Soil consistency and interparticle characteristics of xanthan gum biopolymer–containing soils with pore-fluid variation

Ilhan Chang, Yeong-Man Kwon, Jooyoung Im, and Gye-Chun Cho

Abstract: Biopolymer–soil technology is currently recognized as an environmentally friendly soil improvement method for geotechnical engineering practices. However, concerns exist regarding biopolymer fine-soil applications because the performance of biopolymers is based on an electrical interaction with clay or a pore fluid. Thus, the effect of water content and pore-fluid chemistry on biopolymer behavior in soil must first be clarified in terms of biopolymer applications. In this study, the liquid limits of xanthan gum biopolymer–treated clay–sand mixtures (clayey silt, kaolinite, montmorillonite, and sand) were obtained using three chemically distinct pore fluids (deionized water, 2 mol/L NaCl brine, and kerosene). Xanthan gum has contrary effects to the soil consistency, where the liquid limit can decrease via xanthan gum–induced particle aggregation or increase due to xanthan gum hydrogel formation. The clay-mineral type governed the xanthan gum behavior in the deionized water, while the pore-fluid chemistry governed the xanthan gum behavior in the brine and the kerosene.

Key words: xanthan gum biopolymer, pore-fluid chemistry, electrical sensitivity, soil classification, liquid limit.

Introduction

The use of exocultured biological materials, known as “biopolymers”, has been introduced to overcome the limitations of microbe endocultivation in soil in the form of a newly utilized environmentally friendly construction material for geotechnical engineering applications (Chang et al. 2016a; Latifi et al. 2017).

Recent studies have reported a promising biopolymer effect that improves the strength and stability of soils (Chang et al. 2016b; Ferruzzi et al. 2000; Orts et al. 2007). Microbial biopolymers show erosion reduction against surface water runoff in water-fronts (Ham et al. 2016, 2018) and agricultural farm lands (Orts et al. 2000), while strengthening and interparticle interaction behaviors of the polysaccharide-type biopolymers, such as dextran, beta-glucan, xanthan gum, and gellan gum, with different soil types have been investigated by many researchers (Chang and Cho 2012, 2014; Chang et al. 2015a, 2015c, 2016b, 2017; Kwon and Ajo-Franklin 2013). Biopolymers synthesized from lignin, starch, and acrylamide can decrease the decomposition of plant residues (Awad et al. 2012) or even promote vegetation growth in soils (Chang et al. 2015d). Moreover, biopolymers produced from food waste (e.g., bovine milk) can contribute to the reduction of dairy and relevant food waste (Chang et al. 2018). Furthermore, biopolymers can become an alternative earth construction binder in regions where ordinary cement is scarce and expensive, such as Africa (Chang et al. 2015b).

However, application of biopolymers to soil treatment requires considerations of the biopolymer rheology, phase transfer with water content variation, biopolymer to soil ratio, soil type, and pore-fluid condition, which affects the biopolymer–soil matrix formation (Chang and Cho 2019; Chang et al. 2015c, 2017). Biopolymer-treated soils with a high water content show low strength due to the low viscous biopolymer hydrocolloids surrounding soil particles (Chang et al. 2017), while dehydration increases the soil strength due to the condensation of the biopolymer hydrogels forming high-tensile-strength biofilms through the soil particulate network (Chang et al. 2016a). For biopolymer-treated sands, the concentration of biopolymer hydrogels be-
comes the dominant factor controlling the strength behavior of sands (Chang et al. 2016a; Lee et al. 2017). Meanwhile, electrostatic and chemical bonding characteristics between biopolymers and clayey particles become important for fine soils, where the “biopolymer-to-clay content” (in mass) becomes more critical than the “biopolymer-to-total soil content” (in mass) in terms of the strengthening purpose for biopolymer-treated soils (Chang and Cho 2019; Chang et al. 2015a).

Soil consistency (plastic limit; PL; liquid limit, LL; and plasticity index, PI) not only indicates the soil–water interaction on the particle surfaces, but also represents the pore-fluid chemical environment in pore spaces (Jang and Santamaria 2016). Biopolymer-treated soils show different LL behaviors, where the presence of biopolymer can increase (hydrogel swelling) or decrease (via particle aggregation) the LL of soils (Chang and Cho 2014; Nugent et al. 2009).

Xanthan gum is one of the most common biopolymers used in geotechnical engineering applications, such as soil stabilization (Chang et al. 2015c; Im et al. 2017; Latifi et al. 2017; Lee et al. 2017), mine tailings treatment (Chen et al. 2016), and permeability control (Cabalar et al. 2017; Martin et al. 2016). However, most studies have focused on the macrobehavior of xanthan gum–treated soils without detailed consideration of the microscale soil–fluid interaction. The particle aggregation and water absorption characteristics of xanthan gum hydrogels seem to mainly affect the geotechnical behavior of fine soils. Thus, this study aims to investigate soil behaviors when xanthan gum hydrogels become the main pore-filling medium. The electric conductivity (σel) and relative permittivity (κ′) distinguish pore-fluid chemistry. Deionized water (κ′ = 80, σel < 5 μS/m, pH = 6.5) was obtained using the reverse-osmosis process. The brine solution (κ′ = 55, σel = 12 μS/m; pH = 6.5) was prepared by dissolving sodium chloride (CAS No. 1310-58-3; Junsei Chemical) into deionized water by using a magnetic stirrer to obtain a 2 mol/l NaCl solution. Common kerosene (κ′ = 2, σel = 10−11 S/m; GS Caltex) was used. The pH and σel of deionized water and 2 mol/l NaCl brine were measured with a pH/conductivity meter (S470-USP-K; Mettler Toledo), while the values of σel of kerosene and κ′ of all fluids were obtained from Jang and Santamaria (2016).

Materials and methods

Materials

Sand: jumunjin sand

“Jumunjin” sand (standard sand material of Korea), which is a type of quartz-based (SiO2) poorly graded sand (SP, classified in accordance with ASTM 2017b) (Kim et al. 2017), is used in this study. The surface of jumunjin sand is regarded to be electrically neutral; therefore, the interaction between jumunjin sand and biopolymer is not regarded to be influenced by the pore-fluid chemistry, while soil consistency is mainly governed by the clay fraction (Jang and Santamaria 2016).

Fine soils: clayey silt, kaolinite, and montmorillonite

Clayey silt is known to have low plasticity development when mixed with water, where its LL and PL are mostly lower than those of clay (Mitchell and Soga 2005). The clayey silt used in this study is sampled from Danyang, Korea, and its basic properties are listed in Table 1. Kaolinite clay is more hydrophilic than sand and silt, which forms strong hydrogen bonds with water molecules. However, as kaolinite has a stable 1:1 layer structure of gibbsite and silica sheets, kaolinite becomes less sensitive to fluid chemistry than montmorillonite (Santamaria et al. 2001). Bintang kaolinite (CEC = 6.1 meq/100 g; Belitung Island, Indonesia) is used in this study (Table 1).

Montmorillonite is a smectite clay consisting of two layers of silica sheets and one layer of gibbsite with exchangeable cations (e.g., sodium or calcium) at interlayers, where a large amount of water can occupy the interlayer spaces and swell (Das 2013; Hensen and Smit 2002). The surface of montmorillonite is negatively charged by isomorphous substitutions, resulting in a high cation-exchange capacity (CEC) (Sondi et al. 1996) and accompanying thick double layer (Taylor 1985), which renders the consistency of montmorillonite to be higher than kaolinite. A research grade montmorillonite material (CAS:1302-78-9; CEC = 64.1 meq/100 g; Sigma Aldrich) is used for this study (Table 1).

Table 1. Basic properties of soils.

<table>
<thead>
<tr>
<th>Soil</th>
<th>D50 (μm)</th>
<th>SSA (m2/g)a</th>
<th>CEC (meq/100 g)b</th>
<th>PL (%)</th>
<th>LL10</th>
<th>LLB</th>
<th>LLK</th>
<th>USCSc</th>
<th>Clay-to-sand ratio</th>
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<tbody>
<tr>
<td>Jumunjin sand</td>
<td>421</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>100:0</td>
</tr>
<tr>
<td>Clayey silt</td>
<td>12.5</td>
<td>2.2</td>
<td>—</td>
<td>23.8</td>
<td>30.5</td>
<td>28.8</td>
<td>32.1</td>
<td>SP</td>
<td>50:50</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>3</td>
<td>22.1</td>
<td>6.1</td>
<td>31</td>
<td>69.8</td>
<td>50.9</td>
<td>93.2</td>
<td>CH</td>
<td>20:80</td>
</tr>
<tr>
<td>Montmorillonite</td>
<td>0.07c</td>
<td>220.6</td>
<td>64.1</td>
<td>61.6</td>
<td>398.5</td>
<td>94.2</td>
<td>60.5</td>
<td>CH</td>
<td>10:10</td>
</tr>
</tbody>
</table>

— Specific surface area (SSA) obtained by the methylene blue adsorption method (Santamaria et al. 2001).
— Cation-exchange capacity (CEC) obtained by the methylene blue cation–exchange capacity method (Inglethorpe et al. 1993).
— Unified Soil Classification System (ASTM 2017).

According to the Unified Soil Classification System (ASTM 2017), the surface of jumunjin sand is regarded to be electrically neutral, so the soil–water interaction is not considered. However, most studies have focused on the macrobehavior of xanthan gum–treated soils without detailed consideration of the microscale soil–fluid interaction. The particle aggregation and water absorption characteristics of xanthan gum hydrogels seem to mainly affect the geotechnical behavior of fine soils. Thus, this study aims to investigate soil behaviors when xanthan gum hydrogels become the main pore-filling medium. specifically, the effect of xanthan gum on soil consistency are evaluated through laboratory tests under different pore-fluid conditions.
before experimental procedures, according to ASTM D2216 (ASTM 2007). Then, each type of soil was thoroughly mixed with dry xanthan gum to obtain xanthan gum–soil mixtures with 0.1%, 0.5%, 1.0%, and 2.0% xanthan gum–to-soil mass content \((m_b/m_s)\), while untreated soils \((m_b/m_s = 0\%)\) were prepared simultaneously.

**Atterberg limit (PL and LL) measurements**

The PL values of the soils were determined using a thread-rolling device (unglazed paper attached to the top and bottom plates with a controlled minimum spacing of 3.2 mm) to ensure consistent repeatability and reliability according to ASTM D4318 (ASTM 2017a). Five measurements were conducted for each soil condition to minimize measurement errors.

The LL values of soils were determined through a fall cone test (British Standards Institution 2017; Hansbo 1957; Koumoto and Houlsby 2001) by using a cone with a mass of 80 g and a tip angle of 30°, where the penetration depth was measured for 5 s by using a dial gauge with a precision of 0.01 mm. The LL values were determined according to the water content when the cone penetration was 20 mm into the soil sample. Other testing procedure details were followed by referring to BS EN ISO 17892-6:2017 (British Standards Institution 2017). Fall cone tests were repeated at least three times for a single soil condition to obtain reliable averages.

**Results and analyses**

The values of LL with deionized water \((LL_{DI})\), 2 mol/L NaCl brine \((LL_B)\), and kerosene \((LL_K)\) of xanthan gum–treated soils with variations in xanthan gum content are summarized in Figs. 2, 3, and 4 for clayey silt–sand, kaolinite–sand, and montmorillonite–sand soils, respectively. For all soil conditions, higher fine contents show higher LL values regardless of the pore-fluid condition.

**LL behavior of untreated fine soils with pore-fluid variation**

Particle–pore-fluid interactions of untreated fine-grained soils depend on particle mineralogy and pore-fluid chemistry (Jang and Santamarina 2016). The amount of water absorbed on the clay surface is affected by the amount of surface charge and particle size of clay minerals (Sridharan et al. 1986, 1988; White 1949), which alters the particle packing of clays (Chang and Cho 2010). Untreated clayey silt with low surface charges shows no significant change in LL values (i.e., \(LL_{DI} = LL_B\)) (Jang and Santamarina 2016; Mitchell and Soga 2005). As montmorillonite has a higher CEC than kaolinite, the \(LL_{DI} \rightarrow LL_B\) reduction of montmorillonite (Figs. 4a and 4b) is also larger than that of kaolinite (Figs. 3a and 3b).

Under a nonwetting pore-fluid condition (kerosene), edge charge and van der Waals forces dominantly affect particle–pore-fluid interactions (Jang and Santamarina 2016; Sridharan and Prakash 1999). Kaolinite has a higher proportion of edge charges (12%–36% of total area) than montmorillonite (<5% of total area), which results in \(LL_{DI} < LL_K\) for kaolinite (Fig. 3), while montmorillonite shows \(LL_{DI} \gg LL_K\) (Fig. 4) owing to the higher edge charge and van der Waals force interaction for kaolinite (Brady et al. 2005).
The LL behaviors of untreated fine soils obtained from this study are in accordance with the findings of Jang and Santamarina (2016).

**LLD, LLD, and LLK of xanthan gum–treated clayey silt–sand soils**

For clayey silt–sand soils, xanthan gum mainly interacts with pore fluids in the form of hydrophilic hydrogel owing to negligible surface charges of silt and sand (Mitchell and Soga 2005), where xanthan gum hydrogel gradually increases both LLD (Fig. 2a) and LLD (Fig. 2b) of clayey silt–sand soil with xanthan gum contents. The LLK values remain constant, regardless of the content of xanthan gum (Fig. 2c).

**LLD, LLD, and LLK of xanthan gum–treated kaolinite–sand soils**

Kaolinite–sand soils show distinctive LLD behaviors where LLD varies with xanthan gum content increase, as shown in Figs. 3a and 3d. In particular, LLD initially increases (3) to a peak point (2; $m_b/m_c = 0.5\%$ for K10), decreases to an inflection point (4; $m_b/m_c = 1.0\%$ for K10), and then seems to remain constant up to $m_b/m_c = 2\%$ (4).

Figure 3d represents the variation in LLD in terms of xanthan gum–to-clay (kaolinite) mass content ($m_b/m_c$). Variation of LLD with $m_b/m_c$ implies the hydrogel viscosity increase and particle aggregation effects induced by xanthan gum (Nugent et al. 2009). At low xanthan gum contents, xanthan gum instantly forms hydrogels, which results in the increase in LLD, as shown in Fig. 3d. All kaolinite-sand soils show a peak LLD at $m_b/m_c = 0.5\%$, which is in accordance with the previous finding from Chang and Cho (2019) addressing $m_b/m_c$ to mainly govern biopolymer-clay matrix formation and relevant shear strength properties (i.e., cohesion and friction angle) (Chang and Cho 2019). As the content of xanthan gum increases, xanthan gum initiates kaolinite aggregation via ionic or hydrogen bonding (Laird 1997; Sastry et al. 1995; Theng 2012), which accompanies a decrease in LLD (3 in Fig. 3d). The behavior of LLD for $m_b/m_c > 1\%$ (4; Fig. 3d) seems to be attributed to the equilibrium between xanthan gum hydrogel formation and the simultaneous kaolinite aggregation induced by xanthan gum (Nugent et al. 2009). Meanwhile, LLK increases gradually with the content of xanthan gum (Fig. 3b). Xanthan gum shows no effect on the variation in LLK (Fig. 3c), mainly due to the absence of xanthan gum hydrogel formation.

**LLD, LLD, and LLK of xanthan gum–treated montmorillonite–sand soils**

For montmorillonite–sand soils, LLD decreases gradually with xanthan gum content (Fig. 4b). Montmorillonite shows high hydrophilicity owing to its high SSA and CEC, where xanthan gum hydrogel seems to have less effect on the variation in pore-fluid viscosity, while xanthan gum–induced montmorillonite aggregation seems to be the main factor decreasing LLD. The edge sites of montmorillonite are regarded as negligible owing to the low thickness-to-length ratio (<0.01) (Santamarina et al. 2001; Secor and Radke 1985), where xanthan gum can mainly interact within the interlayer spaces of montmorillonite (Deng et al. 2006; Laird 1997; Theng 2012) or form hydrogen bonds with water molecules in the Gouy diffuse layer (Ng and Plank 2012), resulting in the reduction of the double-layer thickness and accompanying LLD.
decrement with xanthan gum content (Fig. 4a) (Hayes and Swift 1978; Rao et al. 1993).

Meanwhile, LLB (Fig. 4b) follows similar behaviors under relevant pore-fluid conditions of clayey silt–sand and kaolinite-sand soils. Xanthan gum does not seem to have any effect on LLK (Fig. 4c), similar to other types of soils.

Discussion

LL behavior of xanthan gum–treated fine soils with pore-fluid variation

The effect of xanthan gum on LL_{DS} varies with different soil types, as shown in Figs. 3–5. Brine pore fluid reduces the LL of clayey soils from LL_{DS} to LL_{B} (Figs. 3 and 4), while silt shows no effect (LL_{DS} = LL_{B}; Fig. 2). Figure 5 expresses the increment in LL_{B} of soils (LL_{B,0}−LL_{B,X}) according to xanthan gum treatment, where LL_{B,0} denotes the LL_{B} at 0% xanthan gum treatment and LL_{B,X} denotes the LL_{B} of untreated soils. A similar increase in LL_{B} with xanthan gum treatment, regardless of the soil type, implies that xanthan gum mainly forms hydrogels that can be attributed to the increase in viscosity of pore fluids (Nugent et al. 2009) or interacting with concentrated Na^{+} ions in the reduced double-layer under brine pore-fluid conditions (Theng 2012).

The insignificant variation in LL_{K} with xanthangum treatment, shown in Figs. 2c, 3c, and 4c, implies the importance of water for the hydrogel formation of xanthan gum and the accompanying interactions with soil particles and pore fluids. Thus, it can be concluded that hydrogel formation is an important prerequisite for xanthan gum in soil treatment and ground improvement practices.

Xanthan gum biopolymer – pore-fluid effect on classification of soils

Fine-grained soils (L10, L5S5, K10, K5S5, M10, and M5S5) used in this study were plotted on the LL–PI plane (Fig. 6a). Xanthan gum treatment in clayey silt tends to increase both LL and PI, which alters the Unified Soil Classification System (USCS; ASTM 2017b) classification from ML to CL. Both the LL and PI of kaolinite-based soils instantly increase at low xanthan gum contents (m_{bd}/m_{s} = 0.5% for K10; m_{bd}/m_{s} = 0.25% for K5S5) and decrease at higher xanthan gum contents. Most kaolinite-based soils are classified as clay (CH for K10; CL for K5S5), while some are classified as silt (e.g., MH for K10 with m_{bd}/m_{s} = 1%; ML for K5S5 with m_{bd}/m_{s} = 1%). Montmorillonite-based soils are classified as clay with high plasticity (CH), regardless of the content of xanthan gum, while both LL and PI tends to decrease with xanthan gum treatment.

Meanwhile, as xanthan gum treatment seems to alter the USCS classification of fine soils, the pore-fluid effect (e.g., pH, electrical conductivity, and permittivity) has been considered based on the electrical sensitivity (S_E) and LL_{B} values according to Jang and Santamarina (2016).

Figure 6b displays all soils used in this study plotted on the S_E–LL_{B} chart with similar references (silica flour, kaolinite-based soils, montmorillonite-based soils) from previous studies (Jang and Santamarina 2016; Park and Santamarina 2017). LL_{B} distinguishes the plasticity of soils from none to high plasticity, while S_E is classified into low, intermediate, and high S_E conditions (Fig. 6b). Both the plasticity and S_E increase with higher clay contents and soil type variations in the following order: clayey silt → kaolinite → montmorillonite. Xanthan gum treatment induces an increase in soil plasticity owing to the formation of viscous hy-

Fig. 4. Liquid limit of xanthan gum–treated montmorillonite–sand soils with variation in m_{bd}/m_{s}: (a) deionized water pore fluid; (b) 2 mol/L NaCl brine pore fluid; (c) kerosene pore fluid.

Fig. 5. Liquid limit increment by xanthan gum content in brine condition.
drogel when using brine pore fluid, while $S_E$ initially decreases and then increases with xanthan gum contents. The values of $S_E$ of clayey–silt soils are mainly low, while in some cases, these values are found near the boundary between intermediate and low $S_E$. For kaolinite–sand soils, the minimum $S_E$ is observed at a $m_b/m_c$ of 0.2%, 1%, and 2% for K2S8, K5S5, and K10 soils, respectively. With respect to xanthan gum–to-clay mass content ($m_b/m_c$), the lowest $S_E$ points are observed at $m_b/m_c$ values of 1%, 2%, and 2% for K2S8, K5S5, and K10 soils, respectively. For montmorillonite–sand soils, $S_E$ decreases up to $m_b/m_c = 4\%$ (2% xanthan gum content in M5S5). The overall $S_E$ reduction of clays (K10, K5S5, M10, M5S5) with xanthan gum treatment seems to be related to xanthan gum–induced clay particle aggregation, where an advanced technique (e.g., liquid-cell microscopy on aqueous phase xanthan gum–clay–pore fluids) can be used for further investigation.

**Conclusion**

Pore-fluid chemistry and clay-mineral type are salient factors in xanthan gum biopolymer treatment in fine soils. However, xan-
than gum behavior variations according to the pore-fluid chemistry and clay mineral have not been analyzed in previous studies. Therefore, this study addresses this gap in the research.

Liquid limits of xanthan gum–treated soils were measured using chemically distinctive pore fluids. Under deionized-water conditions, xanthan gum behavior was affected by the clay type. With the clayey silt, xanthan gum formed viscous hydrogel in the pore space among clay particles, and LL increased as xanthan gum–clay content increased. With kaolinite, LL peaked at a xanthan–gum–clay content of 0.5%, and decreased until a content of 1.0% was reached, which implies the simultaneous hydrogel formation in pores and particle aggregation induced by xanthan gum. With montmorillonite, LL decreased gradually with an increase in xanthan gum–clay content, which implies xanthan gum mainly aggregates montmorillonite particles rather than affecting the viscosity of the pore fluid. The overall electrical sensitivity reduction with xanthan gum treatment implies that interparticle aggregation is the main phenomenon of clays at low xanthan gum–clay contents. In contrast, xanthan gum behavior was governed by pore-fluid chemistry in brine and kerosene conditions, regardless of clay type. With brine, counter-ion decreased LL of clays significantly, while LL tended to increase with higher xanthan gum–clay content. For kerosene, LL showed no remarkable changes with xanthan gum treatment.

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List of symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tr>
<td>CEC</td>
<td>Cation-exchange capacity</td>
</tr>
<tr>
<td>Dm</td>
<td>Mean grain size</td>
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<tr>
<td>LL</td>
<td>Liquid limit of soils</td>
</tr>
<tr>
<td>LLa</td>
<td>Liquid limit of soils with 2 mol/L NaCl brine pore fluid obtained by laboratory fall cone test</td>
</tr>
<tr>
<td>LLd</td>
<td>Liquid limit of soils with deionized water pore fluid obtained by laboratory fall cone test</td>
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<tr>
<td>LLk</td>
<td>Liquid limit of soils with kerosene pore fluid obtained by laboratory fall cone test</td>
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<tr>
<td>mbi</td>
<td>Mass of biopolymer (xanthan gum, in this study)</td>
</tr>
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<td>mbi/mcs</td>
<td>Xanthan gum–to-clay ratio in mass</td>
</tr>
<tr>
<td>mcs/mci</td>
<td>Xanthan gum–to-soil ratio in mass</td>
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<td>Mass of clay</td>
</tr>
<tr>
<td>mso</td>
<td>Mass of soil</td>
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<tr>
<td>PL</td>
<td>Plastic limit of soils</td>
</tr>
<tr>
<td>PI</td>
<td>Plasticity index (LL – PL) of soils</td>
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<tr>
<td>S_e</td>
<td>Electrical sensitivity</td>
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<td>Specific surface area of soil particles (m²/g)</td>
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<td>Relative permittivity</td>
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