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超临界 CO₂-水驱替过程中 CO₂ 毛细管滞留

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超临界 CO₂ 驱替水驱替 CO₂ 毛细管滞留行为, 基于实验和数值模拟, 研究了 CO₂ 驱替水驱替过程中 CO₂ 毛细管滞留行为。

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3 种毛细管滞留模型 (Land's, Jurauld's 和 Spiteri's) 进行了对比。结果表明, Jurauld's 模型毛细管滞留行为与 Land's 模型毛细管滞留行为相似, 而 Spiteri's 模型毛细管滞留行为与 Land's 模型毛细管滞留行为差异较大。

超临界 CO₂ 驱替水, 驱替水, 毛细管, 滞留

DOI: 10.6052/0459-1879-16-237

363.2, V211.1+7

doi

SUPERCritical CO₂ WATER DISPLACEMENTS AND CO₂ CAPILLARY TRAPPING: MICROMODEL EXPERIMENT AND NUMERICAL SIMULATION¹⁾

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Abstract The CO₂ capillary trapping is an important scientific issue in geological carbon sequestration, but few re-searches focus on the trapping mechanism at pore scale under supercritical CO₂ condition. In this study, based on the high-pressure fluids-microscopy-micromodel experimental system, we performed drainage experiment, i.e. supercritical CO₂ displacing water, and imbibition experiment, i.e. water displacing CO₂, under the conditions of 45 °C and 8.5 MPa. The DSLR camera was used to capture pictures of CO₂-water two-phase immiscible flow and the microscopy was used to capture the capillary trapping behavior for the supercritical CO₂ at the pore scale. The

computational fluid dynamic method was adopted to simulate the two-phase fluid flow processes. The numerical results are generally in agree-

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spacing with the experimental observations, and further provide three-dimensional geometries on the interface during the drainage-imbibition processes and the trapped supercritical CO₂ droplet/cluster. Finally, the capillary trapping curve, i.e. the relationship between the initial CO₂ saturation and the residual saturation, was obtained from the numerical results, and we made an assessment of the three capillary trapping models, i.e. Land's, Jurauld's and Spiteri's trapping models. A comparison of the models performance indicates that Jurauld's model behaves slightly better than Land's model, whereas Spiteri's model behaves poorly. However, given that Land's model only contains one parameter of clear physical meaning, it is recommended for practical use.

Key words geological carbon sequestration, micromodel, two-phase flow, numerical simulation, capillary trapping

space

超临界 CO₂ 驱替水驱替 CO₂ 毛细管滞留行为, 基于实验和数值模拟, 研究了 CO₂ 驱替水驱替过程中 CO₂ 毛细管滞留行为。

CO₂ [1]. [2] CO₂ 2005 — 2050 20%~50%.

CO₂ 800 m CO₂ CO₂

CO₂ (capillary trapping residual trapping)^[5]. GCS CO₂ [6] CO₂ CO₂ CO₂

space [18-20]. Al-Raoush^[8] Chaudhary [10] CO₂ Andrew [21] CO₂ [22] CO₂ (sc CO₂) Niu [17] CO₂ CT CO₂

CT [23-25]. [18-20] [26] [27] Lenormand [20] Zhang [28]

space

CO₂ space CO₂ [7-10]

space Cottin [29]

space [10-12] CO₂ [13-14] CO₂ (1) CT(computed tomography) [15-17]; (2)

space CO₂ CO₂

space [30] [31] [32]

[33-34]. 3 [35].

(8.5 MPa, 45 °C) CO₂ CO₂

CO₂ 2017 49

() 3

1

1.1

1(a) CO₂ CO₂

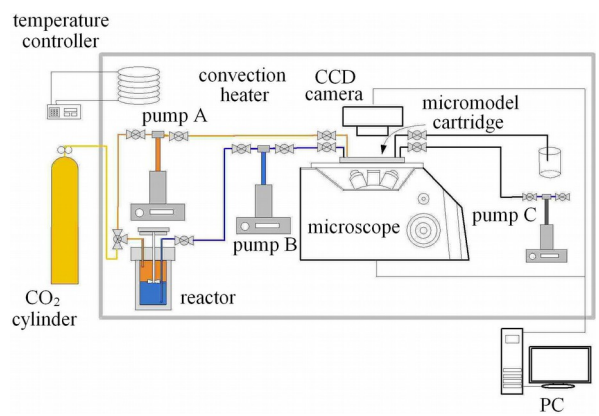
(1) (Carl Zeiss, AxioCam MRc5) (Carl Zeiss, Observer Z1.m) 20 mm 10 mm 50 μm 40 μm (pore volume) 1.97 μL 1(b). (20 mm × 10 mm) (1 / /)

(2) (Tele-dyne ISCO, 500HP×2 65 HP) 1(a) A CO₂ B C (8.5 MPa).

(3) CO₂ (Parr, model 4560) A CO₂ CO₂ (1.97 μL) CO₂ (H₂CO₃) CO₂ [24]. A (8.5 MPa)

space CO₂ ()

(4) (1)(2) (3) (Omega, Type T) (Cole Parmer, EW-89000-10) (45±1)°C.



(a)

(a) Experimental apparatus

(b)

(b) The sizes of the micromodel

Fig. 1 High-pressure fluids-microscopy-micromodel system

1.2

CO₂ CO₂
 CO₂ 4 . 3

(1) CO₂ .
 CO₂ CO₂

space .
 NaCl(Sigma-Aldrich, ACS reagent grade) 0.01 mol/L
 NaCl . CO₂ A. A
 8.5 MPa 45°C.
 24 h CO₂ .

(2) . 500 μL/min
 10 min 120°C 24 h.

(3) CO₂ . C
 8.5 MPa. C
 (0.01 mol/L NaCl) 200 μL/min

space . f_s

$$f_s = \sigma \cdot \kappa \cdot n \cdot \delta_{r_T} \quad (3)$$

σ , κ , n

(δ_{r_T}) Dirac delta 1 0.

(1) (1)
 (volume of fluid VOF) [35].
 α (2)

0, in the nonwetting phase (CO₂)

space 5 min. A
 CO₂ A . 3 h CO₂
. 3 h A 192 μL/min
 CO₂ (drainage). 1
 (1.97 μL)
 2 s . A B
 (192 μ L/min)
 (imbibition). 3 s

(4) CO₂ .
 5 min

space $\alpha = 1$, in the wetting phase (CO₂)

$$0 < \alpha < 1, \text{ on the interface}$$

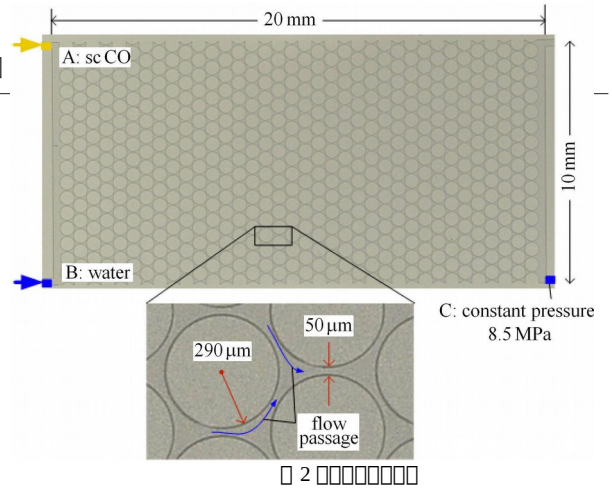


Fig. 2 Illustration of the volume of fluid method

space(4)

space CO₂

[23]
 [23].

2

2.1

CO₂

space

$$\frac{\partial \alpha}{\partial t} + \nabla \cdot (\alpha \mathbf{u}) + \nabla \cdot [\alpha \cdot (1 - \alpha) \mathbf{u}_r] = 0 \quad (5)$$

\mathbf{u}_r [36].

space (4) (1)

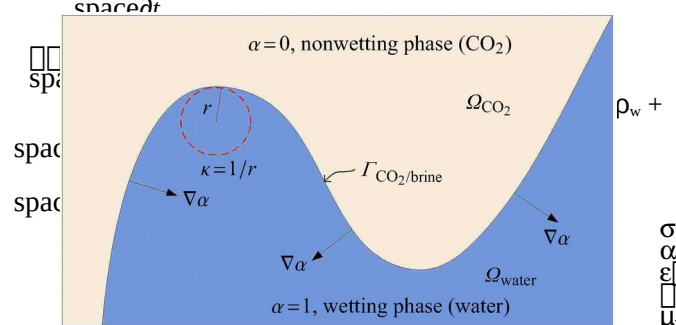
space $\partial(\rho \mathbf{u})$

$$\begin{aligned} & + \nabla \cdot (\rho \mathbf{u}) = \\ & - \nabla p + \\ & \nabla \cdot \mu \\ & (\nabla \mathbf{u} + \\ & \nabla^T \mathbf{u} + \mathbf{f} \end{aligned}$$

$$e \mu = \alpha \mu_w + (1 - \alpha) \mu_{CO_2}$$

space

(6)



σ
 α
 ϵ
 μ_w

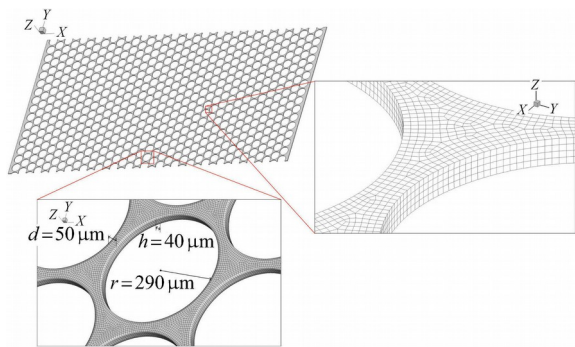


Fig. 3 Finite volume mesh for the micromodel

$$\nabla \cdot \mathbf{u} = 0 \quad (2)$$

ρ_{CO_2}
 CO₂
 \mathbf{u} CO₂ p μ \mathbf{f}_s

$$\mathbf{n} = \nabla \alpha \quad \kappa = \nabla \cdot (\nabla \alpha / |\nabla \alpha|) \quad \mathbf{n}, \kappa \quad \alpha \quad (3) \quad (1)$$

CO₂
 α

$$\nabla \alpha = 1$$

$$\mathbf{n} \cos$$

$$\mathbf{n} \sin$$

$$(8)$$

$$-\nabla \cdot (\rho(\alpha) \mathbf{u}) + \frac{\partial \mu(\alpha)}{\partial t} + \nabla \cdot (\rho(\alpha) \mathbf{u} \mathbf{u}) +$$

$$\nabla \cdot \mathbf{u}$$

$$\nabla p + \sigma \cdot \nabla = 0$$

$$\nabla \alpha$$

$$\nabla \alpha \cdot \delta_r = 0$$

\mathbf{n}_s \mathbf{n}_t

$$\sigma \alpha \chi \epsilon \frac{\partial \alpha}{\partial t} + \nabla \cdot (\alpha \mathbf{u}) + \nabla \cdot [\alpha \cdot (1 - \alpha) \mathbf{u}_r] = 0$$

CO₂
 space (7)
 space (1) (drainage)
 $\nabla p = 0$ $\alpha = 0$
 $\frac{\partial \mu}{\partial x} = \frac{\partial \mu}{\partial y} = \frac{\partial \mu}{\partial z} = 0$ $p = 8.5 \text{ MPa}$ $\nabla \alpha = 0$ (2)

OpenFOAM^[36]
 Open-FOAM (7)
 3 1 962 924
 2 712 905 6~9 μm. 1

Fig. 3 Finite volume mesh for the micromodel

1 CO₂

Table 1 The parameters for the supercritical CO₂-water

two-phase fluid flow	
space (imbibition)	$B \mathbf{u} = 0.32 \text{ m/s}$
	$\nabla p = 0$ $\alpha = 1$ $\frac{\partial \mu}{\partial x} = \frac{\partial \mu}{\partial y} = \frac{\partial \mu}{\partial z} = 0$ $p = 8.5 \text{ MPa}$ $\nabla \alpha = 0$ (3)
	$B \mathbf{u} = 0 \text{ m/s}$ $p = 8.5 \text{ MPa}$ $\nabla \alpha = 0$.
II	
96 CPU	CO ₂
23 040	·
3.3	
768 CPU	115 200 ·

3
 1.2 3 3
 CO₂ CO₂ 1
 CO₂ 3
 $\nabla \alpha$ [39] CO₂

3.1 CO₂

4	θ
45 °C, 8.5 MPa	θ
Water (NaCl 0.01 mol/L)	
Supercritical CO ₂	
3	
3	Ca
density/(kg·m ⁻³)	993.9 ^[37] 281.8 ^a
viscosity/(kg·s ⁻¹ ·m ⁻¹)	5.972 × 10 ^{-4a} 2.275 8 × 10 ^{-5a}
interfacial tension/(N·m ⁻¹)	33.1 ± 0.5 ^[38]

space (M). Ca M

Note: a from National Institute of Standards and Technology (NIST) web-based database (<http://webbook.nist.gov/chemistry/fluid/>)

$$Ca = \frac{\rho_i u_i}{\sigma}$$

$$M = \frac{\mu_i}{\mu_2}$$

$$(9)$$

$$(10)$$

8.5 MPa 45 °C
 10 CO₂ 22 °
 OpenFOAM

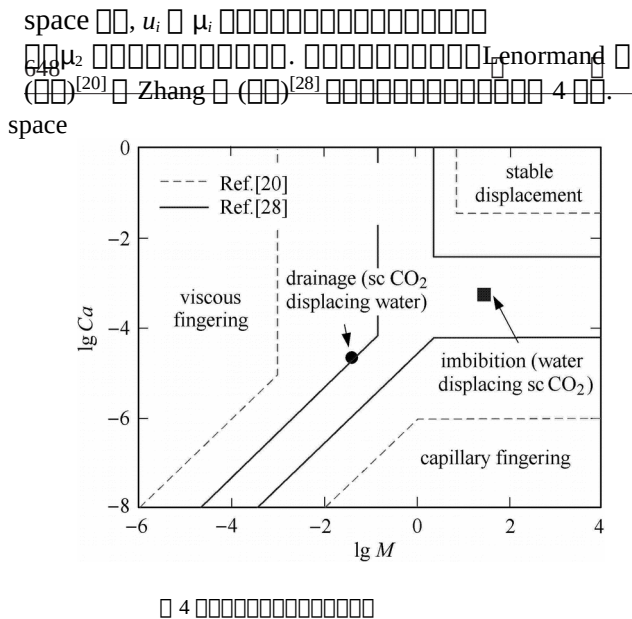


Fig. 4 The phase diagram for displacement pattern within the two-phase immiscible fluid flow

Figure 1 shows the experimental results (8) and (9) for drainage (drainage of CO₂ displacing water) and imbibition (water displacing CO₂) respectively. For drainage, $Ca = 2.24 \times 10^{-5}$, $M = 0.038$ (im- bition) $Ca = 5.87 \times 10^{-4}$, $M = 26.24$. Figure 4 shows the phase diagram (drainage) and imbibition.

Figure 5 shows the numerical results for the two-phase fluid flow after 0.1 s of drainage. The CO₂ displaces water in the pore space. The CO₂ phase is shown in red and water in blue. The CO₂ phase is shown in red and water in blue. The CO₂ phase is shown in red and water in blue.

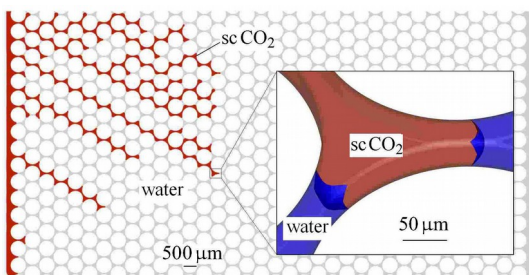


Figure 6 shows the numerical results for the two-phase fluid flow after 1 s of drainage. The CO₂ displaces water in the pore space. The CO₂ phase is shown in red and water in blue. The CO₂ phase is shown in red and water in blue. The CO₂ phase is shown in red and water in blue.

Fig. 6 Comparison between numerical results and the DSLR captured images after 1 s of drainage ((a) experimental result, (b) numerical simulation)

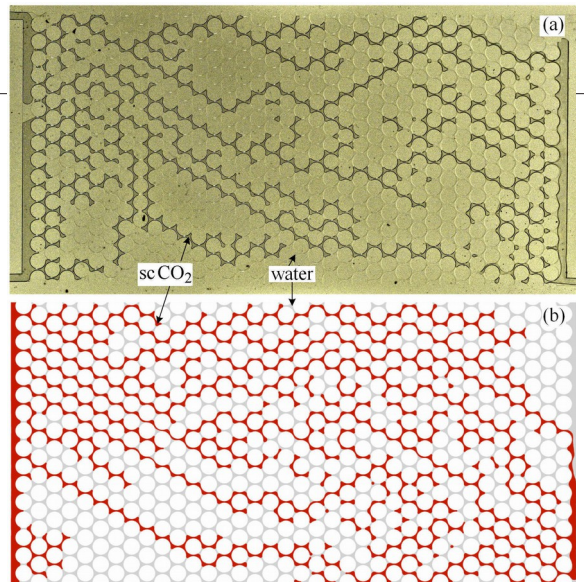


Fig. 6 Comparison between numerical results and the DSLR captured images after 1 s of drainage ((a) experimental result, (b) numerical simulation)

Figure 7 shows the numerical results for the two-phase fluid flow after 0.1 s of imbibition. The water displaces CO₂ in the pore space. The water phase is shown in blue and CO₂ in red. The water phase is shown in blue and CO₂ in red. The water phase is shown in blue and CO₂ in red.

Figure 8 shows the numerical results for the two-phase fluid flow after 1 s of imbibition. The water displaces CO₂ in the pore space. The water phase is shown in blue and CO₂ in red. The water phase is shown in blue and CO₂ in red. The water phase is shown in blue and CO₂ in red.

Fig. 7 The numerical results for the two-phase fluid flow after 0.1 s of drainage

Figure 8 shows the numerical results for the two-phase fluid flow after 1 s of imbibition. The water displaces CO₂ in the pore space. The water phase is shown in blue and CO₂ in red. The water phase is shown in blue and CO₂ in red. The water phase is shown in blue and CO₂ in red.

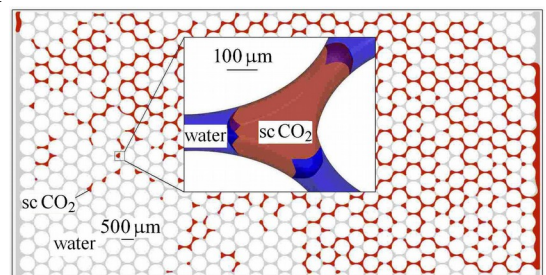


Fig. 7 The numerical results for the two-phase fluid flow after 0.1 s of imbibition

Figure 8(a) shows the CO₂ phase is shown in red and water in blue. The CO₂ phase is shown in red and water in blue. The CO₂ phase is shown in red and water in blue.

Fig. 8(a) shows the CO₂ phase is shown in red and water in blue.

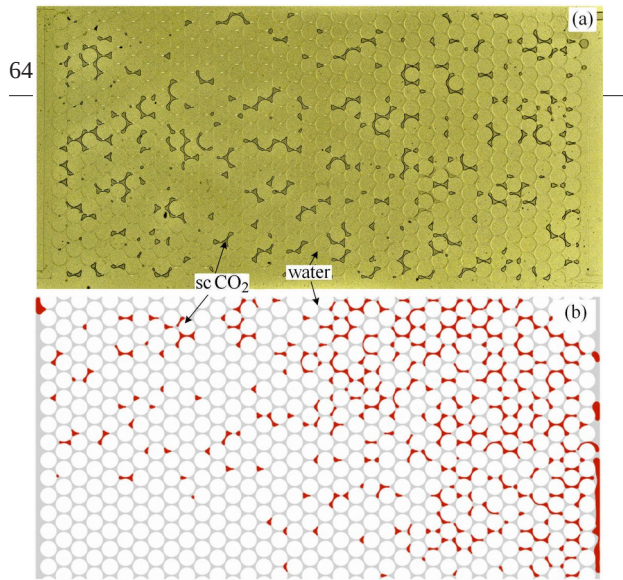


Fig. 8 Comparison between numerical results and the DSLR captured images after 1 s of imbibition ((a) experimental result, (b) numerical simulation)

Fig. 8 Comparison between numerical results and the DSLR captured images after 1 s of imbibition ((a) experimental result, (b) numerical simulation)

space (27.7±2.3)% [8]. The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(a)) and at a single pore (Fig. 9(d)~(g)) is compared with the distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(m)) and at a single pore (Fig. 9(h)~(k)). The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(l) and (n)) and at a single pore (Fig. 9(i)~(j)) is compared with the distribution of trapped CO₂ droplets/clusters at a single pore (Fig. 9(h)~(k)).

3.2 CO₂ droplets/clusters

CO₂ droplets/clusters are observed in the experimental results (Fig. 9(a)) and in the numerical simulation (Fig. 9(b)). The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(a)) and at a single pore (Fig. 9(d)~(g)) is compared with the distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(m)) and at a single pore (Fig. 9(h)~(k)). The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(l) and (n)) and at a single pore (Fig. 9(i)~(j)) is compared with the distribution of trapped CO₂ droplets/clusters at a single pore (Fig. 9(h)~(k)).

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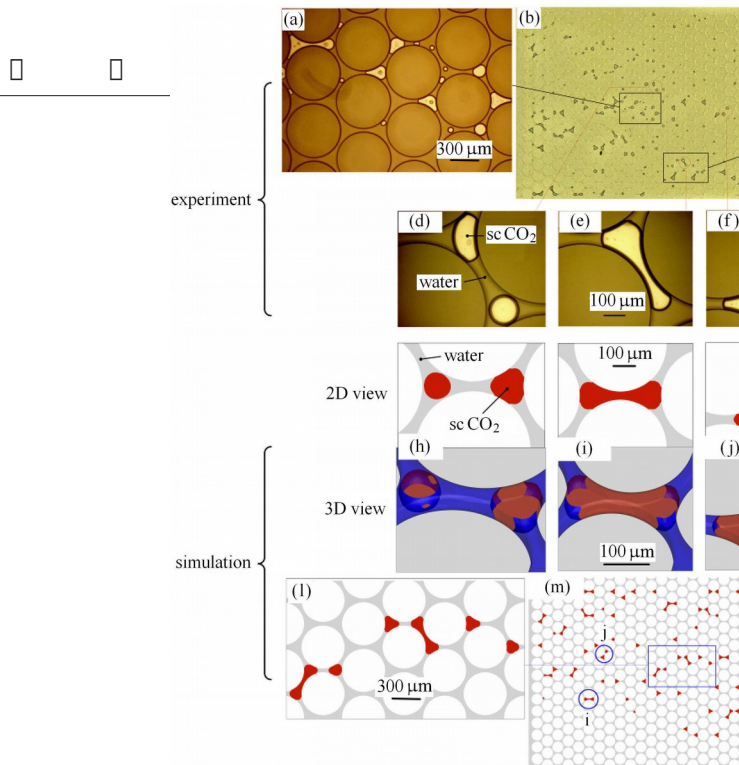


Fig. 9 The observed (a) ~ (g) and simulated (h) ~ (n) shape and distribution of trapped CO₂ droplets/clusters: (a) and (c) the distribution of trapped CO₂ droplets/clusters at multiple pores; (b) the distribution of trapped CO₂ droplets/clusters at the full scale of micromodel; (d) ~ (g) the distribution of trapped CO₂ droplets/clusters at a single pore. (m) the distribution of trapped CO₂ droplet/cluster at the full scale of micromodel; (l) and (n) the distribution of trapped CO₂ droplets/clusters at multiple pores; (h) ~ (k) the distribution of trapped CO₂ droplets/clusters at a single pore

space (27.7±2.3)% [8]. The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(a)) and at a single pore (Fig. 9(d)~(g)) is compared with the distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(m)) and at a single pore (Fig. 9(h)~(k)). The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(l) and (n)) and at a single pore (Fig. 9(i)~(j)) is compared with the distribution of trapped CO₂ droplets/clusters at a single pore (Fig. 9(h)~(k)).

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3.3 Simulation of trapped CO₂ droplets/clusters

CO₂ droplets/clusters are observed in the experimental results (Fig. 9(a)) and in the numerical simulation (Fig. 9(b)). The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(a)) and at a single pore (Fig. 9(d)~(g)) is compared with the distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(m)) and at a single pore (Fig. 9(h)~(k)). The distribution of trapped CO₂ droplets/clusters at the full scale of micromodel (Fig. 9(l) and (n)) and at a single pore (Fig. 9(i)~(j)) is compared with the distribution of trapped CO₂ droplets/clusters at a single pore (Fig. 9(h)~(k)).

space S_{nwr}

$$= \frac{1 + CS_{nwi}}{space}$$

(11)

space 8.5 MPa

(6 h)

“ ”

space, S_{nwi} S_{nwr}

C Land, S_{nwi}

Land

s

p

a

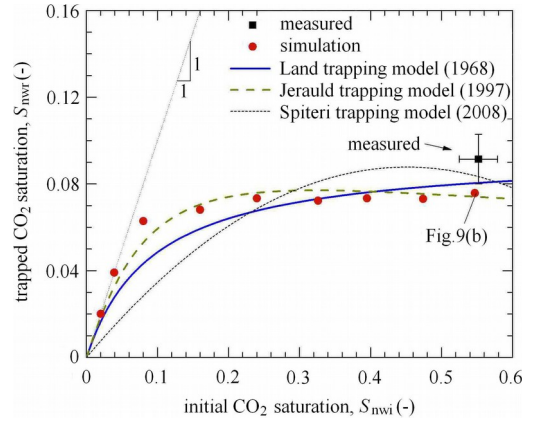
c

e

a
*
S
n
w
,
m
a
x

J
e
r
a
u
l
d
[
4
6
]
L
a
n
d

L
a
n
d



$$spaceC = \frac{1}{S_{nwr,max}}$$

$$space1 = \frac{1}{S_{nwr,max}}$$

space(12)

s

p

a

c

e

648

space 3

CO₂ 2017. CO₂ 5 s S_{nwi} S_{nwr} (2 s)

10 2 2 Jerauld Land 0.006 6 0.01 Spiteri 0.019 0. Jerauld Land Jerauld Land Spiteri

2

Table 2 The fitting parameters and the root-mean-square-error (RMSE) for the three models

Prediction model	Optimal fitting coefficient	Mean-squared error
Land mode, Eq.[10]	C = 10.62	0.010 0
Jerauld mode Eq.[12]	C = 14.09, b = 4.42	0.006 6
Spiteri mode Eq.[13]	α = 0.389 4, β = 0.431 8	0.019 0

Spiteri 30% Jerauld Land Spiteri

10 S_{nwi} = (58.3 ± 2.1)% S_{nwr} = (9.2 ± 1.1)% S_{nwi} = 61.3% S_{nwr} = 7.9%

3 Raeini [33] LV60 CO₂ 0.5 CO₂ 2%~4% 1.3%

4

CO₂ OpenFOAM CO₂ 3

(1) CO₂

(2) CO₂ CO₂

(3) 3 Jerauld Land Land Land

CO₂ CO₂ GCS CO₂

$$S_{nwr} = \frac{S_{nwi}}{1 + CS^{1+b/C}}$$

(13)

b = 0 Jerauld Land Spiteri [47]

10 CO₂ Fig. 10

comparison of the capillary trapping curve between the numerical simulation/experimental data and three different models

10 7%

$$S_{nwr} = \alpha S_{nwi} - \beta S^2$$

α β

(14)

1:1 CO₂ 0.07 CO₂ Jerauld Land

- □ □ □
- 648 Zhang W, Li Y, Xu T, et al. Long-term variations of CO₂ trapped in different mechanisms in deep saline formations: a case study of the Songliao Basin, China. *International Journal of Greenhouse Gas Control*, 2009, 3(2): 161-180
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