Impact of multiphasic pore-scale interactions on gas hydrate formation and dissociation characteristics and kinetics: a microfluidic study

Supplementary Information

The image processing software employed in this study was ImageJ, which includes the Trainable Weka Segmentation (TWS) plugin. TWS is a machine learning-based segmentation tool that integrates the Weka library, enabling complex image segmentation through interactive classifier training. While TWS is widely used in biomedical image analysis (e.g., cell and tissue segmentation), it has proven equally effective for gas-hydrate research. Further details about TWS are available at https://imagej.net/plugins/tws/.

Figure S1 shows the gas-liquid distribution within the microfluidic chip before the formation of methane hydrate and the processing flow of each phase separation. As shown in Figure S1a, before the formation of methane hydrate, the microfluidic chip contains a blue liquid phase, a white gas phase and some white etched particles. After threshold segmentation of Figure S1a in the TWS plugin based on machine learning, Figure S1b is obtained, where white represents the liquid phase and black represents the gas phase and etched particles. Before the experiment, the chip was saturated with a liquid phase containing methyl blue. Similarly, after threshold segmentation through the TWS plug-in, the threshold segmentation map of the initial etched particles could be obtained, as shown in the black area of Figure S1c. Based on the Image Calculator in ImageJ, subtracting Figure S1c from Figure S1b can obtain the gas phase, as shown in the black area of Figure S1d. Therefore, before the formation of gas hydrates, the liquid phase (Figure S1b white), etched particles (Figure S1c black) and gas phase

(Figure S1d black) can be easily distinguished and quantified.

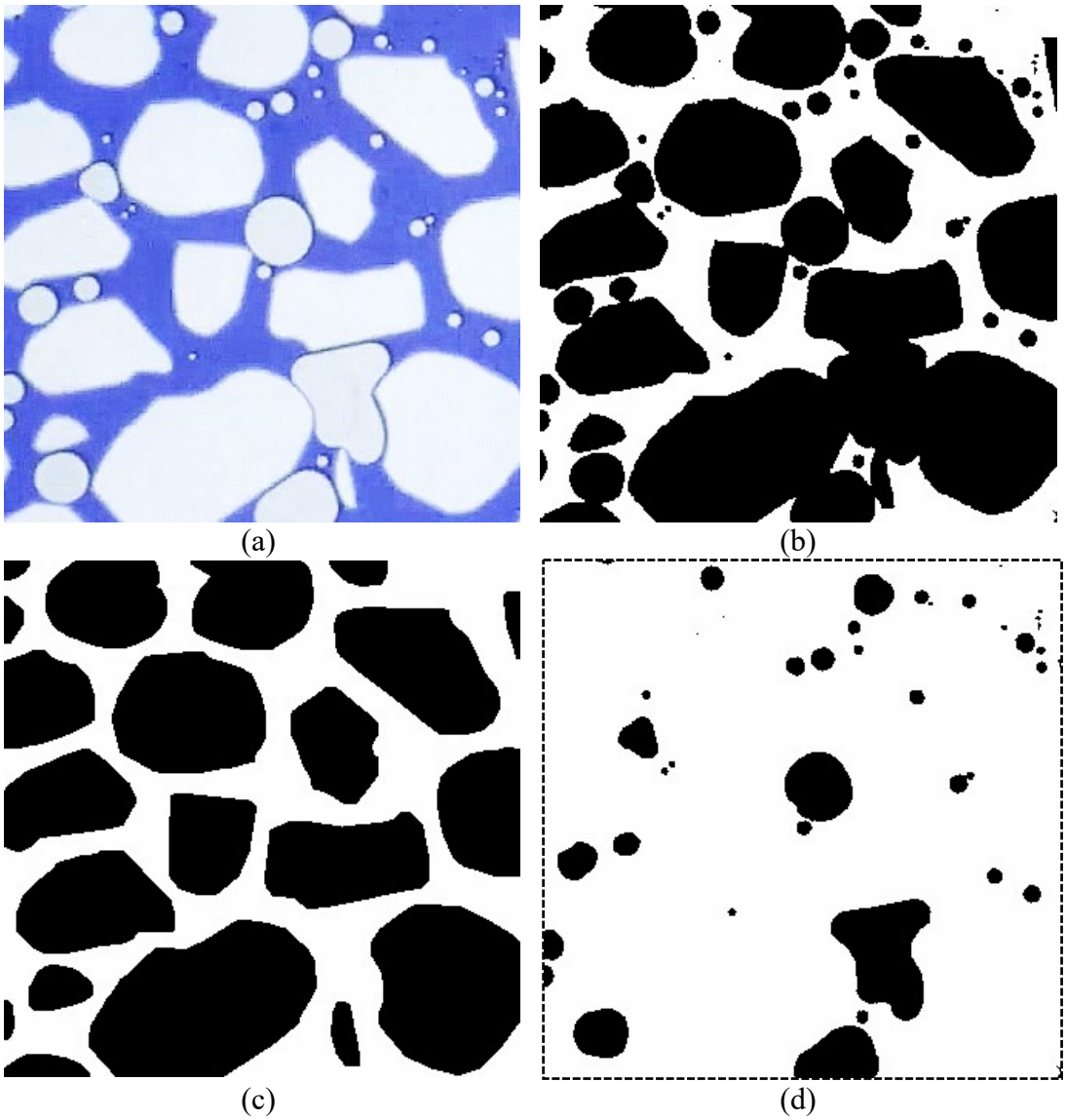


Figure S1 Phase differentiation before gas hydrate formation (a) initial image; (b) liquid; (c) etched particles; (d) gas

Figure S2 shows the liquid and gas hydrate distribution within the microfluidic chip after the formation of methane hydrate and the processing flow of each phase separation. As shown in Figure S2a, after the formation of methane hydrate, the microfluidic chip contains a blue liquid phase, a white gas hydrate phase and some white etched particles. After threshold segmentation of Figure S2a in the TWS plugin based on machine learning, Figure S2b is obtained, where white represents the liquid phase and black represents the gas hydrate phase and etched particles. During the

hydrate formation, the morphology and size of the etched particles will not change. Therefore, the etched particles are still as shown in Figure S1c. Based on the Image Calculator in ImageJ, subtracting Figure S1c from Figure S2b can obtain the gas hydrate phase, as shown in the black area of Figure S2c. Therefore, after the formation of gas hydrates, the liquid phase (Figure S2b white), etched particles (Figure S1c black) and gas hydrate phase (Figure S2c black) can also be easily distinguished and quantified.

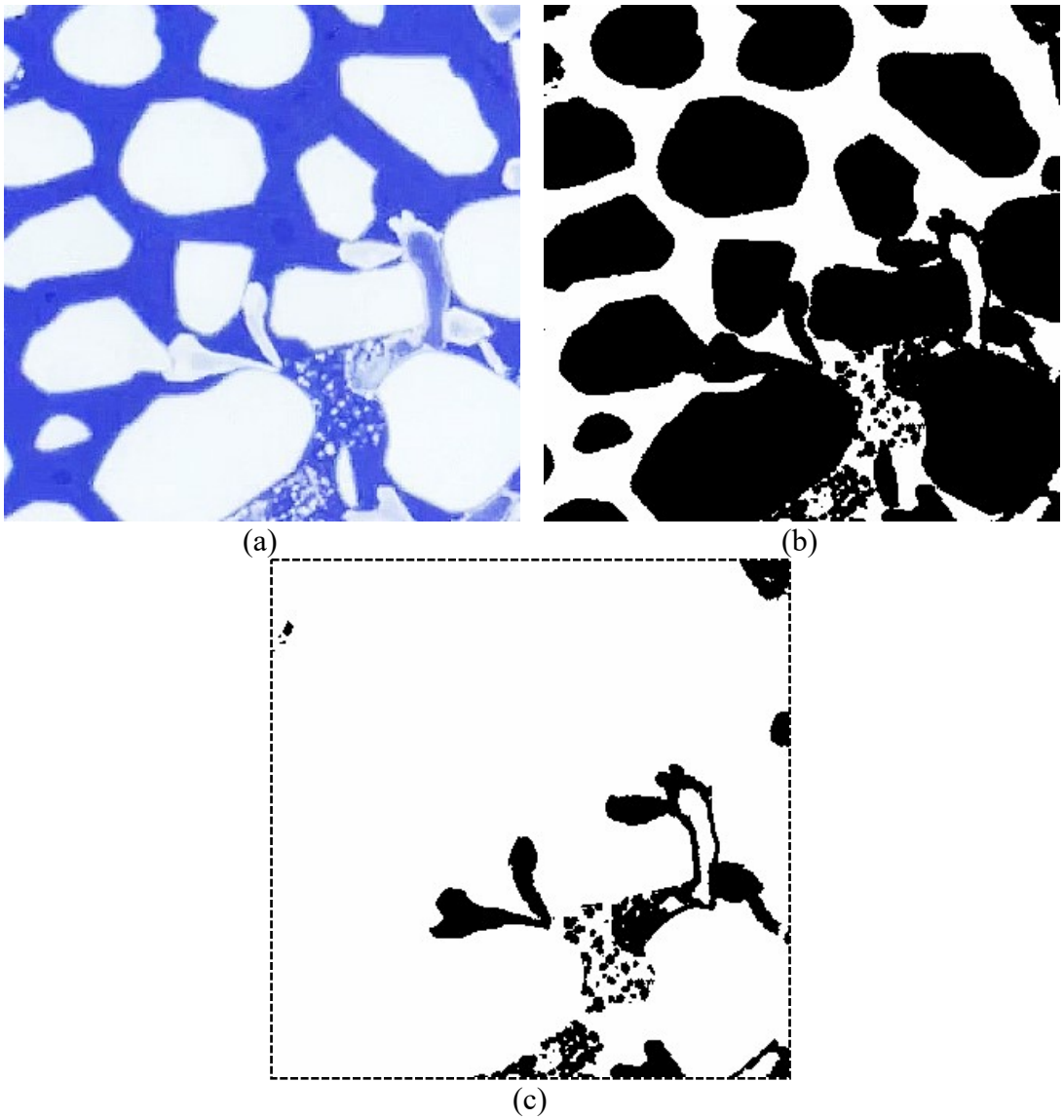
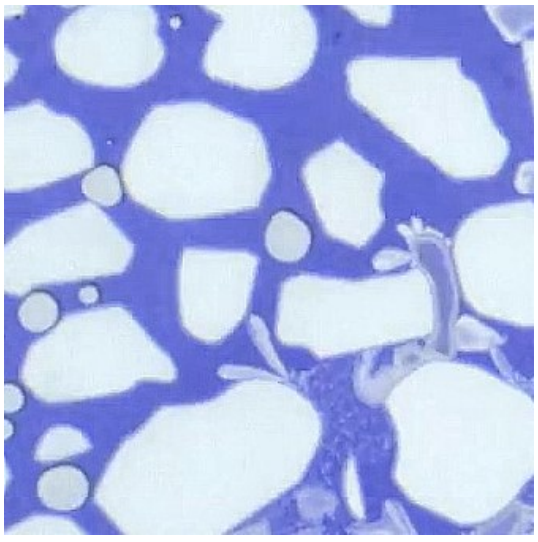


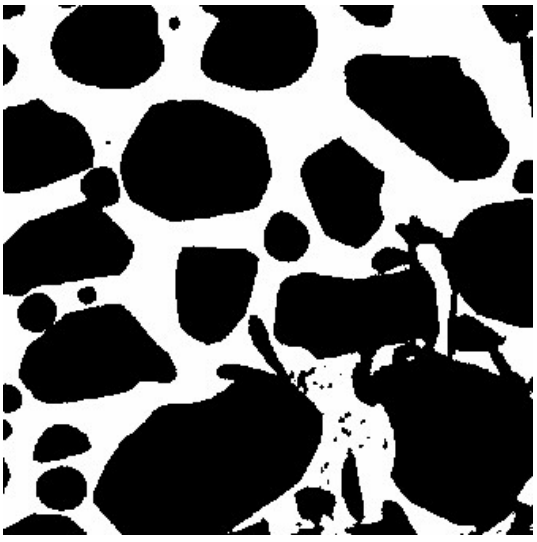
Figure S2 Phase differentiation after gas hydrate formation (a) initial image; (b) hydrate and etched particles; (c) hydrate

Figure S3 shows the liquid,gas and gas hydrate distribution within the microfluidic chip

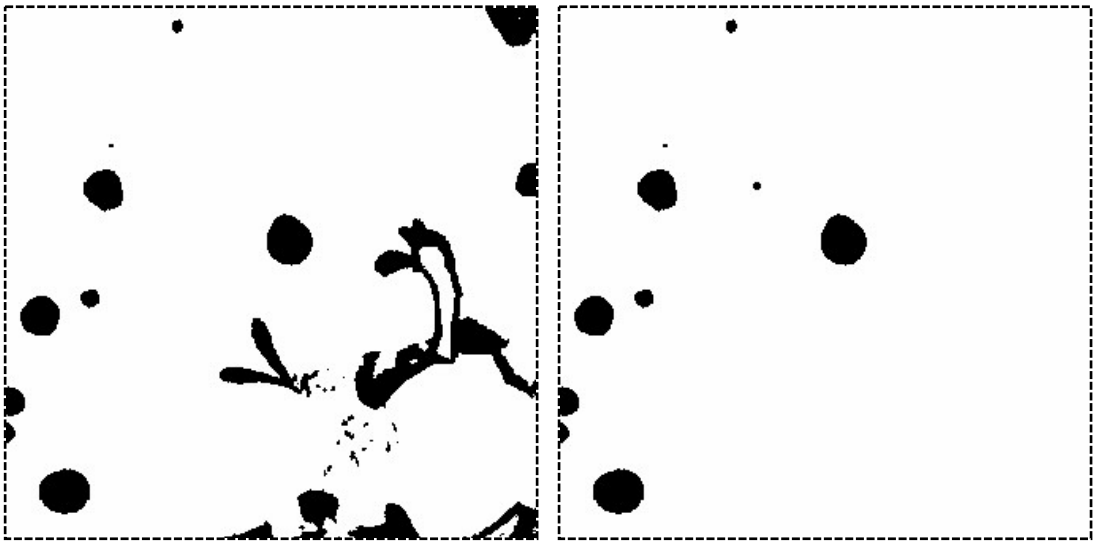
during the formation of methane hydrate and the processing flow of each phase separation. As shown in Figure S3a, the microfluidic chip contains a blue liquid phase, a white gas phase, a white gas hydrate phase and some white etched particles. After threshold segmentation of Figure S3a in the TWS plugin based on machine learning, Figure S3b is obtained, where white represents the liquid phase and black represents the gas phase, gas hydrate phase and etched particles. Based on the Image Calculator in ImageJ, subtracting Figure S1c from Figure S3b can obtain the gas phase and gas hydrate phase, as shown in the black area of Figure S3c. The morphological differences between the gas phase and the hydrate are significant. The gas phase distribution can be obtained by manually selecting the gas phase area, as shown in the black area of Figure S3d. Based on the Image Calculator in ImageJ, subtracting Figure S3d from Figure S3c can obtain the gas hydrate phase, as shown in the black area of Figure S3e. Therefore, during the formation of gas hydrates, the liquid phase (Figure S3b white), etched particles (Figure S1c black), gas phase (Figure S3d black) and gas hydrate phase (Figure S3e black) can also be easily distinguished and quantified.



(a)



(b)



(c) (d)

(e)

Figure S3 Phase differentiation during gas hydrate formation (a) initial image; (b) gas, hydrate and etched particles; (c) gas and hydrate; (d) gas; (e) hydrate