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23 Abstract: Soil hydraulic conductivity depends not only on the state variables such as void ratio 24 and pore size distribution, but also on its physical/geotechnical properties. In this study, Atterberg limits, hydraulic conductivity and mercury intrusion porosimetry (MIP) tests were 25 performed on MX80 bentonite/Callovo-Oxfordian (COx) claystone mixtures. Based on the 26 27 experimental results obtained in this study together with the test data compiled from literature, a 28 general capillary model considering  $N_p$  pores in series was used to evaluate the influence of soil 29 plasticity on the relationship between hydraulic conductivity and pore size distribution. It was found 30 that the best pore interconnection parameter  $N_p$  was highly dependent on the soil property. The 31 larger the plasticity index, the larger the best  $N_p$  value and the more complex the pore 32 interconnection. The relationship between the best  $N_p$  value and plasticity index could be well described by an exponential equation. The general capillary model was then improved to estimate 33 34 the hydraulic conductivity of different soils. The estimated values using the improved general capillary model were finally compared with the measured ones and the good agreement between the 35 measurement and estimation revealed the good performance of the proposed model. 36

37 Keywords: bentonite/claystone mixture; plasticity index; pore size distribution; hydraulic
38 conductivity; general capillary model

#### 39 **1 Introduction**

Water flow through soils is a matter of concern in many geotechnical, agricultural and 40 environmental engineering practices, for instance, in the coupled consolidation analysis of clay 41 foundation improvement and land reclamation with dredged clay (Yin 2009; Zeng et al. 2019a), in 42 43 the design of isolating barriers in waste disposal facilities (Chapuis 1990) as well as in the 44 assessment of water infiltration rate and percolation depth into agricultural land (Saffih-Hdadi et al. 45 2009). Obviously, this percolation process is dominated by the hydraulic conductivity of soils based on Darcy' law. It is therefore essential to quantitatively evaluate the hydraulic conductivity of soils 46 47 for the geotechnical, agricultural and environmental issues.

48 In general, the hydraulic conductivity of saturated soils is experimentally measured using 49 constant-head or fall-head methods (Deng et al. 2011). It can also be indirectly determined from the 50 consolidation curve based on the Terzaghi's consolidation theory (Terzaghi 1943). It has been well documented that the hydraulic conductivity can be influenced by many factors, including the 51 chemical properties of fluid, void ratio, mineralogy, grain size and shape, grain packing and 52 53 orientation, pore size distribution and pore interconnection (Yin et al. 2009; Ren et al. 2016; Teng et 54 al. 2019). Several investigators attempted to develop some empirical and theoretical models to predict the hydraulic conductivity from easily determinable physical characteristics of soils 55 56 (Lapierre et al. 1990; Dolinar 2009). In most empirical models, the hydraulic conductivity was related to the selected properties of soils, such as void ratio (Berilgen et al. 2006; Dolinar 2009), 57 effective grain size (Chapuis 2004) and representative pore size (Deng et al. 2015). However, the 58 59 applicability of these empirical models to other soils needs further evaluation. Taking the channels 60 of water flow through a cross section as uniform tubes, Kozeny (1927) proposed a theoretical model

61 known as Kozeny-Carman equation, which was further improved by Carman (1956):

62 
$$k = C_{\rm F} \frac{1}{S_{\rm s}^2} \frac{\rho_{\rm w}g}{\mu \rho_{\rm s}^2} \frac{e^3}{1+e}$$
(1)

63 where k is the hydraulic conductivity;  $C_{\rm F}$  is a dimensionless shape constant;  $S_{\rm s}$  is the specific 64 surface area;  $\rho_{\rm w}$  and  $\mu$  are the density and viscosity of water, respectively; g is the gravitational 65 acceleration. Its performance was validated for sand (Ren et al. 2016). For clayey soils, which were 66 generally characterized by an aggregated structure, pores could vary from very fine (intra-aggregate) 67 to very coarse (inter-aggregate) (Kong et al. 2018). In that case, the pore size distribution became a 68 crucial factor to be accounted for.

Based on the Poiseuille's equation for laminar flow through a cylindrical capillary, a capillary
 model was developed by Garcia-Bengochea et al. (1979) assuming parallel and cylindrical
 nonintersecting pores with different diameters:

72 
$$k = \frac{\rho_{\rm w}gn}{32\mu} \sum_{i=1}^{m} d_i^2 f(d_i)$$
(2)

73 where *n* is the porosity of soil; *m* is the total number of pore intervals; *i* is the counter from 1 to *m*;  $d_i$  and  $f(d_i)$  are the pore diameter and volumetric probability corresponding to i, respectively. In 74 75 addition to the pore size distribution, the pore interconnection between soil sections could also influence the water flow. Childs and Collis-George (1950) and Marshall (1958) introduced a 76 77 probabilistic approach to address this issue. Fig. 1a shows the water flow path through a capillary network in a soil fabric with two in-series sections having the same porosity n for a given length. 78 The probability that a pore of diameter  $d_1$  from one section joins a pore of diameter  $d_2$  of another 79 80 section is  $[nf(d_1)][nf(d_2)]$ . According to the basic idea of Poiseuille's equation, the smallest 81 capillary between two capillaries in series governs the water flow through the capillaries (e.g.  $d_1$ 

from a series of d<sub>1</sub> and d<sub>2</sub> in Fig. 1a) (Watabe et al. 2006). Correspondingly, Garcia-Bengochea et al.
(1979) derived the following equation (known as the Marshall Model) for hydraulic conductivity:

84 
$$k = \frac{\rho_{w}gn^2}{32\mu} \sum_{i_1=1}^{m} \sum_{i_2=1}^{m} \overline{d}^2 f(d_{i_1}) f(d_{i_2})$$
(3)

where  $i_1$  and  $i_2$  are the counters from 1 to m;  $d_{i_1}$  and  $d_{i_2}$  are the pore diameters corresponding to 85  $i_1$  and  $i_2$ , respectively;  $\overline{d}$  is the smaller one of  $d_{i_1}$  and  $d_{i_2}$ ;  $f(d_{i_1})$  and  $f(d_{i_2})$  are the volumetric 86 probabilities of occurrence of pores with diameters  $d_{i_1}$  and  $d_{i_2}$ , respectively. When a more complex 87 88 soil fabric is considered, the water flow path through a capillary network in the soil fabric is schematically shown in Fig. 1b. The probability that a pore of diameter  $d_{i_1}$  from one section joins 89 other pores of diameters  $d_{i_2}, d_{i_3}, \dots, d_{i_{N_p}}$  from other sections is  $[nf(d_{i_1})][nf(d_{i_2})]\cdots[nf(d_{i_{N_p}})]$  (e.g. 90  $[nf(d_1)][nf(d_2)][nf(d_3)][nf(d_4)][nf(d_5)][nf(d_6)]$  in Fig. 1b) and the diameter of the dominant flow 91 channel  $\overline{d}$  is the smallest one of  $d_{i_1}, d_{i_2}, \dots, d_{i_{N_p}}$  (e.g.  $d_6$  from a series of  $d_1, d_2, d_3, d_4, d_5$  and  $d_6$  in 92 Fig. 1b). N<sub>p</sub> refers to the number of in-series pores in different sizes along a capillary water path for 93 94 a given length. Under this circumstance, a general capillary model was proposed by Watabe et al. 95 (2006; 2011):

96 
$$k = \frac{\rho_w g n^{N_p}}{32\mu} \sum_{i_1=1}^m \sum_{i_2=1}^m \cdots \sum_{i_{N_p}=1}^m \overline{d}^2 f(d_{i_1}) f(d_{i_2}) \cdots f(d_{i_{N_p}})$$
(4)

97 where  $i_1, i_2, \dots, i_{N_p}$  are the counters from 1 to *m*. The capillary and Marshall models correspond to 98 Eq. (4) when  $N_p=1$  and  $N_p=2$ , respectively. As a first attempt, Watabe et al. (2011) applied the 99 general capillary model to the hydraulic conductivity prediction for glacial tills, sandy soils and 100 clayey soils. They found that the  $N_p$  value which gives the best prediction was not the same for 101 different soils. However, the influence mechanism of soil property on the hydraulic conductivity 102 was not investigated in-depth.

103	In France, the clay-rich Callovo-Oxfordian (COx) sedimentary rock formation situated in the
104	Meuse/Haute Marne site between 400 and 600 m depth was selected as the host rock to install the
105	future repository for disposing radioactive wastes by the French National Agency for Nuclear Waste
106	Management (ANDRA) (Andra 2005). The excavation of the underground tunnels for waste storage
107	will produce a large amount of excess COx claystones to be dealt with. To reduce the financial costs
108	and to better ensure the compatibility of chemistry with the host rock, the mixtures of the excavated
109	COx claystone and MX80 bentonite were proposed to seal and backfill the underground drifts and
110	shafts (Zeng et al. 2019b; 2020). After the installation in the repository, the selected mixtures are
111	expected to exhibit permeability low enough to ensure the safety functions of the deep geological
112	repository. Therefore, a comprehensive understanding of the permeability of bentonite/COx
113	claystone mixtures with different bentonite fractions is essential to meet the performance target and
114	to optimize the sealing/backfilling element design.

115 In this study, a series of Atterberg limits, constant-head hydraulic conductivity and mercury 116 intrusion porosimetry (MIP) tests were carried out on MX80 bentonite and COx claystone mixtures. The plasticity index, hydraulic conductivity and pore size distribution of the mixtures with various 117 118 bentonite fractions were determined. The general capillary model was applied to evaluate the hydraulic conductivities of bentonite/claystone mixtures together with those of other soils compiled 119 120 from literature. The effect of plasticity index on the best  $N_{\rm p}$  value was quantitatively determined, 121 allowing an improved general capillary model to be proposed for the hydraulic conductivity 122 estimation of different soils.

#### 123 2 Materials and methods

124 2.1 Materials

125 The MX80 bentonite was taken from Wyoming in the USA, while the COx claystone was extracted 126 at a depth of around 490 m in the Underground Research Laboratory (URL) operated by ANDRA. The bentonite and claystone were crushed to less than 2 mm. Their grain size distributions 127 determined by dry-sieving and hydrometer methods are shown in Fig. 2. The fractions of fine grains 128 129 (< 0.08 mm) determined by dry-sieving method are 19% and 0.3%, while the clay contents (< 2  $\mu$ m) 130 determined by hydrometer method are 86% and 26%, for bentonite and claystone respectively. 131 Based on the X-ray diffraction analysis, the bentonite consists of montmorillonite (86%) and quartz (7%), whereas the claystone comprises interstratified illite-smectite (40-45\%), carbonates (30\%) as 132 133 well as quartz and feldspar (25-30%).

Table 1 summarizes the chemical composition of the pore water extracted from the ANDRA URL. To simulate the working environment of the bentonite/claystone mixtures, synthetic water was used in the Atterberg limits and hydraulic conductivity tests. It was prepared by mixing the components (Table 1) with deionized water until full dissolution.

#### 138 2.2 Determination of index properties

139 Prior to the experiments, the bentonite and claystone, at their initial water contents of 11.4% and 140 6.1%, were first mixed together with different proportions of 10/90, 20/80, 30/70, 50/50 and 70/30 in dry mass. Subsequently, the specific gravity  $(G_s)$  values of bentonite, claystone and their 141 142 mixtures were measured using the pycnometer method in accordance with ASTM D854-10 (2010). 143 Referring to ASTM D4318-10 (2014), the liquid limit ( $w_L$ ) and plastic limit ( $w_P$ ) were determined using percussion-cup and rolling test methods, respectively. Note that the bentonite, claystone and 144 145 their mixtures were wetted with synthetic water in the Atterberg limits test. For comparison, the 146 Atterberg limits of pure bentonite and claystone were also measured with de-ionized water.

#### 147 2.3 Hydraulic conductivity tests

148 The hydraulic conductivity tests were performed on cylindrical samples of 50 mm diameter and 10 mm height. The pure claystone and mixtures were poured into a stainless steel ring and statically 149 compacted at a controlled displacement rate of 0.05 mm/min to various dry densities. Subsequently, 150 151 the compacted samples were transferred into the testing cell of 50 mm diameter and confined by a 152 cap which was blocked using a screw to prevent axial swelling deformation during hydration (Fig. 153 3). Afterwards, synthetic water was supplied from the bottom of the cell and the swelling pressure during the hydration was monitored by the force transducer installed at the bottom of the cell. After 154 155 stabilization of the swelling pressure, the constant-head method was adopted to determine the 156 hydraulic conductivity of the compacted bentonite/claystone mixture in this study to gain good accuracy (Pedescoll et al. 2012; Sandoval et al. 2017). The constant water pressure applied at the 157 158 bottom of samples was 10-200 kPa (about 1/10 of the stabilized swelling pressure), defining a hydraulic gradient ranging from 100 to 2000. The evolution of injected water volume with time was 159 160 recorded. As the flow rate became steady, the tests were terminated and the corresponding hydraulic 161 conductivity k (m/s) was computed according to Darcy' law:

$$k = \frac{q}{iA} \tag{5}$$

where *q* is the steady flow rate (m<sup>3</sup>/s); *i* is the hydraulic gradient; *A* is the cross-section area of sample (m<sup>2</sup>). All the tests were carried out at an ambient temperature of  $20\pm1$  °C.

#### 165 2.4 Mercury intrusion porosimetry tests

162

After the hydraulic conductivity tests, the samples were extracted from the cell for the analysis of pore size distribution by MIP. Freeze-drying method was applied for the sample preparation: the saturated samples were carefully cut into small cubes with a side approximately equal to 1 cm; each 169 cube was rapidly immersed into liquid nitrogen previously vacuum-cooled below its freezing point 170 (-196 °C) and then dehydrated in a vacuumed chamber for about 24 hours (Wang et al. 2014). After 171 that, the samples were put into the low-pressure chamber of porosimeter with a working pressure 172 from 3.6 to 200 kPa, prior to being transferred to the high-pressure chamber with a working 173 pressure from 0.2 to 210 MPa (Bian et al. 2018). According to the Washburn equation, the pore 174 entrance diameter d (µm) intruded by mercury at an applied pressure p (MPa) could be calculated, 175 as follows:

$$d = \frac{4T_{\rm s} \cos \alpha}{p} \tag{6}$$

177 where  $T_s$  is the interfacial tension (taken as 0.485 N/m);  $\alpha$  is the contact angle between the 178 mercury-air interface and soil (taken as 130°). The identified pore entrance diameter ranged from 179 350 to 0.006 µm.

#### 180 **3 Experimental results**

#### 181 *3.1 Index properties*

The index properties of the bentonite, claystone and their mixtures are shown in Table 2. As the bentonite fraction increased from 0 to 100%, the liquid limit determined with synthetic water increased from 41 to 291% while the plastic limit increased from 23 to 53%. The variation of the corresponding plasticity index with bentonite fraction is illustrated in Fig. 4. It clearly showed that the plasticity index determined with synthetic water increased from 17 to 238% with the increase of bentonite fraction from 0 to 100%. From Fig. 4, it could also be observed that the plasticity index of the mixtures lied below the dashed line, which could be expressed by the following equation:

189 
$$I_{P-M} = I_{P-B} B / 100 + I_{P-C} (1 - B / 100)$$
 (7)

190 where  $I_{P-B}$  and  $I_{P-C}$  and are the plasticity indices of bentonite and claystone, respectively;  $I_{P-M}$  is the

191 mean weighted plasticity index of the bentonite and claystone in the mixture. This was consistent 192 with the observation of Pandian et al. (1995) on bentonite/clayey soil mixtures. It was suspected that the mixing of two clays with various particle sizes and non-uniform clay plate sizes influenced 193 194 development of the diffuse double layer (Schanz et al. 2013) and a smaller thickness of diffuse double layer surrounding the clay particles was expected in the bentonite/claystone mixture 195 196 (Sridharan et al. 1986). Accordingly, lower absorbed water on the external clay surface at the liquid 197 limit (Sridharan et al. 1986; Dolinar et al. 2007) and a lower plasticity index were observed 198 compared to the mean weighted value. With de-ionized water, the plasticity indices of bentonite and 199 claystone were found to be 488 and 17%, respectively. The plasticity index of bentonite with 200 synthetic water was significantly lower than that with de-ionized water. Yukselen-Aksoy et al. (2008) found that the saline water could considerably reduce the plasticity index of soils with a 201 202 plastic index larger than 70%. This water chemistry effect could be explained by the diffuse double layer theory. For the soils treated with saline water, the distance between two particles was 203 204 inversely with the square root of the salt concentration (Zhu et al., 2013). Thereby, a larger 205 inter-particle spacing and a larger amount of adsorbed water could be expected for the bentonite 206 with deionized water. As a result, a higher plastic index was observed compared to that with synthetic water. 207

208 3.2 Hydraulic conductivity

The hydraulic conductivities of the samples with different bentonite fractions and void ratios are summarized in Fig. 5. For comparison, the data of Karnland et al. (2008) on pure MX80 bentonite is also presented. For the samples with 70% bentonite, it decreased from  $1.00 \times 10^{-12}$  to  $9.91 \times 10^{-14}$  m/s as the void ratio decreased from 0.99 to 0.61; for the samples without bentonite, it decreased from

 $3.65 \times 10^{-10}$  to  $3.14 \times 10^{-11}$  m/s as the dry density decreased from 0.50 to 0.36. Overall, there was a 213 214 linear relationship between hydraulic conductivity and void ratio. Similar phenomenon was observed by Berilgen et al. (2006) and Dolinar (2009) when they studied the hydraulic conductivity 215 216 of crystallized kaolinite and Ca-montmorillonite. As the bentonite fraction decreased from 100% to 217 0%, the slope of the fitting lines (represented by the solid lines in Fig. 5) kept almost constant, with a mean value of 6.577, while the intercept increased from  $10^{-12.722}$  to  $10^{-7.401}$ . This phenomenon 218 219 indicated that the addition of claystone significantly reduced the sealing performance of the 220 mixture.

221 *3.3 Pore size distribution* 

222 Fig. 6 presents the pore size distribution curves of samples with various bentonite fractions and void ratios. On the whole, the cumulative curves of samples with larger void ratio lied above those of 223 224 samples with lower void ratio (Fig. 6a, c, e, g, i and k). Regarding the density function curves, the samples with large bentonite fractions and low void ratios showed a typical bimodal pore size 225 226 distribution, with small pores at 0.25 µm and large pores at 10-22 µm. For the samples with low 227 bentonite fractions and large void ratios, another pore population at 0.17-0.52 µm (designated as 228 medium pores) could be observed apart from the small-pore and large-pore populations (Fig. 6b, d, f, h, j and l). Following the definition of Bian et al. (2018) and Zeng et al. (2020), the pore sizes of 2 229 230 and 0.04 µm were adopted to delimit the large (2-350 µm), medium (0.04-2 µm) and small 231 (0.006-0.04 µm) pores. According to the accumulative curves, the corresponding void ratios of large, medium and small pores were determined. Their variations with the total void ratio are 232 233 illustrated in Fig. 7. It appeared that the large-pore and medium-pore void ratios decreased while the 234 small-pore void ratio increased with the decrease of total void ratio. For the samples at the same 235 total void ratio, as the bentonite fraction increased, the large-pore and small-pore void ratios 236 increased whereas the medium-pore void ratio decreased. Additionally, the void ratios for various populations became more uniform, showing a more complex pore size distribution. This 237 phenomenon could be explained as follows: as a rule, the samples compacted at dry state were 238 239 characterized by a bimodal porosity, with small and large pores (Wang et al. 2014; Bian et al. 2018). 240 When the clay particles were saturated with water, the water molecules would be successively 241 placed on the clay surface, increasing the inter-layer space (Wang et al. 2014; Bian et al. 2018). 242 During this process, some initial small pores would become medium pores. Therefore, a decrease of 243 the small-pore void ratio and an increase of the medium-pore void ratio could be expected after 244 saturation. In the meanwhile, the placement of water molecules on the clay surface could also lead to clay particle exfoliation, clogging the large pores (Wang et al. 2014). Note that this process was 245 246 strongly dependent on the dry density of soil. For the samples with low bentonite fraction, the void ratio of bentonite was relatively large and the placement of water molecules could be significant. 247 248 Thereby, greater large-pore and medium-pore void ratios and smaller small-pore void ratio could be 249 expected. On the contrary, with a high bentonite fraction, the void ratio of bentonite was relatively 250 low; the limited void space did not allow full placement of water molecules, thereby limiting the 251 microstructure change. For the samples with the same bentonite fraction, the decrease of total void 252 ratio in the as-compacted state reduced the inter-aggregate pore volume (defined as the pores larger than 2 um by Wang et al. (2013)), but did not change the intra-aggregate pore volume. This 253 explained why a greater large-pore (2-350 µm) void ratio was observed for the samples with a 254 255 greater total void ratio after saturation, despite the much significant clogging process of large pores, 256 thanks to the greater void ratio of bentonite.

#### **4 Influence of plasticity index on hydraulic conductivity based on the general capillary model**

From the general capillary model (Eq. (4)), the relationship between the hydraulic conductivity and the pore-size parameter (PSP) can be expressed by Eqs. (8) and (9) in the double-liner and double-logarithmic forms, respectively:

$$k = \frac{\rho g}{32\mu} \text{PSP} \tag{8}$$

$$\log(k) = \log(\frac{\rho g}{32\mu}) + \log(\text{PSP}) \tag{9}$$

263 where PSP is calculated from the pore size distribution using Eq. (10):

261

264 
$$PSP = n^{N_p} \sum_{i_1=1}^{m} \sum_{i_2=1}^{m} \cdots \sum_{i_{N_p}=1}^{m} \overline{d}^2 f(\mathbf{d}_{i_1}) f(\mathbf{d}_{i_2}) \cdots f(\mathbf{d}_{i_{N_p}})$$
(10)

265 The parameter  $N_p$  refers to the pore interconnection and reflects the pore tortuosity. To further analyse the influence of  $N_p$ , Eq. (10) with  $N_p=1, 2, 3, 4, 5, 6, 7, 8, 9$  and 10 was applied to compute 266 the PSP of MX80 bentonite/COx claystone mixtures, Louiseville clay (Lapierre et al. 1990), 267 268 Shanghai muddy clay (Tang and Yan 2015), Al-Ghat soil (Elkady et al. 2016), Wuhan clay (Zhao et al. 2016), Nanyang clay (Zhao et al. 2016), Maryland clay (Yuan et al. 2019) and Rajasthan 269 270 monovalent bentonite (Jadda and Bag 2020) using the respective pore size distributions. As an 271 example, the details of calculating the PSP value with  $N_p=4$  for the compacted MX80 272 bentonite/claystone mixture with a bentonite fraction of 30% and void ratio of 0.62 are described in Appendix A. In Fig. 8, the measured hydraulic conductivity is presented versus the PSP value for 273 274  $N_{p}=1, 2, 3, 4, 5, 6, 7, 8, 9$  and 10, together with estimated hydraulic conductivity using Eq. (8). For 275 clarity, all the data are presented in the double-logarithmic plane and the relationship between the estimated hydraulic conductivity and PSP value can be described by a straight line with a slope of 1. 276 For comparison, also plotted in Fig. 8 is the relationship between the measured hydraulic 277 conductivity and PSP determined by Watabe et al. (2006; 2011) for Québec glacial tills (Np=1, 2 278

279 and 3), sandy soils ( $N_p=2$ , 3, 4 and 5) and clayey soils ( $N_p=2$ , 3, 4 and 5). For all the samples, the 280 larger the value of  $N_p$ , the lower the PSP value and the lower the estimated hydraulic conductivity 281 using Eq. (8), suggesting that the samples with a more complex pore interconnection had a lower 282 hydraulic conductivity. When the  $N_p$  value was lower than 4, the measured hydraulic conductivities 283 of the MX80 bentonite/COx claystone mixtures appeared on the right-hand of the estimated line. It 284 appeared that the theoretical estimations with  $N_p=5$ , 6, 6, 7, 9 and 10 were the best fit for the 285 hydraulic conductivities of MX80 bentonite/COx claystone mixtures with 0%, 10%, 20%, 30%, 50% 286 and 70% bentonite, respectively. This phenomenon indicated that the pore structure was more 287 complex for the samples with a higher bentonite fraction, which was consistent with the MIP results. 288 For the Louiseville clay, Québec glacial tills, sandy soils, clayey soils, Shanghai muddy clay, Al-Ghat soil, Wuhan clay, Nanyang clay, Maryland clay and Rajasthan monovalent bentonite, the 289 290 theoretical equation showed the best performance for the hydraulic conductivity estimation when 291 the  $N_p$  was equal to 5, 3, 4, 5, 4, 5, 4, 6, 6 and 10, respectively.

292 As an easily determinable property, the plasticity index  $I_{\rm P}$  represents the water quantity 293 adsorbed to the internal and external clay surfaces (Dolinar 2009). Generally, the higher the clay 294 fraction (< 2  $\mu$ m), the larger the plasticity index (Seed 1964; Shi and Herle 2015). As illustrated in Fig. 1,  $N_p$  represents the number of in-series pores in various diameters for a given length. 295 296 Obviously, the larger the clay fraction, the smaller the grain size, the more complex the pore interconnection and the larger the  $N_p$  value. To quantitatively evaluate the influence of soil property 297 on the  $N_p$  value, the best  $N_p$  values are plotted versus the plasticity index in Fig. 9 for various soils. 298 299 Note that the plasticity indices of sandy and clayey soils reported by Watabe et al. (2011) ranged 300 from 0 to 15.3 and from 20.5 to 30.7, respectively. In Fig. 9, the mean plasticity indices of 7.6 and 301 25.4 were considered for the sandy and clayey soils, respectively. This relationship could be
302 expressed by the following exponential function, with a correlation coefficient of 0.940:

$$N_{\rm p} = 11.587 - 8.423 \exp^{-0.0096I_{\rm p}}$$
(11)

According to Eq. (11), the best  $N_p$  values of MX80 bentonite/COx claystone mixtures with 0%, 304 305 10%, 20%, 30%, 50% and 70% bentonite, Québec glacial tills, sandy soil, clayey soil, Louiseville 306 clay, Shanghai muddy clay, Al-Ghat soil, Wuhan clay, Nanyang clay, Maryland clay and Rajasthan monovalent bentonite were found to be 4.50, 5.22, 6.18, 7.25, 8.55, 9.90, 5.75, 3.16, 3.76, 4.99, 307 308 4.65, 4.97, 4.63, 5.11, 6.16 and 10.20, respectively. Note that the value of  $N_p$  applied to Eq. (10) 309 must be a natural number. In this study, two limit values of hydraulic conductivity were first 310 determined using Eq. (8) and (10) with two adjacent natural numbers of the estimated  $N_p$  and then the hydraulic conductivity of samples was estimated by a linear interpolation. The relationship 311 312 between the estimated and measured hydraulic conductivities for different soils is shown in Fig. 10. 313 The estimated hydraulic conductivities agreed well with the measured values, with errors in the 314 range from 1/5 to 5 times the measured values in most cases. This good agreement confirmed the 315 performance of the proposed method in the estimation of hydraulic conductivity.

#### 316 **5 Conclusions**

In this study, the hydraulic conductivity of bentonite/claystone mixtures with different bentonite fractions and void ratios were experimentally determined, together with the pore size distribution. The experimental results obtained and the data compiled from literature were interpreted using the general capillary model considering  $N_p$  pores in series, allowing the influence of plasticity index on the relationship between the hydraulic conductivity and the pore size distribution to be investigated.

322 The following conclusions can be drawn.

(1) There was a liner relationship between the logarithms of hydraulic conductivity and void ratio.
The slope of the fitting lines, kept almost constant for the samples with various bentonite fractions
whereas the intercept increased with the decreasing bentonite fraction, suggesting that the addition
of claystone reduced the sealing performance of the mixture.

327 (2) According to the microstructure investigation, the dry density of bentonite is lower for the 328 samples with lower bentonite fractions and larger void ratios and therefore, a larger medium-pore 329 (0.04-2  $\mu$ m) volume but a lower small-pore (0.006-0.04  $\mu$ m) volume was observed after saturation 330 due to the more significant swelling of bentonite.

(3) Based on the general capillary model, it was found that the higher the plasticity index, the larger the best value of  $N_p$ , indicating a more complex pore interconnection for the soils with higher plasticity. The relationship between  $N_p$  and plasticity index could be described by an exponential equation. Based on this equation, an improved general capillary model was proposed. The results estimated using this model were then compared with the measured ones and the good agreement of hydraulic conductivity between the measurement and estimation revealed the performance of the proposed model.

338

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343 Appendix A. The details of calculating the PSP value with  $N_p$ =4 for the bentonite/claystone 344 mixture sample with a bentonite fraction is 30% and void ratio of 0.62 345 According to Eq. (10), the pores should be divided into discrete intervals before the calculation of 346 PSP. Generally, the smaller the discretized intervals, the more precise the calculation of PSP. 347 However, more discretized intervals will increase the computation time. After performing several trial computations, the pores were divided into 33 groups in this study, as shown in Fig. 1A. 348 349 According to the cumulative pore volume curve of the bentonite/claystone mixture sample with a 350 bentonite fraction of 30% and void ratio of 0.62, the volumetric probability (f(d)) of each group 351 (corresponding to different average diameters of d) was calculated. The pore volumetric probability 352 distribution of the sample is summarized in Fig. 1A. Afterwards,  $N_p=4$ , m=33, n=0.62/(1+0.62)=0.38 and the pore volumetric probability distribution were substituted into Eq. 353 (10). Fig. 2A is a flowchart showing the process of calculating the PSP value with  $N_p=4$ . The 354 corresponding PSP value of the sample was calculated to be 0.0225  $\mu m^2$ . 355

356

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#### 466 List of Tables

- 467 Table 1 Chemical composition of the synthetic water
- 468 Table 2 Index properties of MX80 bentonite, COx claystone and their mixtures

#### 469 List of Figures

- 470 Fig. 1 Typical water flow paths in soils consisting of (a) two sections and (b) six sections (modified
- 471 after Watabe (2006)). Note: the grey areas represent the solid particle and the arrows indicate the
- 472 water flow paths
- 473 Fig. 2 Grain size distributions of MX80 bentonite and crushed COx claystone
- 474 Fig. 3 Layout of the experimental setups for hydraulic conductivity tests
- 475 Fig. 4 Variation of plasticity index determined using synthetic water with bentonite fraction
- 476 Fig. 5 Relationship between hydraulic conductivity and void ratio
- 477 Fig. 6 Pore size distributions of samples: (a) cumulative curves of samples with 0% bentonite, (b)
- 478 density function curves of samples with 0% bentonite, (c) cumulative curves of samples with 10%
- 479 bentonite, (d) density function curves of samples with 10% bentonite, (e) cumulative curves of
- 480 samples with 20% bentonite, (f) density function curves of samples with 20% bentonite, (g)
- 481 cumulative curves of samples with 30% bentonite, (h) density function curves of samples with 30%
- 482 bentonite, (i) cumulative curves of samples with 50% bentonite, (j) density function curves of
- 483 samples with 50% bentonite, (k) cumulative curves of samples with 70% bentonite and (l) density
- 484 function curves of samples with 70% bentonite
- 485 Fig. 7 Variations of (a) large-pore, (b) medium-pore and (c) small-pore void ratios with respect to
- 486 total void ratio
- 487 Fig. 8 Relationship between hydraulic conductivity and PSP
- 488 Fig. 9 Variation of  $N_P$  with plasticity index  $I_p$
- 489 Fig. 10 Comparison between measured and estimated hydraulic conductivities using Eqs. (8), (10)490 and (11)
- 491 Fig. A1 Pore volumetric probability versus pore diameter for the sample with a bentonite fraction of
- 492 30% and void ratio of 0.62. The number above each bar indicates the pore volumetric probability in493 percent
- 494 Fig. A2 The flow chart of calculating the PSP value using Eq. (10) with  $N_p=4$
- 495

496 Table 1 Chemical composition of the synthetic water

Component	NaCl	NaHCO <sub>3</sub>	KCl	CaSO <sub>4</sub> •2H <sub>2</sub> O	MgSO <sub>4</sub> •7H <sub>2</sub> 0	CaCl <sub>2</sub> •2H <sub>2</sub> O	Na <sub>2</sub> SO <sub>4</sub>
Content (g/L)	1.950	0.130	0.035	0.630	1.020	0.080	0.700

Soil monorty	Claystone	Bentonite/cl	- Dontonito				
Son property		<i>B</i> =10%	<i>B</i> =20%	<i>B</i> =30%	<i>B</i> =50%	<i>B</i> =70%	- Bentonne
Water content (%)	6.1	6.6	7.2	7.7	8.8	9.8	11.4
Specific gravity	2.70	2.71	2.71	2.72	2.73	2.74	2.76
Liquid limit (%)	40	54	73	98	140	208	291
Plastic limit (%)	23	25	27	29	34	41	53
Plasticity index (%)	17	29	46	69	106	167	238

**Table 2** Index properties of MX80 bentonite, COx claystone and their mixtures



Fig. 1 Typical water flow paths in soils consisting of (a) two sections and (b) six sections (modified after Watabe (2006)). Note: the
 grey areas represent the solid particle and the arrows indicate the water flow paths



Fig. 2 Grain size distributions of MX80 bentonite and crushed COx claystone





Fig. 3 Layout of the experimental setups for hydraulic conductivity tests



Fig. 4 Variation of plasticity index determined using synthetic water with bentonite fraction



Fig. 5 Relationship between hydraulic conductivity and void ratio







516 Entrance pore diameter D (μm)
517 Fig. 6 Pore size distributions of samples: (a) cumulative curves of samples with 0% bentonite, (b) density function curves of samples
518 with 0% bentonite, (c) cumulative curves of samples with 10% bentonite, (d) density function curves of samples with 10% bentonite,
519 (e) cumulative curves of samples with 20% bentonite, (f) density function curves of samples with 20% bentonite, (g) cumulative
520 curves of samples with 30% bentonite, (h) density function curves of samples with 30% bentonite, (i) cumulative curves of samples
521 with 50% bentonite, (j) density function curves of samples with 50% bentonite, (k) cumulative curves of samples with 70% bentonite
522 and (l) density function curves of samples with 70% bentonite



Fig. 7 Variations of (a) large-pore, (b) medium-pore and (c) small-pore void ratios with respect to total void ratio



Fig. 8 Relationship between hydraulic conductivity and PSP



**Fig. 9** Variation of  $N_p$  with plasticity index  $I_p$ 



Fig. 10 Comparison between measured and estimated hydraulic conductivities using Eqs. (8), (10) and (11)



532 Pore diameter (μm)
 533 Fig. A1 Pore volumetric probability versus pore diameter for the sample with a bentonite fraction of 30% and void ratio of 0.62. The
 534 number above each bar indicates the pore volumetric probability in percent







Fig. A2 The flowchart of calculating the PSP value using Eq. (10) with  $N_p=4$