Anchoring Multi-Scale Models to Micron-Scale Imaging of Multiphase Flow in Rocks

Shan Wang¹, Leonardo C. Ruspini², Paal-Eric Oeren², Stefanie Van Offenwert¹, and Tom Bultreys¹

¹Ghent University ²Petricore Norway AS

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Abstract

Image-based pore-scale modeling is an important method to study multiphase flow in permeable rocks. However, in many rocks, the pore size distribution is so wide that it cannot be resolved in a single pore-space image, typically acquired using micro-computed tomography (micro-CT). Recent multi-scale models therefore incorporate sub-voxel porosity maps, created by differential micro-CT imaging of a contrast fluid in the pores. These maps delineate different microporous flow zones in the model, which must be assigned petrophysical properties as input. The uncertainty on the pore scale physics in these models is therefore heightened by uncertainties on the representation of unresolved pores, also called sub-rock typing. Here, we address this by validating a multi-scale pore network model using a drainage experiment imaged with differential micro-CT on an Estaillades limestone sample. We find that porosity map-based sub-rock typing was unable to match the micrometer-scale experimental fluid distributions. To investigate why, we introduce a novel baseline sub-rock typing method, based on a 3D map of the experimental capillary pressure function. By incorporating this data, we successfully remove most of the sub-rock typing uncertainty from the model, obtaining a close fit to the experimental fluid distributions. Comparison between the two methods shows that in this sample, the porosity map is poorly correlated to the multiphase flow behavior of the microporosity. The validation method introduced in this paper serves to separate and address the uncertainties in multi-scale models, facilitating simulations in complex geological reservoir rocks important for e.g. geological storage of CO2 and renewable energy.

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3	Shan Wang ^{1,2} , Leonardo C. Ruspini ³ , Pål-Eric Øren ³ , Stefanie Van Offenwert ^{1,2} and Tom					
4	Bultreys ^{1,2}					
5	¹ Department of Geology, Pore-Scale Processes in Geomaterials Research (PProGRess), Ghent					
6	University, Krijgslaan 281/S8, B-9000, Ghent, Belgium					
7	² Centre for X-ray Tomography (UGCT), Ghent University, Proeftuinstraat 86, B-9000, Ghent,					
8	Belgium					
9	³ Petricore Norway A.S., Trondheim, Norway					
10	Corresponding author: Shan Wang (Shan.Wang@UGent.be)					
11	Key Points:					
12	• Improved differential imaging technique is used to quantify porosity and saturation					
13	distribution of a heterogeneous rock sample					
14	• A new sub-rock typing method based on the drainage experiment is presented					
15	• The breakthrough pressure plays an important role in multiphase distribution within					
16	heterogeneous rocks during capillary drainage					
17						

18 Abstract

19 Image-based pore-scale modeling is an important method to study multiphase flow in permeable 20 rocks. However, in many rocks, the pore size distribution is so wide that it cannot be resolved in a 21 single pore-space image, typically acquired using micro-computed tomography (micro-CT). 22 Recent multi-scale models therefore incorporate sub-voxel porosity maps, created by differential 23 micro-CT imaging of a contrast fluid in the pores. These maps delineate different microporous 24 flow zones in the model, which must be assigned petrophysical properties as input. The uncertainty 25 on the pore scale physics in these models is therefore heightened by uncertainties on the 26 representation of unresolved pores, also called sub-rock typing. Here, we address this by validating 27 a multi-scale pore network model using a drainage experiment imaged with differential micro-CT 28 on an Estaillades limestone sample. We find that porosity map-based sub-rock typing was unable 29 to match the micrometer-scale experimental fluid distributions. To investigate why, we introduce 30 a novel baseline sub-rock typing method, based on a 3D map of the experimental capillary pressure 31 function. By incorporating this data, we successfully remove most of the sub-rock typing 32 uncertainty from the model, obtaining a close fit to the experimental fluid distributions. 33 Comparison between the two methods shows that in this sample, the porosity map is poorly 34 correlated to the multiphase flow behavior of the microporosity. The validation method introduced 35 in this paper serves to separate and address the uncertainties in multi-scale models, facilitating 36 simulations in complex geological reservoir rocks important for e.g. geological storage of CO₂ and 37 renewable energy.

38

39 Plain Language Summary

40 Understanding multiphase flow within heterogeneous reservoir rocks is crucial for geological 41 reservoir management. These rocks usually have intricate microstructures (unresolved or sub-42 resolution pores) which are difficult to quantify and have a strong impact on fluid flow. Pore-scale 43 modelling combined with imaged-based experiments can be a useful tool to describe complex pore 44 structures, which is of key importance in the subsequent simulation and prediction of multiphase 45 flow behavior. In this study, we focus on improving the representation of unresolved porous 46 regions of a heterogeneous rock sample (also called sub-rock typing). A drainage experiment was 47 performed and imaged by micro-computed tomography (micro-CT) to characterize the multiphase

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48 distribution at increasing capillary pressures. The predictions of two multi-scale models, which

- 49 were generated according to distinct sub-rock typing methods on the same sample, were compared
- 50 with the drainage experimental data. We found that the model obtained by the "classical" sub-rock
- 51 typing method was unable to simulate the correct arrangement of fluids in this sample, while the
- 52 new method performed better, which illustrates the importance of rock type identification to pore-
- 53 scale modelling. The validation workflow presented in this paper can be extended and served as a
- 54 reliable reference to improve simulations in other complex geological materials.

55 **1 Introduction**

56 Multiphase flow through rocks plays an important role in numerous earth science applications, 57 such as hydrocarbon recovery (Olaviwola & Dejam, 2019; Wang et al., 2020), carbon dioxide 58 storage (Arif et al., 2017), remediation of polluted aquifers (Bortone et al., 2013) and subsurface 59 energy storage in the form of hydrogen or compressed air (Amid et al., 2016, Mouli-Castillo et al., 2019). Many reservoir rocks, notably carbonates and clay-bearing sandstones, exhibit complex 60 61 pore geometries with very wide pore size distributions. The petrophysical properties of such rocks 62 often do not obey classical correlations (Prodanović et al., 2015; Shanley et al., 2004), spurring 63 pore-scale studies of their fluid flow behavior (Mehmani et al., 2020). This can be done based on 64 images of the pore space, obtained with for example micro-computed tomography (micro-CT) and 65 (FIB-)SEM imaging (Bultreys et al., 2016b; Bera et al., 2011; Ciobanu et al., 2011; Cnudde & 66 Boone, 2013; Wirth, 2009).

67 Despite the wide interest in simulating fluid flow in complex, multi-scale pore spaces, the trade-68 off between image size and resolution in most imaging techniques complicates the development 69 of suitable image-based approaches (Blunt et al., 2013). Typical imaging workflows identify 70 resolved pores and zones with unresolved porosity (below the µm scale) in micro-CT images of 71 mm-scale samples, which usually capture the largest pore features. Unresolved porosity is visible 72 in these images as zones with grey values that are intermediate between solid and void, due to their 73 intermediate density (Cnudde & Boone, 2013). These regions can then be imaged by higher-74 resolution techniques, that are then correlated back to the lower-resolution micro-CT scan (De 75 Boever et al., 2015; Devarapalli et al., 2017; Lin et al., 2019). The unresolved pores, which we 76 will by definition refer to as the microporosity, can play a crucial role in the sample's multiphase 77 flow behavior (Bultreys et al., 2016c; Mehmani et al., 2020). Therefore, specialized multi-scale 78 models are required that fuse the resolved pores with information on the microporosity features, 79 typically obtained using higher-resolution images on small sub-sections of the sample (Menke et 80 al., 2019).

Multi-scale models are an extension of pore-scale, image-based modelling techniques, consisting of either direct numerical simulations (Alhashmi et al., 2015; Pan et al., 2004; Raeini et al., 2012) or pore network models (PNMs) (Blunt et al., 2013; Dong & Blunt, 2009). Direct numerical simulations of multiphase flow usually require large computational resources and therefore

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85 struggle to capture capillary-dominated flow on large images (Blunt et al., 2013). PNMs feature 86 higher computational efficiencies due to simplifications on the geometry and the fluid 87 displacement physics, which makes them well-suited for multi-scale simulations. Networks 88 obtained at different resolutions or scales can be extrapolated in space and fused to reconstruct a 89 multi-scale pore network (Jiang et al., 2013; Mehmani & Prodanovic, 2014), resulting in detailed 90 but very large networks. Alternatively, the micropores can be treated as a continuous porous 91 medium with specific petrophysical properties to reduce the size of the network while still representing the connectivity caused by microporosity (Bultreys et al., 2015; Bauer et al., 2012; 92 93 Youssef et al., 2008).

94 Despite the progress in representing multi-scale pore networks, it remains difficult to assess the 95 model uncertainties. Multi-scale pore network models depend on a significant amount of uncertain 96 input information to describe the microporosity behavior, which complicates the validation of the 97 physical assumptions in the model itself (e.g. quasi-static fluid displacement). An important aspect 98 of the input uncertainty is that only a limited volume of microporosity is typically imaged at the 99 highest resolutions, while there is often significant heterogeneity in its properties. Recent 100 approaches have addressed this information gap by incorporating sub-voxel porosity maps 101 (Ruspini et al., 2016, 2021). The porosity map can be generated based on differential imaging: 102 measuring the calibrated grey value change when the pore space is filled with a high-contrast fluid 103 such as high-concentration potassium iodide or cesium chloride (Boone et al., 2014; Ghous et al., 104 2007, Lin et al., 2016). The resulting map is then used for "sub-rock typing": identifying and 105 characterizing zones with different microporosity properties in the model. The common approach 106 is to perform a segmentation on dry images or differential images directly to separate distinct 107 phases, sometimes followed by a series of image processing operations to alleviate artifacts at 108 phase boundaries (Bauer et al., 2012; Bultreys et al., 2015). However, there is still a lack of 109 validation to reveal whether these approaches can provide a reasonable representation for 110 unresolved pores and how much uncertainty they introduce into the multi-scale PNM simulations. 111 Recent work on single-scale pore network models has illustrated how pore-by-pore validation of 112 the fluid distributions during drainage or imbibition can serve to study model uncertainties 113 (Bultreys et al., 2018, 2020, Øren et al., 2019). A similar principle used to generate porosity maps 114 can be used to track the fluid saturation in microporous regions during multiphase flow 115 experiments (Lin et al., 2016, Gao et al., 2017). In this paper, we propose a pore-by-pore validation

116 workflow for multi-scale PNM by comparing fluid distributions in the model to a differential 117 micro-computed tomography (micro-CT) based drainage experiment on a heterogeneous 118 Estaillades limestone (as shown in Figure 1). Using this data, we validate the porosity-map based 119 workflow, and explain its uncertainties by comparing to a novel data-based model, which takes its 120 microporosity information from the micro-CT drainage experiment. The experiment and the image 121 processing workflows are introduced in Section 2.1. Then, the multiscale PNM workflow 122 including the two sub-rock typing methodologies (porosity-based and drainage-based methods) 123 are explained in Section 2.2. In Section 3, we compare the predictions from the two models with 124 experimental data and discuss the reasons that may lead to simulation uncertainties. Section 4 125 discusses the conclusion and the outlook for our future research.

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Figure 1. The validation workflow for multi-scale pore network model.

- 129
- 130 2 Materials and Methods
- 131 2.1 Experiment

132 The main goal of the experiment presented here is to measure the brine and decane distribution

133 within both macropores and microporous regions of the sample during capillary drainage. In the

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following, the sample and fluid preparation, the set-up, the experimental procedure and dataprocessing workflow will be discussed in detail.

136 2.1.1. Rock samples and fluid preparation

137 The rock sample used in the experiment is Estaillades limestone, which has a complex pore 138 structure featuring a bi-modal pore size distribution. The broad pore size distribution has two peaks 139 with modes of respectively 390 nm and 19 μ m (Han et al., 2007). Estaillades limestone is 140 composed of 99% calcite (Alyafei & Blunt, 2016). A sample with a diameter of 6 mm and a length 141 of 20 mm was cored and vacuum-saturated with deionized water to ensure that all the air was 142 removed from the pores before the experiment.

As fluid phases in the drainage experiment, we used KI-brine and decane. The former acted as wetting phase, while the latter was the non-wetting phase. The brine was made from deionized water doped with 25 wt% potassium iodine (KI) as a contrast agent due to its high X-ray attenuation coefficient. This solution provided a strong contrast to identify the fluid phases in the micro-CT images.

148 2.1.2. Drainage experiment

149 The experimental apparatus and flow lines are shown in Figure 2. The rock sample was placed on 150 top of a water-wet ceramic porous plate (Cobra Technologies B.V., NL) and then wrapped in a 151 Viton sleeve. The hydrophilic porous plate had a breakthrough pressure of 1300 kPa to prevent 152 early breakthrough of the non-wetting phase. This assembly was placed in an X-ray transparent 153 flow cell made out of PEEK (RS Systems, Norway), connected to high-precision syringe pumps 154 supplying the experimental fluids. A differential pressure transducer (Keller PD-33X) was 155 connected to the inlet and outlet of the sample. The flow cell was then placed on the Environmental 156 Micro-CT (EMCT) scanner at Ghent University's Centre for X-ray Tomography (UGCT) (Dierick 157 et al., 2014; Bultreys et al., 2016a). This scanner consists of a rotating source-and-detector gantry, 158 meaning the flow cell remained static during the full experiment. Throughout the experiment, the 159 X-ray beam was filtered with 1 mm aluminium to reduce the beam hardening effect. The 160 experiment was performed by executing the following steps:

- A confining pressure of 3500 kPa was set to compress the Viton sleeve, to avoid fluid
 bypassing along the wall of sample.
- 164
 2. The water-saturated sample was scanned by micro-CT at room temperature and pressure.
 165 The imaging settings were: 6.5 μm voxel size, 2400 projections, 1150 ms integration time
 166 per radiograph; 110 kV and 8 W X-ray tube settings. Since water has a low grey value
 167 similar to air in the images, we will refer to this as the "dry scan" or "dry image" in
 168 following sections for convenience.
- 3. The water was flushed out by brine, which was injected through the sample with a maximum flow rate of 0.075 ml/min for 4 hours and left overnight. A high-quality microCT scan was conducted to capture the sample's 100% brine saturated state (same imaging settings as step 2).
- 173 4. The drainage was started by injecting decane from the top of the flow cell at a low flow 174 rate (0.001 ml/min). To set a constant pressure drop over the sample, which at vanishing 175 flow rates yields a set capillary pressure in the sample, the flow rate was subsequently 176 gradually lowered based on manual inspection of the pressure transducer reading. This 177 proved to be more reliable than using automated constant-pressure settings on the syringe 178 pump, particularly at low set pressures. The experiment remained capillary dominated at 179 all times during this equilibration procedure, as the maximum capillary number was $6.2 \times$ 180 10⁻⁸.
- 5. Radiographs of the sample were collected and subtracted from each other to track the saturation change in the sample during the equilibration (supporting information Figure S1). In addition, a short micro-CT scan (6.5 µm voxel size, 2400 projections, 115 ms exposure time) was performed every hour to further compare flow distribution changes.
 When no more changes were found from both the differential radiographs and the subsequent micro-CT images at the target pressure, a high-quality scan (imaging settings see step 2) was taken.
- 6. Steps 4 and 5 were repeated with gradually increasing pressure. We performed 6 capillary
 pressure steps during the experiment, at 8, 14, 80, 180, 220 and 400 kPa. Equilibration took
 between 4 and 7 hours for the different pressure steps, in all cases having reached very low
 final flow rates (below 0.0003 ml/min).



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- 194

Figure 2. Experimental apparatus used in the drainage experiment.

196 2.1.3. Micro-CT image processing

197 The acquired tomograms, which represent the 3D distribution of X-ray attenuation coefficients in 198 the sample, were reconstructed using Octopus Reconstruction software (Tescan-XRE, Belgium). 199 After reconstruction, the image processing was performed using Avizo 2020.2 (ThermoFisher, 200 France). The brine-saturated and drainage-step images were all registered to the dry image using 201 normalized mutual information and resampled using the Lanczos algorithm to make sure all the 202 images are aligned in space. The images were then filtered with a non-local means edge-preserving 203 filter to reduce the image noise. As shown in Figure 3, the pores in the dry scan image are dark 204 grey and have significantly lower grey values than solid calcite grains. In the brine-saturated image, 205 the brine-invaded pores are brighter than other phases in the sample. A cross-section of the fluid 206 distribution in the 6 capillary pressure steps is shown in Figure 3(c). With the increase of drainage 207 pressure, the brine is displaced by decane, and brighter pores gradually become "black".



(a)











208

Although the KI-brine visually already indicates the presence of different fluid phases within macropores, further processing is necessary to quantify the brine saturation in the unresolved porosity. We propose an improved workflow here based on differential imaging with normalized water-filled and brine-filled images to calculate the sub-resolution saturation in voxels that contain microporosity.

The image normalization is based on selecting fixed grey values for two known materials, setting them to the same value in the different images, and doing a linear rescaling of everything in between. To normalize the brine-saturated image to the dry image, we first cropped 3 region-ofinterests (ROI) of solid grain to determine the upper normalization value, and an ROI in the sleeve (which has a low grey value) for the lower value. The rescaled image was calculated following equation (1), which is adapted from Lin et al, (2017):

$$I_{new} = (I - p_{s_bine}) \cdot \frac{p_{g_dry} - p_{s_dry}}{p_{g_brine} - p_{s_brine}} + p_{s_dry}$$
(1)

Where I_{new} is the rescaled image, I is the image before normalization, p_{g_dry} is the average mode of the 3 solid grain ROIs from the dry image, p_{s_dry} is the mode of the grey value histogram of the sleeve ROI from the dry image, p_{g_brine} is the average mode of the 3 solid grain ROIs from the brine saturated image, and p_{s_brine} is the mode of the sleeve ROI from the brine saturated image. For the drainage images, we first cropped 3 ROIs in solid grains and 3 ROIs in water-filled resolved pores from the dry and the 6 drainage images, extracted their grey value histograms and calculated their modes. The drainage images were then rescaled according to equation (2):

229
$$I_{new} = (I - p_{d_drain}) \cdot \frac{p_{g_dry} - p_{w_dry}}{p_{g_drain} - p_{d_drain}} + p_{w_dry}$$
(2)

Where p_{g_dry} is the average mode of 3 solid grain ROIs in the dry image, p_{w_dry} is the average mode of 3 water-invaded-pore ROIs from the dry image, p_{g_drain} is the average mode of 3 solid grain ROIs from the drainage images, p_{d_drain} is the average mode of 3 decane-invaded-pore ROIs from drainage images.

Then, the differential image was obtained by calculating the differences between the rescaled drainage- or brine saturated images and the water-filled image:

$$I_{diff_drain} = I_{new_drain} - I_{dry}$$
(3)

$$I_{diff_brine} = I_{new_brine} - I_{dry}$$
(4)

238 The dry-brine differential image was used to quantify sub-resolution porosity in the image, due to 239 the assumed linear dependence of the grey value in the differential image and the volume 240 percentage of brine present in each voxel. We denote the thresholds set for solid (0% porous) and 241 open porosity (100% porous) voxels as CT1 and CT2, respectively. CT1 was determined based on 242 the valley between the grey value distributions of the solid and microporous phase in the histogram. 243 CT2 was found by masking the differential image with the macropores segmented from the dry 244 image, and finding the peak of the associated grey value histogram (supporting information Figure 245 S2). Voxels with grey values equal to or less than CT1 are assigned 0% porosity, those equal to or 246 higher than CT2 are assigned 100% porosity, the microporous region between CT1 and CT2 is 247 assigned 0~100% porosity, using:

- 248 $\varphi_{micro} = \frac{I_{diff_brine} CT1}{CT2 CT1}$
- 249 Where φ_{micro} is the porosity within microporous voxels. The total porosity can be calculated by:
- 250
- 251

$$\varphi_{total} = \frac{V_{macro} + V_{micro} * \varphi_{micro}}{V_{sample}} \tag{6}$$

(5)

252 Where V_{macro} is the number of macropore voxels, V_{micro} is the number of micro-pore voxels, V_{sample} 253 is the number of voxels in the whole sample.

Simarly to the porosity map, we also calculated the brine saturation map at each capillary pressurestep using the equation (7):

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$$S_{w_drain} = \frac{\varphi_{drain}}{\varphi_{micro}} = \frac{I_{diff_drain} - CT1}{I_{diff_brine} - CT1}$$
(7)

With this method, the porosity distribution map was obtained, shown in Figure 4(b), which provided a 3D distribution of the porosity variation within the microporous phase. Table 1 shows the porosity calculation results. The total porosity of this sample was 25.43%, of which the subresolution porous regions contributed 78.53%. The 3 components, macropores, microporous regions and solid grains, were extracted from this porosity map (Figure 4(c) and supporting information Figure S3).

263



264 265

(a)

(c)

Figure 4. Porosity distribution map. (a) A slice from the filtered dry image (b) The corresponding porosity
distribution map at the same slice (c) Three-phase segmentation of the sample into macropores (red),
microporous regions (yellow) and solid (blue).

(b)

269

270

Table 1. Porosity calculation

	Phase	Threshold	Voxel	Voxel*Porosity	Average	Volume	Contribution	Total
			(×10 ⁸)	(×10 ⁸)	porosity	fraction		porosity
	macro	>1	0.42576		1	0.05463	0.05463	
	micro	0-1	5.01773	1.55637	0.31017	0.64381	0.19969	0.2543
	grain	<0	2.35033		0	0.30156	0	
$271^{$								

272 2.2 Multi-scale model

273 2.2.1 Sub-rock typing

274 Multi-scale models of multiphase flow depend on the classification of regions with sub-resolution 275 porosity. To identify these regions, and inform their description in the model, a recently proposed 276 method is to segment a sub-resolution porosity map of the sample into different "sub-rock types". 277 We will refer to this as the "porosity-based" method. A crucial first step is to generate an accurate 278 porosity map (Figure 4(b)). Figure S4 in supporting information depicts the histogram of the 279 porosity distribution in the sample. Two thresholds (0.2 and 0.4) were manually selected to divide 280 the sample into 3 microphase regions or 3 rock types (3RT), based on trial-and-error to obtain 281 realistic simulation results for the capillary pressure curve (see further). The sub-rock typing result 282 is shown in Figure 6.

283 The downside of porosity-based sub-rock typing is that it is based on the assumption that porosity 284 is closely correlated to the multiphase flow properties of the microporosity. This is not necessarily 285 the case. Therefore, we introduce a new method here to perform rock typing based directly on 286 experimental multiphase flow data. To this end, we determined the invasion capillary pressure (Pct) 287 distribution within the microporous phase. First, the saturation map at each drainage pressure step 288 was calculated (Section 2.1.3). The saturation variation with the increase of pressure of every 289 individual voxel was thus obtained. Next, these saturation maps were mean-filtered and 290 downsampled by a factor 2 to reduce the noise dependency and computational load. Then, a 291 Brooks-Corey-type P_c formulation (Brooks & Corey, 1964) was fitted to the capillary pressure-292 saturation data points of each voxel using a least-squares approach in Matlab. This yielded a 293 relation of the following form for each voxel:

294

$$P_c = P_{ct} \left(\frac{1}{S_w}\right)^{\frac{1}{\lambda}} \tag{8}$$

Where P_c is the capillary pressure, P_{ct} is the fitted invasion capillary pressure, λ is a fitted parameter related to the pore size distribution and S_w is water saturation.

In this way, P_{ct} and λ values were derived for all microporous voxels. The fitted curves for two representative voxels are shown in Figure 5(a). Figure 5(c) presents the Pct map and the λ map, showing the 3D variation of the capillary pressure behavior in the sample. We primarily used the Pct map here due to its easier interpretability and lower noisiness than the λ -map. We considered 301 both porosity and Pct together by performing a k-means clustering on the (ϕ , Pct) points of all the

302 voxels (Figure 5(b)). This was used to divide the voxels into 5 clusters, representing 5 microporous

303 subrock types, next to the microporous and solid voxels in the image.





310

311 Figure 5. Pct-based sub-rock typing workflow. (a) The diagram of data fitting of two representative voxels. (b)

- 312 A plot of Pct versus porosity. (c) Porosity map (left), Pct distribution map (middle) and λ distribution map (right).
- 313

314 2.2.2 Pore network extraction and simulations

315 Two multi-scale PNMs of the sample were extracted using the porosity map and the sub-rock type 316 maps as input. These PNMs consist of four types of network elements: resolved nodes ("pores") 317 and links ("throats") that represent the macroporosity and unresolved ("Darcy") nodes and links 318 that represent the microporosity. The extraction is based on skeletonization and maximal ball 319 clustering of the resolved pore space and each of the microporosity sub-rock types in order to find 320 the centers of nodes and links, as described in Øren et al. (2019). These are then connected together 321 to honour the connectivity of the multi-scale pore-space. Geometrical properties (e.g. inscribed 322 radius, volume, shape factor) of the network elements are subsequently determined. For Darcy 323 nodes and links, the geometrical properties are supplemented by the local porosity determined 324 from the porosity map, and local petrophysical properties based on user input for each sub-rock 325 type (see Section 2.2.3). Full details on the method can be found in Ruspini et al. (2021). We 326 extracted two different PNMs of the same sample, based on respectively the porosity-based sub-327 rock typing map and the Pct-based sub-rock typing map (Figure 6). The properties of these PNMs 328 are shown in Table 2. The total porosity was 26.2% with 5.6% contributed by macropores and 329 20.6% by micropores, which honours the micro-CT results (Table 1).



Figure 6. Dry image (left), porosity-based sub-rock typing result (middle) and Pct-based sub-rock typing result (right). The red and black in both sub-rock typing maps refer to macropores and solid grain. In porosity-based

map: RT1, RT2 and RT3 show high, middle and low microporosity regions respectively. In Pct-based map: RT1

to RT5 represent lower to higher Pct microporous regions.

335

336 After extraction, the resulting PNMs were used to predict permeability and pore-scale fluid 337 distributions during drainage. Permeabilities were calculated by assigning a conductivity value to 338 each element (based on Poisseuille flow in resolved network elements and Darcy's law in Darcy 339 elements, taking the local microporosity's permeability) and solving a set of mass balance 340 equations (Ruspini et al., 2021). Drainage was simulated quasi-statically by calculating an 341 intrusion capillary pressure for each network element, and then performing an invasion-percolation 342 (i.e. progressively invading accessible network elements in ascending order of their intrusion 343 capillary pressure). Invasion capillary pressures for resolved pores and throats were calculated by 344 assuming triangular, square or cylindrical cross-sectional shapes (Mason & Morrow, 1991). In 345 Darcy elements, the invasion pressure was found from input capillary pressure curves of the sub-346 rock type (scaled according to the local porosity and permeability using the Leverett-J-curve). The 347 simulation took fluid connectivity through wetting layers into account, and assumed a fluid to be 348 connected through a Darcy element as long as the local relative permeability to the fluid was larger 349 than zero. Full details are provided in Ruspini et al. (2021).

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- 352

Table 2. Properties of extracted PNMs

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Properties	Porosity-based model	Pct-based model
Nodes	666543	658120
Links	4334582	5006058
Darcy pore	4898310	5561272
Total porosity	0.2625	0.2624
Resolved porosity	0.0564	0.0563
Unresolved porosity	0.2061	0.2061

353

354 2.2.3 Petrophysical properties

355 The petrophysical properties taken into account to represent unresolved porosity in each Darcy 356 node of the PNM are porosity, permeability, relative permeability and capillary pressure curve (P_c-357 curve). The local porosity in each element was determined from the porosity map. Compared with 358 the traditional way of assigning an average porosity value to the whole microporosity phase, the 359 introduction of the porosity map captures more realistic porosity heterogeneity. The permeability for each node was calculated from a power correlation $k = a * \varphi^b$, where a and b are supplied as 360 361 input values for each sub-rock type. Parameter b was set to 3.37 for all types based on a nm-scale 362 imaging study performed by Menke et al. (2019) on microporosity in Estaillades. Since the 363 microporosity's permeability values affect the intrusion capillary pressure curve of Darcy nodes 364 through Leverett-J scaling, parameter "a" was tuned to match the output P_c -curve of the PNM 365 simulations with our experimental P_c -curve measurement (Section 2.1). Note that anchoring 366 simulations to available experimental data is common practice for multi-scale models, in order to 367 allow better predictions of more difficult-to-obtain properties.

The determination of the relative permeability curve for each rock type is a challenging task, as this property is difficult to measure directly. Here, the Brooks-Corey model (Brooks & Corey, 1966) was used to assign the same relative permeability curves to all Darcy elements, in order to decrease its uncertain influence on the drainage simulation results. Approaches to obtain more accurate relative permeability curves for different micro-regions are the subject of future investigations.

For the porosity-based PNM, the Brooks-Corey-type P_c formulation (Brooks & Corey, 1964) was fitted to the experimental Pc-measurement to obtain P_c -curves for each microporosity type. The fitted curve was split into three parts, corresponding to the three microporosity regions from the porosity map (see Section 2.2.1). The saturation range of each of these parts was then rescaled to 378 a range between 0 and 1, to serve as input P_c -curves for the sub-rock types, where the curve with 379 the lowest Pc-range was assigned to the rock type with the highest porosity. Note that this 380 workflow represents a coarse fit of the input properties to the experimental Pc-data (see e.g. 381 Bultreys et al. 2015), but that this may not generally yield good results for all samples, and that 382 there is no set way to divide the experimental Pc-curve into the individual sections (in this work, 383 this was done by trial-and-error, until a satisfactory fit was found between the simulated and the 384 experimental Pc-curves). For the Pct-based PNM, the input P_c -curves of each rock type were 385 calculated directly from the imaging data, that is, the average saturation within each sub-rock type 386 was determined at each capillary pressure step. This significantly reduced the input uncertainty of 387 the simulations compared to the porosity-based workflow.

388 **3 Results**

389 3.1 Simulation results

390 The permeability values simulated with the Pct-based model and porosity-based model were 69.93 391 mD and 9.54 mD respectively, compared to literature values of 95.5 mD to 283.6 mD (Alyafei et 392 al., 2015; Bultreys et al., 2015; Nono et al., 2014) and minipermeameter values 202.4 ± 86.9 mD. 393 To validate the experimental workflow and the image analysis results from it, we compared the 394 drainage results with a capillary pressure curve obtained using mercury intrusion capillary pressure 395 (MICP) method (Bultreys et al., 2015), rescaled to the water-decane interfacial tension (48.3 mN/m) 396 and contact angle (0°) . As shown in Figure 7, the evolution of the saturation from the imaging with 397 the capillary pressure imposed in our experiment agreed well with the MICP data, indicating that 398 the experimental operation and data processing methods were reasonable and reliable. Furthermore, 399 the simulated capillary pressure curves also showed a good match with our experiment, and the 400 input parameters were thus considered to be reasonable for the further pore-by-pore evaluation of 401 the model.



404 Figure 7. Comparison of capillary pressure curve obtain from MICP experiment, our drainage experiment, and405 simulations.

403

407 3.2 Model validation of fluid distributions

408 Figure 8 shows the slice-average probile of the brine saturation along the flow direction in the 409 sample. Visual comparision indicates that the distribution simulated by the Pct-based model 410 generally fits more closely to the experimental measurements.

411





416 Figure 8. Brine saturation slice-average profiles along fluid flow direction. (a) Experimental results. (b)417 Comparision of experiment and model predictions at 1 to 4 drainage steps.

414 415

419 The simulations were then compared to each step of the imaging experiment at a matching total 420 saturation. Figure 9 shows the comparison between saturation distributions of the experiment and 421 the two model cases. In the first and second drainage stage, due to the low capillary pressure, large 422 macro pores and well connected micropores are invaded by oil, while other macro pores remain 423 uninvaded or partially invaded. From the third capillary pressure on, the displacement takes place 424 almost completely in the microporous regions. This general trend was captured by both models. 425 However, visual inspection of the fluid saturation maps shows that the new Pct-based model 426 performs significantly better than the classical porosity-based model in predicting the fluid 427 distribution.

428 The observed discrepancy in the porosity-based model is related to the fact that the porosity-based 429 model (similarly to most classical multi-scale models) assumes that regions with higher porosity 430 have larger pore sizes and are more easily drained. Contrary to this classical assumption, the 431 experiment shows that the sub-rock type with the lowest average porosity has the lowest water 432 saturation at high capillary pressure steps (Figure 10). While the model contained enough degrees 433 of freedom for it to be tuned to the sample-averaged capillary pressure curve in Figure 7, this is 434 not the case for the pore-scale distribution of the fluids. Matching this distribution is known to be 435 a crucial issue to obtain reliable relative permeability curves from the model (Bultreys et al., 2020; 436 Gharbi & Blunt, 2012; Ruspini et al., 2017). Comparison to the novel Pct-based method, which is

437 directly based on the breakthrough pressure of each pore and provides a much better match, shows

that the fluid distribution discrepancy can be resolved by avoiding the basic assumption that higher

- 439 porosities are related to lower intrusion capillary pressures in the sub-rock typing.
- 440





441 Figure 9. Saturation map comparison at each drainage pressure. The left column is experimental results. The





Figure 10. Average experimental saturation for the 3 micro-phase regions determined by the porosity-based subrock typing, for each capillary pressure step. These are the high (0.4-1), middle (0.2-0.4) and low (0-0.2) porosity regions respectively.

448

449 3.3 Saturation error

To investigate the accuracy of the predicted saturations further, we compared the simulations to the experiment directly on the image by calculating the squared saturation error in each voxel: $(S_{w_exp} - S_{w_mod})^2$. Cross-sections of these saturation error maps are are shown in Figure 11(a). The Pct-based model generally shows higher accuracy than the porosity-based model, especially unresolved regions at high capillary pressures.

455 The saturation error in certain open pores and in pores just above the image resolution is relatively 456 large in the beginning of drainage in both models (pressure steps 1-3). To explain this, Figure 12 457 shows the pore radius distribution of water filled macropores and throats in the experiment and in 458 the Pct-based model. We define the discrepancy of the filling state as the percentage of network 459 elements that were occupied by a different fluid in the simulation than in the experiment (Bultreys 460 et al., 2018). If more than half of the fluid within a single pore or throat was wetting phase, the 461 pores or throats were considered to be wetting phase occupied, otherwise, they were classified as 462 non-wetting phase filled. As expected, the non-wetting phase invaded large open pores and throats 463 in the beginning, and almost all the brine within them was displaced before increasing to the fourth

464 pressure. Similar to single-scale models (Bultreys et al., 2018), the pores and throats with 465 intermediate size had the largest filling discrepancy. At the first drainage pressure, the occupancy 466 for pores with large radius (>12 µm) had a close match. However, the simulation predicted that all 467 small pores were wetting-filled while the experiment did not show this behaviour. The throat filling 468 error also reached 18.14% in this step. As shown in previous studies (Bultreys et al., 2018), the 469 experimental fluid distribution and filling states at the pore scale are generally not fully 470 reproducible, even in repeated experiments. Some of the pore-by-pore errors are likely caused by 471 wrong intrusion pressure predictions in throats that are only just above the resolution. Furthermore, 472 we only imaged the central part of the sample and the boundary conditions are therefore not exactly 473 the same in the model and the experiment, which increased the pore-by-pore uncertainty of the 474 simulation.

475 To quantitatively compare the saturation errors at each drainage step, we calculated the absolute 476 average root mean square error (δ_{Abs}) and the volume weighted average root mean square error 477 (δ_{VolWei}) of the models over the entire image:

478
$$\delta_{Abs} = \sqrt{\frac{\Sigma (S_{w_exp} - S_{w_mod})^2}{N}}$$
(9)

Г

479
$$\delta_{VolWei} = \sqrt{\frac{\Sigma \left((S_{w_exp} - S_{w_mod}) \cdot \varphi \right)^2}{N}}$$
(10)

480 Where S_{w_exp} and S_{w_mod} are brine saturation of each voxel from experimental measurments and 481 simulations respectively, φ is the corresponding porosity of each voxel, *N* is the total number of 482 pore voxels.

Figure 11(b) indicates that the saturation error in the porosity-based model at high drainage pressures is significantly higher than that at low pressures, while the Pct-based model shows a smoother trend and lower errors, decreasing from 0.34 to 0.26 with the increase of pressures. The volume-weighted error in the porosity-based model is almost twice as high as in the Pct-based model in drainage steps 3 to 6.

Step	Pct-based mo	del error map	Porosity-based model error map		
	X-Y plane	X-Z plane	X-Y plane	X-Z plane	





Figure 11. Saturation error. (a) Saturation error map. The left two columns (X-Y plane and X-Z plane maps respectively) are saturation discrepancy between experiment and Pct-based model simulations. The right two columns (X-Y plane and X-Z plane maps respectively) are saturation discrepancy between experiment and porosity-based model simulations. The balck is solid grain. The blue color represents the simulated saturation error is very small, while the red color denotes the error is large. (b) Absolute saturation error (left) and volume weighted saturation error (right) predicted by the two models.





498

Figure 12. Water-filled open pore (top) and throat (bottom) size distribution after each drainage step.

500 4 Conclusions

501 In this work, we proposed a multi-scale PNM validation workflow using drainage experiments 502 imaged with micro-CT, to study the factors that influence the uncertainty of pore-scale modelling 503 and simulation. The differential imaging technique was used to quantify the porosity map and the 504 saturation distribution during capillary-dominated drainage in an Estaillades limestone sample. A 505 "classical" porosity-based and a novel invasion-capillary-pressure-based sub-rock typing methods 506 were used to characterize the microporosity, followed by a multi-scale PNM extraction. The 507 continuum scale properties and pore-scale multiphase distribution from the two models were then 508 compared to experimental data.

509 We showed that the P_c -curves simulated by both models matched our image-based capillary 510 pressure curve and an MICP curve of the rock type. The porosity-based model performed poorly 511 in simulating multiphase fluid distribution at the pore-scale, while the novel Pct-based model 512 significantly improved the prediction of pore filling states. This indicated that the multiphase 513 transport behavior within sub-resolution pores was poorly correlated to the sub-voxel porosity. 514 Further research should indicate if this is due to micropore geometry variations or due to dynamic 515 effects inside the microporosity, e.g. inhomogeneous drainage due to extremely low brine 516 conductivities that slow down the invasion to very long time scales. The methodology presented 517 in this work was proven to be a robust approach in decreasing the uncertainty of pore-scale 518 modelling and can be extended to other complex reservoir rocks modelling, to provide more 519 insights on, for example, CO₂ sequestration and reservoir management.

520 Despite the improved results obtained with the Pct-based method, the determination of input 521 petrophysical properties for microporous flow zones is still challenging and was shown to be a key factor affecting the reliability of the simulation. Furthermore, this current "best-fit" model is generated based on the fact that we have the full information of the drainage experiment. While this may be useful as a hybrid workflow to calculate properties that are difficult or impossible to measure in unsteady-state experiments, notably relative permeability, performing the experiment itself is time-consuming and complex. Therefore, further work should point out if the Pct-based rock-typing can serve as a baseline to develop more straightforward sub-rock typing methods.

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Supporting Information for

Anchoring Multi-Scale Models to Micron-Scale Imaging of Multiphase Flow in Rocks

Shan Wang^{1,2}, Leonardo C. Ruspini³, Pål-Eric Øren³, Stefanie Van Offenwert^{1,2} and Tom Bultreys^{1,2}

1Department of Geology, Pore-Scale Processes in Geomaterials Research (PProGRess), Ghent University, Krijgslaan 281/S8, B-9000, Ghent, Belgium

2Centre for X-ray Tomography (UGCT), Ghent University, Proeftuinstraat 86, B-9000, Ghent, Belgium

3Petricore Norway A.S., Trondheim, Norway

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Introduction

Figure S1 shows fluid flow variation on X-ray radiograph.

Figure S2 shows the grey value histogram of differential image between dry scan and brine saturated scan.

Figure S3 shows the three-phase segmentation based on the porosity distribution map. Figure S4 shows microporosity distribution of the sample.



Figure S1. Fluid flow variation on X-ray radiograph to track the saturation change in the sample during the equilibration.



Figure S2. The grey value histogram of differential image between dry scan and brine saturated scan. The CT1 and CT2 are denoted as the thresholds for solid (0% porous) and open porosity (100% porous) respectively.



Figure S3. Segmentation based on the porosity distribution map. (a) Segmentation for the whole sample. The red is macropore, the yellow represents microporous region and the blue is solid grain. (b) Macropore overlap on the image. (c) Boundary of the microporous region.



Figure S4. Microporosity distribution of the sample.