydrogels through modifying physicochemical properties (e.g., surface tension) of soil solution. In this study, we simulated water retention curve and hydraulic conductivity of hydrogel-affected soils by applying a roughness-triangular pore space model (R-TPSM) developed in Zheng et al. (2015). As this model takes into account the effective soil pore size distribution, surface tension of soil solution, and soil surface roughness, hydrogel effects are assessed by modifying effective model parameters that quantify the effective soil pore size distribution and physicochemical properties. In this work, we accomplished the following: (a) we compared modeling results with experimental data from the literature and provided interpretation of the model parameters; (b) we conducted sensitivity analysis to evaluate how the model parameters are, to the extent possible, related to different physical processes that collectively control water dynamics in hydrogel-mediated soil, and (c) we discussed implications of the modeling results.

2 MODEL DESCRIPTION

2.1 The roughness-triangular pore space model

Soil hydraulic properties (i.e., water retention curve and hydraulic conductivity) are modeled by the R-TPSM, which was presented in Zheng et al. (2015); thus, only a brief introduction of the model is provided here. Soil pores can be represented by a bundle of triangular pores with a statistical pore size distribution (Diamantopoulos & Durner, 2013, 2015; Tuller & Or, 2001). The construction of the model includes two parts: (a) the geometric representation, retention curve, and hydrodynamic consideration for a single pore, and (b) upscaling from the pore scale to sample scale using a pore size distribution function.

2.1.1 Single-pore representation

In the R-TPSM, a single pore is characterized by an equivalent side length $L$ (m). Within the pore, water is held in small channels or corner regions by capillary force (capillary-dominated) and on the surface of solid particles as water film by van der Waals force. Only the drainage process is considered here because the experimental data under testing were obtained using the evaporation method, which corresponds to a drainage process. Upon drainage, as the matric potential $\mu$ (J kg$^{-1}$; note that the matric potential is usually defined in units J cm$^{-2}$; however, to apply the shifted Young–Laplace equation, we converted J cm$^{-3}$ into J kg$^{-1}$ by dividing the matric potential by the density of water $\rho$, 998.21 kg m$^{-3}$, but keeping the same name, matric potential) decreases (i.e., becomes more negative) and reaches the threshold of $\mu_t$, the liquid in a single pore is displaced by air from the central region, leaving liquid wedges in corners and liquid films (i.e., corner flow and film flow) on solid surfaces. Under these conditions, the radius of the capillary interface curvature, $r(\mu)$, at the onset of pore snap-off determines the size length $L_j$, below which the pores are fully filled. The curvature of the arc $r(\mu)$ increases and the thickness of water film, $h(\mu)$, decreases as the matric potential continues to decrease. The configuration of the liquid in a single pore is described by the shifted Young–Laplace equation (Tuller & Or, 2001). In the shifted Young–Laplace equation, adsorptive films and liquid wedges in corners are treated separately. The film adsorbed on all exposed solid surfaces is controlled by van der Waals forces, whereas the radius of the interface curvature for liquid wedges, calculated...
from the classic Young–Laplace equation, is shifted by the film thickness $h$.

Assuming zero gravity for small capillaries and for perfect wetting systems, the radius of the curved liquid–gas interface of liquid wedges $r(\mu)$ is expressed as a function of matric potential $\mu$ (J kg$^{-1}$) based on the Young–Laplace relationship (Mason & Morrow, 1991):

$$r(\mu) = -\frac{\sigma}{\rho \mu} \tag{1}$$

where $\sigma$ is the surface tension of the liquid (N m$^{-1}$), and $\rho$ is the density of water (kg m$^{-3}$). The adsorptive film thickness $h_{ad}(\mu)$ by adsorptive forces is written as

$$h(\mu) = X h_{ad}(\mu) = X \left( \frac{A_{wvl}}{6\pi \rho \mu} \right)^{\frac{1}{3}} \tag{2}$$

where $A_{wvl}$ is the Hamaker constant for solid–gas interactions through the intervening liquid (J), and $X$ is the roughness factor (–), which quantifies the magnitude of increased film adsorption due to roughness effects.

With the expression of $r(\mu)$ and $h(\mu)$, the saturation ($S_w$) and conductance of a single pore under a certain $\mu$ is readily calculated. A pore is completely filled before the pore snap-off during drainage (i.e., $S_w = 1$). Under unsaturated conditions, $S_w$ equals the ratio of the area occupied by liquid (the sum of liquid films and wedges) to the total area on the cross-sectional surface of the capillary tube:

$$S_w(\mu) = \frac{3h(\mu) [L - \sqrt{3}h(\mu)]}{A_3 L^2} + \frac{r^2(\mu) F_3}{A_3 L^2} (\mu < \mu_d) \tag{3}$$

where $A_3$ and $F_3$ are pore-shape-dependent parameters. For an equivalent triangle, $A_3 = \sqrt{3}/4$ and $F_3 = 3\sqrt{3} - \pi$.

The flow in a single pore is calculated based on the assumption of stable and fully developed laminar flow (Tuller & Or, 2001). Therefore, the flow conductance ($K_D$, $K_F$, and $K_C$ for ducts, films, and corners, respectively) is written as

$$K_D = \left( \frac{\rho g}{\eta} \right) \left( \frac{1}{80} \right) L^2 \tag{4}$$

$$K_F = \left( \frac{\rho g}{\eta} \right) \frac{h^2(\mu)}{3} \tag{5}$$

$$K_C = \left( \frac{\rho g}{\eta} \right) \frac{r^2(\mu)}{\varepsilon} \tag{6}$$

where $g$ is the acceleration due to gravity (m s$^{-2}$), $\eta$ is the viscosity of the bulk phase (kg m$^{-1}$ s$^{-1}$), $\eta = \eta_0 = 1.002 \times 10^{-3}$ kg m$^{-1}$ s$^{-1}$ for the control soil samples. In this study, we do not consider changes of viscosity due to hydrogels for duct and corner flow; however, the viscosity for film flow is set as 100 times higher than $\eta_0$, and $\varepsilon = 31.63$ is the dimensionless flow resistance parameter for the corner flow with a no-shear air–water interface and a corner angle of 60°. This is an oversimplification of the effects of viscosity and needs to be improved in future efforts.

### 2.1.2 Sample-scale expressions

The statistical property of a population of pores is described using the gamma distribution function:

$$f(L) = \frac{L^{\frac{5}{2}}}{\xi^3 \omega^{\frac{3}{2} + 1}} \exp \left( -\frac{L}{\omega} \right) \tag{7}$$

where $\xi$ is the shape factor (–) and $\omega$ is the scale factor (m). We used $\xi = 1$ in this study, which led to a statistical mean of $2\omega$ and variance of $2\omega^2$.

The sample-scale saturation $S_w$ is expressed as the sum of water fully filled in ducts, retained in corners and as water films. The three terms that are related to filled ducts ($S_{wd}$), film ($S_{wf}$), and corners ($S_{wc}$) are expressed as

$$S_w(\mu) = S_{wd}(\mu) + S_{wf}(\mu) + S_{wc}(\mu) \tag{8}$$

$$S_{wd}(\mu) = \int_{L_{min}}^{L_{1}(\mu)} f(L) dL \tag{9}$$

$$S_{wf}(\mu) = \int_{L_{1}(\mu)}^{L_{max}} \frac{3h(\mu) [L - \sqrt{3}h(\mu)]}{A_3 L^2} f(L) dL \tag{10}$$

$$S_{wc}(\mu) = \int_{L_{1}(\mu)}^{L_{max}} \frac{r^2(\mu) F_3}{A_3 L^2} f(L) dL \tag{11}$$

where $L_{min}$ and $L_{max}$ are the smallest and largest pore size, respectively, and $L_{max}$ is related to the air-entry value $\mu_d$ as

$$L_{max} = L_1(\mu_d) + 2\sqrt{3} h(\mu_d) \tag{12}$$

$$L_1(\mu) = -\frac{\sigma}{\rho \mu} C_3 \tag{13}$$
where \( L_1 \) is calculated from the radius of the capillary interface curvature at the onset of pore snap-off. \( C_3 \) is a geometric parameter, \( C_3 = 2(\sqrt{3} + \sqrt{\pi/\sqrt{3}}) \).

The sample-scale hydraulic conductivity \( K_w \) (m s\(^{-1}\)) is determined based on an upscaling scheme of adding each contribution from the duct flow \( (K_{wd}) \), film \( (K_{wf}) \), and corners \( (K_{wc}) \) calculated at the pore scale and saturated hydraulic conductivity \( K_s \):

\[
K_t (\mu) = \frac{K_{wd} (\mu) + K_{wf} (\mu) + K_{wc} (\mu)}{K_s} \tag{14}
\]

\[
K_s = \int_{L_{min}}^{L_{max}} \phi K_D f (L) \, dL \tag{15}
\]

\[
K_{wd} (\mu) = \int_{L_{min}}^{L_{1}(\mu)} \phi K_D f (L) \, dL \tag{16}
\]

\[
K_{wc} (\mu) = \int_{L_{1}(\mu)}^{L_{max}} \phi K_C \frac{r^2 (\mu) F_3}{A_3 L^2} f (L) \, dL \tag{17}
\]

\[
K_{wf} (\mu) = \int_{L_{1}(\mu)}^{L_{max}} \phi K_F \frac{3h (\mu) [L - \sqrt{3}h (\mu)]}{A_3 L^2} f (L) \, dL \tag{18}
\]

where \( \phi \) is the porosity. We also tabulated equations for calculating saturation and conductivity in the form of duct, film, and corner flow in Supplemental Table S1.

### 2.2 Incorporating hydrogel effects into the R-TPSM

In the R-TPSM, a hydrogel-affected sample is assumed to be homogeneous, and soil pores are simplified as a bundle of triangular pores. The hydrogel effects modifying the effective pore size distribution, surface tension, and film adsorption are reflected by the model parameters: scale factor \( \omega \), surface tension \( \sigma \), and roughness factor \( X \), respectively. The R-TPSM implemented in this study builds on a continuous scale so that the 3D pore-scale and transient behaviors could not be fully incorporated. Therefore, we treated the model as a semiphysical model, with the assumptions and our justifications discussed below.

1. Hydrogels swell at high soil moisture levels due to their hygroscopic properties, shrink, and then form filaments as the polymer molecules retreat with the retreat-

2. We assume that the surfactant molecules contained in bacteria EPS or plant mucilage decrease surface tension of soil solution. We also assume that the surfactant concentration reaches the CMC (critical micelle concentration) so that water loss during drainage or evaporation does not further reduce the surface tension of soil solution.

3. We assume a contact angle of 0 for the simulated drainage process. Diamantopoulos and Durner (2013) showed that the receding contact angle, which is more relevant to drainage, was 0 for both hydrophilic and hydrophobic soils.

4. We assume that hydrogels change water film adsorption by influencing the roughness factor \( X \). The roughness factor was used to represent the effects of physical roughness on water film thickness based on van der Waals interactions (Zheng et al., 2015). For hydrogel-treated soil samples, we use \( X \) to capture additional effects that lead to changes in film adsorption on soil surface due to hydrogel presence, which include (a) that hydrogels may increase film thickness due to polymer interactions induced by steric forces (Israelachvili, 1991) in addition to van der Waals forces, and (b) that the surfactants contained in hydrogels may decrease the thickness of adsorbed water films, which are capillary-held and induced by surface roughness (Kibbey, 2013).

5. We assume that hydrogels are immobile during the simulated drainage process. The mobilization of EPS or mucilage could happen under certain scenarios but is unlikely for typical soil conditions, where biopolymers are attached on soil particles and water flow velocities are low (Tong, Long, Jiang, & Kim, 2010).

### 3 DATA DESCRIPTION

The R-TPSM was tested using five different datasets: a Hamra sand (Chromic Luvisol), a Pepperbox (loamy,
mixed, semiactive, mesic Aquic Arenic Paleudults) sand, a Lenni (fine, mixed, active, mesic Typic 141 Endoaquults) clay, and two sandy loams collected from Reinhausen and Bullion, obtained from the literature. Particle size fractions and soil organic matter contents of the five soils are listed in Table 1. For Hamra sand, the water retention curves and unsaturated hydraulic conductivity values of the control and soil incubated with Pseudomonas putida (with the average viable count of \(2.3 \times 10^7\) colony-forming units g\(^{-1}\) soil) were determined by a combination of Hyprop and WP4C dewpoint methods, and the saturated hydraulic conductivities were measured using a constant head permeameter (Volk et al., 2016). For Pepperbox sand and Lenni clay, the treated soil samples were incubated with a Bacillus subtilis strain FB17 (with the commercial name UD1022). The Reinhausen sandy loam was treated with chia seed mucilage (Benard et al., 2019). The Bullion sandy loam was also treated with chia seed mucilage and water retention curve was measured using the polyethylene glycol (PEG) approach (Naveed et al., 2019).

We fitted the R-TPSM to the measured water retention curves by optimizing the model parameters (i.e., \(\sigma\), \(\omega\), \(X\), and \(\mu_3\)), and compared the differences between control and treatment. The model parameters were optimized using the covariance matrix adaptation evolution strategy (CMA-ES; Hansen, Muller, & Koumoutsakos, 2003), which is an evolutionary algorithm for nonlinear multiparameter optimization problems in a continuous domain. The code implemented in Matlab was used and can be found at [http://cma.gforge.inria.fr/cmaes_sourcecode_page.html#matlab](http://cma.gforge.inria.fr/cmaes_sourcecode_page.html#matlab). The unsaturated hydraulic conductivity was calculated based on the fitted model parameters from water retention curves and measured saturated hydraulic conductivity values.

### RESULTS

#### 4.1 Comparison of modeling and experimental results

The measured and fitted water retention curves of control and treated soil samples are plotted in Figure 1 for Hamra sand, Pepperbox sand, Lenni clay, and Reinhausen and Bullion sandy loams. Figure 1 shows that the R-TPSM is able to fit all experimental data very well, with RMSE values of \(\sim 0.01\) cm\(^3\) cm\(^{-3}\) for all soil samples (Table 2), which are comparable with the measurement error of Hyprop (0.008 cm\(^3\) cm\(^{-3}\)). The error of Hyprop was estimated based on the precision of the balance (0.02 g) used in the Hyprop setup (e.g., for a soil sample \([250\ \text{cm}^3]\) with a water content of 10%, the experimental error is \(0.02\ g/[250\ \text{cm}^3 \times 10\% \times 1\ \text{g cm}^{-3}] = 0.008\)). Higher saturated water contents, an increase of 2.74, 1.22, 12.3, and 9.95%, respectively, are found for bacteria-incubated Hamra sand, Pepperbox sand, Lenni clay, and mucilage-treated Reinhausen sandy loam compared with their controls. Figure 1 also shows that the values of water content at the drier end are higher and the bubbling pressures (i.e., the matric suction at which air starts to enter the soil), are lower (i.e., more negative) for treated soil samples than their controls. Furthermore, the capillarity- and adsorption-controlled water retention curves, shown separately in Figure 1, demonstrate that adsorption dominates at the drier end, and this observation is more significant (adsorptive water retention increased at the drier end) for treated soil samples than for their controls.

Figure 2 shows that hydrogel treatment reduced the unsaturated hydraulic conductivity for Hamra sand and Reinhausen sandy loam over the tested matric potential range. In addition, there is a change in the slope of the hydraulic conductivity curve when plotting on the log scale, implying that capillary flow dominates at relatively
FIGURE 1  Measured (Meas.) and calculated water retention curves using the roughness-triangular pore space model (R-TPSM) for control (C) and treated (T) Hamra sand, Pepperbox sand, and Lenni clay, Reinhausen sandy loam, and Bullion sandy loam. The capillary curve includes water in filled ducts and corners whereas the adsorption curve represents water in water films.

TABLE 2  Saturated water content $\theta_s$ and fitted model parameters with the roughness-triangular pore space model (R-TPSM), including surface tension $\sigma$, scale factor $\omega$, roughness factor $X$, the threshold matric potential during drainage $\mu_d$, RMSE, and measured or estimated saturated hydraulic conductivity $K_s$.

| Soil                  | $\theta_s$ (cm$^3$ cm$^{-3}$) | $\sigma$ (N m$^{-1}$) | $\omega$ | $X$ | $\mu_d$ (J kg$^{-1}$) | RMSE (cm$^3$ cm$^{-3}$) | $K_s$ (cm d$^{-1}$) |
|-----------------------|--------------------------------|------------------------|----------|-----|------------------------|--------------------------|---------------------|
| Hamra sand-C          | 0.329                          | 0.0721                 |          |     | 4.427 $\times$ 10$^{-5}$ | 836                      | 0.011               | 173.0               |
| Hamra sand-T          | 0.338                          | 0.0540                 |          |     | 3.419 $\times$ 10$^{-5}$ | 650                      | -4.912              | 0.013               | 10.4               |
| Pepperbox sand-C      | 0.410                          | 0.0729                 |          |     | 5.456 $\times$ 10$^{-5}$ | 795                      | -4.825              | 0.013               |
| Pepperbox sand-T      | 0.415                          | 0.0460                 |          |     | 3.423 $\times$ 10$^{-5}$ | 756                      | -2.794              | 0.014               |
| Lenni clay-C          | 0.478                          | 0.0784                 |          |     | 2.552 $\times$ 10$^{-5}$ | 423                      | -3.720              | 0.011               |
| Lenni clay-T          | 0.537                          | 0.0590                 |          |     | 1.739 $\times$ 10$^{-5}$ | 765                      | -3.305              | 0.009               |
| Reinhausen sandy loam-C | 0.412                        | 0.0703                 |          |     | 4.040 $\times$ 10$^{-5}$ | 631                      | -3.212              | 0.019               | 1.21 $\times$ 10$^{-2}$ |
| Reinhausen sandy loam-T | 0.453                        | 0.0524                 |          |     | 1.440 $\times$ 10$^{-5}$ | 280                      | -0.527              | 0.006               | 4.13 $\times$ 10$^{-7}$ |
| Bullion sandy loam-C  | 0.546                          | 0.0714                 |          |     | 2.160 $\times$ 10$^{-5}$ | 883                      | -15.805             | 0.012               |
| Bullion sandy loam-T  | 0.546                          | 0.0516                 |          |     | 1.020 $\times$ 10$^{-5}$ | 526                      | -14.749             | 0.021               |

*C, control; T, treated.
FIGURE 2 Measured (Meas.) and calculated hydraulic conductivity using the roughness-triangular pore space model (R-TPSM) for control (C) and treated (T) Hamra sand and Reinhause sandy loam

FIGURE 3 The effective pore size distribution calculated with the fitted model parameters for all control (C) and treated (T) soils

wet conditions, whereas film flow controls at the dry end. Although the overall matching between the simulated results and their experimental data was not quantitatively very satisfactory, our model correctly predicts the trend of reduced hydraulic conductivity for treated samples vs. their controls, which agrees with the experimental data.

4.2 Model parameters

Fitted model parameters ($\sigma$, $\omega$, $X$, and $\mu_d$) from the water retention curves are listed in Table 2. Smaller values of surface tension $\sigma$ and scale factor $\omega$, and less negative air entry values $\mu_d$, were found for treated samples compared with their controls. Specifically, the fitted surface tension values are consistently lower for hydrogel-treated sand or clayey soil than the values of their controls. The fitted scale factor was reduced by 23, 38, 32, 64, and 53% for treated soils (Hamra sand, Pepperbox sand, and Lenni clay, Reinhause sandy loam, and Bullion sandy loam, respectively) compared with their controls. The effective pore size distributions of treated and control samples (Figure 3), calculated using the fitted scale factors, demonstrate that one of the effects of hydrogels is equivalent to a shift in pore sizes. The mean pore size of each treated sample shifted to a smaller value compared with its control (i.e., the number of bigger pores decreased and the number of smaller pores increased). An inconsistent trend was found between treatments for roughness factor (i.e., increased roughness factor value [by 81%] for bacteria-treated clay and decreased
values [by 22, 5, 56, and 48%] for other treated soils [Hamra sand Pepperbox sand, Reinsausen sandy loam, and Bullion sandy loam, respectively] than their controls.

An independent two-sample $t$ test was applied to model parameters, which was obtained by eight realizations of optimization (Supplemental Table S2). The differences in surface tension and the scale factor between treatment and control samples were statistically significant, as shown by the significantly larger $t$ values (Supplemental Table S3) than the critical $t$ value for both parameters. In addition, the correlation between any two of the model parameters was insignificant.

### 4.3 Sensitivity analysis

For illustrative purposes, we conducted sensitivity analysis, taking the parameters from Pepperbox sand control ($\xi = 1, \omega = 5.456 \times 10^{-5}, X = 794.9, \mu_d = -4.825$ cm, and $\sigma = 0.0729$ N m$^{-1}$) and changing one parameter at a time while holding others constant and calculated the corresponding water retention curve (left panel in Figure 4) and hydraulic conductivity (right panel in Figure 4). Figure 4 shows that the scale factor and surface tension influenced the water retention curve at both the wet and dry ends within the tested water content range, whereas the roughness factor mainly affected the dry end. Decreasing surface tension or increasing the scale factor led to earlier air entry and higher water content at the dry end. The roughness factor did not affect air entry; however, it dramatically increased the water content at the intermediate potential range. Changing the air-entry value from $-4.59$ to $-1.15$ cm did not significantly affect the water retention curve.

The relative hydraulic conductivity was sensitive to changes of the four parameters as well. Although reducing surface tension decreased hydraulic conductivity at the wet end, decreasing the scale factor or increasing the roughness factor mainly increased hydraulic conductivity at more negative potentials (i.e., less than $-10$ cm). Decreasing the air-entry value did not significantly affect the relative hydraulic conductivity over the whole range of the matric potentials.

### 5 DISCUSSION

#### 5.1 Mechanisms of hydrogel effects on soil hydraulic properties

Water is retained in soil by capillary force at the relatively high water content, and by adsorptive forces due to the presence of exchangeable cation hydration, mineral surface, or crystal interlayer surface hydration at the relatively dry conditions (Tuller & Or, 2001). Our hypothesis is that the hydroscopic or viscous nature of hydrogels changes capillary force, as well as the adsorptive water films. The effective pore size distribution is used as a measure of the capillary force. In addition, biopolymers normally contain substances acting as surfactants, which reduce surface tension of soil solution and play an important role in changing the shape of water retention curve. In this work, we used the R-TPSM model to fit experimentally measured water retention curves of hydrogel-mediated soil samples and their controls reported in literature. Analysis of the fitted model parameters provides insight to the possible mechanisms that are responsible for hydrogels’ effects on modifying water retention curve and hydraulic conductivity, which are discussed below.

#### 5.1.1 Hydrogels reduce surface tension

The fitted surface tension values from experimentally measured water retention curves consistently show reduction in surface tension for hydrogel-mediated soil samples compared with their controls. The model-fitted surface tension values (45.96 and 59.03 mN m$^{-1}$) of the treated Pepperbox sand and Lenni clay, which were incubated using *Bacillus subtilis* strain FB17 (Zheng et al., 2018), are similar to the values measured from the supernatants of suspensions prepared with the same bacteria (42.43 ± 1.30 mN m$^{-1}$) (see the supplemental material for experimental protocol); both show that surface tension of bacteria-incubated samples were significantly reduced compared with the control. The consistency between modeling and measurement confirms the important role of surface tension reduction in controlling water movement in soil. It also indicates that the R-TPSM correctly implements hydrogels’ effects on surface tension. Some discrepancies in the modeled and measured surface tension values are not surprising, as surface tension changes with the length of time and growth stage of bacteria, which were different in different studies.

The results of the sensitivity analysis highlight the important role that surface tension reduction plays on modifying soil water retention curve over the entire water content range. At the wet end, reduced surface tension facilitates drainage of large pores, as indicated by the less negative air-entry values for the treated soil samples than their controls (Table 2). On the dry end, lower surface tension reduces the amount of water retained in ducts and corners, thus reducing the overall retention (Figure 4a). These observations are consistent with experimental data from a study using Triton X100 as a model surfactant (Karagunduz, Young, & Pennell, 2015). Lower surface tension facilitates water drainage and thus decreases the
FIGURE 4  Sensitivity analysis of the roughness-triangular pore space model (R-TPSM) for water retention curve and relative hydraulic conductivity: the effects of (a, e) surface tension $\sigma$, (b, f) the roughness factor $X$, (c, g) the air-entry value $\mu_d$, and (d, k) the scale factor $\omega$. Other parameters were fixed at shape factor $\xi = 1$, $\omega = 5.456 \times 10^{-5}$, $X = 794.9$, $\mu_d = -4.825$ cm, and $\sigma = 0.0729$ N m$^{-1}$.

relative hydraulic conductivity (Figure 4e), and this effect is less pronounced at the drier end where conductivity is dominated by film flow and surface tension is not directly involved. It should be noted that we only considered the process of drainage in this study thus did not include the potential effects of hydrogels on wetting and wettability of a dry soil (e.g., contact angle change in the presence of hydrogels), which are relevant to imbibition.
5.1.2 Hydrogels induce changes in the effective pore size distribution

The treated soil samples show a general trend of shifting to a smaller mean pore size upon hydrogel impact (Figure 2). The effective pore size reduction could be due to swelling of hydrogels within pores under wetter conditions and shrinkage and formation of filaments as the polymer molecules retreat with the retreating air–water interface as soil dries due to drainage or evaporation. Formation of filaments has been directly visualized using scanning electron microscopy (SEM) or optical microscopy (Benard et al., 2019; Carminati et al., 2017; Zheng et al., 2018). Carles Brangari et al. (2017) accounted for this phenomenon and proposed a mechanistic modeling framework that assumed biofilm produced structure channels by converting a single cylindrical channel into multiple channels.

Reduction in the mean pore size as a result of a shifted effective pore size distribution in the presence of polymers reflects increased water retention dominated by capillary force. The shift to a smaller average pore size mainly affects the intermediate range of matric potential on the water retention curve and leads to higher saturation (Figure 4b). As the matric potential becomes more negative, the differences between water retention curves at various scale factors become less significant. Another observation is that although the changes in the effective pore size distribution in Hamra sand and Lenni clay were significant, water retention change in Hamra sand was not nearly as dramatic as in the Lenni clay soil (Figure 1). This may be due to the fact that the portion of pores clogged, especially of bigger pores, account differently for clay and for sand. In other words, clogging of bigger pores in clay will significantly change the water retention curve shape, whereas, because sand has much bigger pores, the shape of water retention curve may not be changed as significantly as clay even though some big pores are clogged.

Reduction in the mean pore size leads to a smaller saturated and unsaturated hydraulic conductivity, as the conductivity is proportional to the square of the pore radius (Equations 4 and 15). However, the relative hydraulic conductivity increases (Figure 4f) because of enhanced capillary water retention as a result of decreased pore sizes.

5.1.3 Hydrogels affect water film thickness and morphology

Water retention as adsorbed film is greater for hydrogel-treated soil samples than for their controls (Figure 1). Total film adsorption depends on both the thickness and total surface area of the water film. The total surface area is larger for hydrogel-treated soils than for their controls because the pore size distribution is shifted to have more smaller pores. On the other hand, the film thickness is calculated based on van der Waals interactions multiplying an effective model parameter X, which was originally used to represent the effects of physical roughness on film thickness (Zheng et al., 2015). We found that the treated samples had greater X values (i.e., increased film thickness) than their controls for Lenni clay, whereas the opposite trend was shown for the coarser soil samples. The discrepancy in the observed X values is due to the fact that when used to evaluate the effects of hydrogels in this study, the parameter X essentially encompasses two possible effects that lead to changes in film adsorption and conductance due to the existence of hydrogels on soil surfaces. The two possible effects counteract each other as follows: (a) hydrogels may increase film thickness due to polymer interaction (e.g., steric forces) in addition to van der Waals forces (Israelachvili, 1991), and (b) the surfactant molecules contained in hydrogels may decrease adsorption of thick films, which are controlled by capillary force due to surface roughness (Kibbey, 2013). The relative hydraulic conductivity increases due to enhanced film water retention.

5.1.4 Hydrogels increase viscosity of soil solution

Hydrogels’ viscous and hygroscopic nature is responsible for the increased saturated water content (Chenu & Roberson, 1996). The saturated water content is higher in the treated soil samples than in their controls, and the difference is more significant for fine-textured than for coarse-textured samples. Consistently, Kroener et al. (2018) reported that finer textured soil samples required less mucilage to increase saturated water content by 5%.

Recent measurements clearly suggest that hydrogels reduce saturated hydraulic conductivity (Kroener, Zarebanadkouki, Kaestner, & Carminati, 2014; Volk et al., 2016), because hydrogels are highly viscous with limited mobility and permeability in transporting water (Bottero et al., 2013; Knutson, Werth, & Valocchi, 2005; Rosenzweig et al., 2013). The viscous polymeric network also reduces flow conductance in ducts, corner, and films (Equations 4–6). However, the hydrogel effects on unsaturated hydraulic conductivity are more complex (Benard et al., 2019; Carminati et al., 2017). For Hamra sand, hydrogel reduced hydraulic conductivity across the measured potential range (Volk et al., 2016), but the conductivity at the drier end was greater for Reinhausen sandy loam amended with seed mucilage tested in Benard et al. (2019). Our modeling results show that the difference between treatments was smaller at the dry end for sandy loam than for sand. This
observation can be explained by the fact that there are two compensating factors (i.e., enhanced water retention and increased viscosity) controlling the unsaturated hydraulic conductivity. The enhanced water retention due to the abovementioned pore structure and size change can compensate for the higher viscosity, thereby increasing the unsaturated conductivity at the dry end (Carminati et al., 2017).

5.2 | The advantages and limitations of the R-TPSM

Two advantages of using the R-TPSM approach for modeling hydraulic properties of hydrogel-affected soils can be identified. First, the R-TPSM accounts for retention of water in soil pores in the form of water film and its contribution to flow. This improves the accuracy in estimating soil water retention curve and hydraulic conductivity, particularly in the low-water-content region. The significant contribution of water films to soil water retention and hydraulic conductivity has been increasingly recognized (Almquist, Brueck, Clarke, Wanzek, & Dragila, 2018; Kibbey, 2013; Tuller & Or, 2001; Zheng et al., 2015). Taking into account film flow provides an effective description of water dynamics from full saturation to complete dryness (Peters, 2013; Tuller & Or, 2001). Peters (2013) reported significant underestimation of water fluxes when neglecting film conductivity. Almquist et al. (2018) studied “thick film” due to surface roughness and addressed its importance in water availability to plants and contaminant transport. Water retention in the form of films has been attributed to adsorptive van der Waals forces (Tuller & Or, 2001; Zheng et al., 2015) or capillary force due to surface roughness (Almquist et al., 2018; Kibbey, 2013). The approach of using a roughness factor to account for the effects of roughness on water film was successfully applied in modeling hydraulic properties (Zheng et al., 2015) and estimating fluid–fluid interfacial area in unsaturated “clean” soils (Jiang, Guo, & Brusseau, 2020). When applied to hydrogel-mediated soils, the current R-TPSM treats the roughness factor X as an effective and fitting model parameter, which encompasses the possible hydrogel effects on changing water film thickness and area. An improved representation of hydrogels’ effects on water film thickness and morphology and a rigorous physical interpretation will require better understanding and quantification of the physical-chemical properties of soil-hydrogel systems.

The second advantage of the R-TPSM is that it accounts for multiple soil physicochemical properties (i.e., the effective pore size distribution, surface roughness, and surface tension) when assessing the effects of hydrogels. Changes in these factors due to hydrogels have been extensively discussed in hydrogel-mediated systems in experimental and theoretical studies. However, modeling efforts to date have mostly considered the factors individually and rarely simultaneously. For example, Carles Brangari et al. (2017) considered pore geometry change with suction due to swelling–shrinkage, and Kroener, Zarebanadkouki, Kaehtner, and Carminati (2014) included changes in osmotic potential induced by mucilage. By taking into account the factors simultaneously, the R-TPSM captures how hydrogels affect soil water behavior. At higher water contents, reduced surface tension facilitates water drainage and reduces hydraulic conductivity, whereas increased water film enhances water retention at the drier region.

The main limitation of the R-TPSM is that this approach builds on the capillary bundle model, which oversimplifies the complexity of porous media, so the possible interpor transport mechanisms (tortuosity and connectivity) are not considered (Hunt, Ewing, & Horton, 2013). In addition, the model in its current form is incapable of taking into account the transient nature of viscosity and surface tension, which depends on hydrogel concentration and water content. As we learn to better characterize the physical-chemical properties of biopolymer systems (e.g., surface tension, contact angle, the surface interaction involving polymers, viscosity), the model will need to be modified to improve its ability to capture the physics of the hydrogel-modified systems more accurately and to make the model parameters more meaningful, rather than mostly empirical.

6 | CONCLUSIONS

In this study, we extended the application of a previously developed triangular pore space model that takes into account surface roughness (i.e., the R-TPSM) to simulate soil water retention function and hydraulic conductivity in hydrogel-mediated soils. We demonstrated the R-TPSM’s capability to account for and study hydrogels’ effects on the effective pore size distribution and surface tension (capillary water retention) as well as film adsorption through comparison between modeling and experimental results. Hydrogels’ hygroscopic properties (i.e., swelling when wet), as well as their ability to form filaments between particles that creates additional media and capillary force (when dry), are likely responsible for reducing mean pore size. The shift in pore size distribution illustrates the capillary effects induced by the formation of a hydrogel–soil network matrix. Additionally, the model captures the hydrogels’ effect on surface tension, which has not been considered explicitly in previous modeling attempts, and shows how surface tension can significantly affect the shape of water retention curve and unsaturated hydraulic conductivity. Despite the semiempirical nature of the R-TPSM, this study nevertheless clearly demonstrates the
necessity of considering surface property changes when modeling hydraulic properties in hydrogel-affected soils.

This study improves the mechanistic understanding of how hydrogels modify soil hydraulic properties and highlights the importance of hydrogel-induced changes in surface tension and effective pore size distribution in shaping soil water retention function and hydraulic conductivity. The findings contribute to the overall efforts in revealing biophysical control in rhizosphere hydrology, mediated by exudates from roots, bacteria, fungi, and soil fauna. Such efforts will aid development of water management strategies to improve water use efficiency by root zone soil and to increase crop production under restricted water availability.

CONFLICT OF INTEREST
The authors declare no conflict of interest.

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