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NEW METHOD FOR INFERRING THE MEAN PORE SIZE OF SHALE USING IMBIBITION-DIFFUSION DATA

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ABSTRACT

After the multistage hydraulic fracturing operations, it is generally observed that the shale gas well is characterized by low flowback rate and high-salinity flowback water. It can be explained by the high water imbibition and ion diffusion capacity of gas shale that is significantly different from conventional sandstone reservoirs. The research on imbibition and ion diffusion is significant for the understanding of shale formation characterization. A new method to evaluate the mean pore size was developed using the experimental data of water imbibition and ion diffusion in gas-saturated shale. The mathematical model was derived theoretically depending on Darcy's law and continuum equation. The results show that the imbibition and diffusion rate can be calculated by imbibition-diffusion curves, and the ratio of imbibition rate to diffusion rate is related to mean pore size. The value of mean pore size is inferred, which is verified by the mercury injection test. This research is significant for volumetric analysis and chemical analysis after fracturing operations to understand the characteristics of gas shale.

KEYWORDS

Shale gas, Imbibition, Diffusion, Fracturing

INTRODUCTION

Shale gas, one of the unconventional natural gas resources, becomes the focus of the world. The economical explorations have been taken in several basins of America, Canada and China. As shale gas formation has the characteristics of low-porosity and low-permeability, the multistage fracturing technology must be taken to realize economical exploration (Vera and Ehlig-Economides, 2014).

The field studies of network fracturing illustrate that the flowback efficiency of fracturing fluid is generally low. The flowback efficiency of shale gas wells in America is 20~40%, and that of several shale gas wells in Fuling of China is evenly lower than 5~10% (Zhong, 2011). Many researches explain that this is caused by spontaneous imbibition. High capillary pressure due to micro-nano pores and ultra-low initial water saturation ("super dry") leads to the strong imbibition capacity of shale formation that substantially exceeds conventional sandstone formation. (Lal, 1999). The clay mineral content of shale is high, which will result in strong chemical effects. When shale contacts with water, the water is imbibed into shale under the chemical effects. So, the imbibition mechanism in shale is more complicated than that in conventional sandstone. In particular, the existence of smectitie and illite can greatly enhance the water imbibition rate (Yang et al., 2016).

Another observation is that the salinity of recovered water rises quickly over time and can reach to 20%. The salinity of the slick water fracturing fluid is about 0.1% (Fakcharoenphol et al., 2014). The salinity of recovered water is mainly caused by the ion diffusion into fracturing fluid. The saline ions in flowback fluid are mainly from the dissolution of the minerals and crystal salt on pore wall. The clay mineral is the key factor for the saline ion content in shale formation (Bohacs et al., 2013). On the one hand, clay minerals have abundant soluble ions, and they will immediately dissolve when contacting with water to increase the fracturing fluid salinity. On the other hand, the clay crystal layer could act as semipermeable membrane. The water molecules can pass freely, and saline ions pass in part or cannot pass (Ghanbari et al., 2013). During deposition and compaction of shale formation, the salinity of extracted water is relatively low due to semipermeable membrane effects. So much ions are detained in pore space of shale and gather in the clay mineral surface, forming high salinity water film or crystal salt (Haluszczak et al., 2013).

The flowback rate and recovered water salinity contribute to understanding the reservoir characteristics and evaluating the development of fracturing network. Hence, the research on shale reservoir imbibition and ion diffusion is in favor of understanding the flowback characteristics of shale gas well. The paper will establish the

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imbibition-ion diffusion model and propose a new method to evaluate the mean pore size based on the experimental data of water imbibition and ion diffusion in gas-saturated shale.

1. MATHEMATICAL BACKGROUND

In order to establish the fracturing fluid imbibition and ion diffusion model, the authors propose the following assumptions for the physical process:

- Only one face can contact with water
- The capillary bundles are used to describe shale pore structure.
- Saline ions are evenly distributed on pore walls
- When imbibition front contacting with pore wall, the ions on wall surfaces dissolve rapidly and diffuse into water to increase the fluid salinity.

According to Darcy's formula, the gas and water flow during water imbibition is given by

$$q_g\left(x\right)=\frac{kk_{rg}}{\mu_g}A_c\frac{dP_g}{dx}$$
$$q_w\left(x\right)=\frac{kk_{rw}}{\mu_w}A_c\frac{dP_w}{dx} \quad (1)$$

where $q_g\left(x\right)$ and $q_w\left(x\right)$ are gas and water flux respectively; k_{rg} and k_{rw} are the relative permeability of gas and water respectively; k is absolute permeability; P_g and P_w are the pressure of gas and water respectively, A_c is the sectional area; μ_g and μ_w are the viscosity of gas and water respectively.

Only one face contacts with water, the gas flow is equal to water flow.

$$q_g\left(x\right)=-q_w\left(x\right) \quad (2)$$

On gas-water interface, the gas-water two phase pressures can be given by

$$P_g=P_w+\left(P_\pi+P_c\right) \quad (3)$$

where P_c is capillary pressure, and P_π is osmotic pressure.

Simultaneous Eq. (1), (2) and (3), the variation of imbibition front x with time can be given by

$$x=\sqrt{\frac{2k\left(P_c+P_\pi\right)t}{\left(\frac{\mu_g}{k_{rg}}+\frac{\mu_w}{k_{rw}}\right)\phi\left(S_{wf}-S_{wi}\right)}} \quad (4)$$

where ϕ is porosity; S_{wf} , S_{wi} are the front water saturation and initial water saturation; t is imbibition time.

During shale imbibition, the variation of imbibed water volume with time is

$$V_{imb}/A_c=\sqrt{\frac{2k\left(P_c+P_\pi\right)\phi\left(S_{wf}-S_{wi}\right)t}{\left(\frac{\mu_g}{k_{rg}}+\frac{\mu_w}{k_{rw}}\right)}}=A\sqrt{t} \quad (5)$$

Where V_{imb} is the imbibed water volume; A is the imbibition rate.

During shale imbibition, the variation of fluid conductivity caused by ion diffusion is given by

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$$G/A_c = 2\frac{C}{V} + \frac{4\pi rnc}{V}\sqrt{\frac{2k\left(P_c + P_\pi\right)}{\left(\frac{\mu_g}{k_{rg}} + \frac{\mu_w}{k_{rw}}\right)\phi\left(S_{wf} - S_{wi}\right)}}\sqrt{t} \tag{6}$$

$$G/A_c = 2\frac{C}{V} + D\sqrt{t} \tag{7}$$

where G is fluid conductivity; C is the ion adhesion amount in unit area; V is the fluid volume; D is the ion diffusion rate.

The ratio of imbibition rate to ion diffusion rate is given by

$$\frac{A}{D} = \frac{Vr\Delta S_w}{4C} \tag{8}$$

As the shale is compact, the front water saturation is high, and is assumed to be 1. Meanwhile, the samples are dried on the oven before experiments, thus the initial water saturation is 0, so ΔS_w is nearly to 1. The average pore size of shale is given by

$$r = \frac{4CA}{DV\Delta S_w} = 4\cdot\frac{A}{D}\cdot\frac{C}{V} \tag{9}$$

2. EXPERIMENTS

2.1. Materials

The shale samples are from China typical shale formation-Longmaxi formation of Sichuan Basin. Sichuan Basin is the most potential shale gas area in China. At present, Fuling District in Chongqing has made great breakthroughs for shale gas exploitation.

The mineral composition of shale is measured by D/MAX 2500X X-ray diffract meter. The total content of clay minerals is 36.9%, and the clay mineral types are mostly illite/smectite mixed-layer. The quartz mineral content is 40.3%. Therefore, Longmaxi shale has the characteristics of high clay mineral content and high quartz content (Figure 1). Figure 2 illustrates that shale develops organic materials that develop large amount of pores with well pore connectivity.

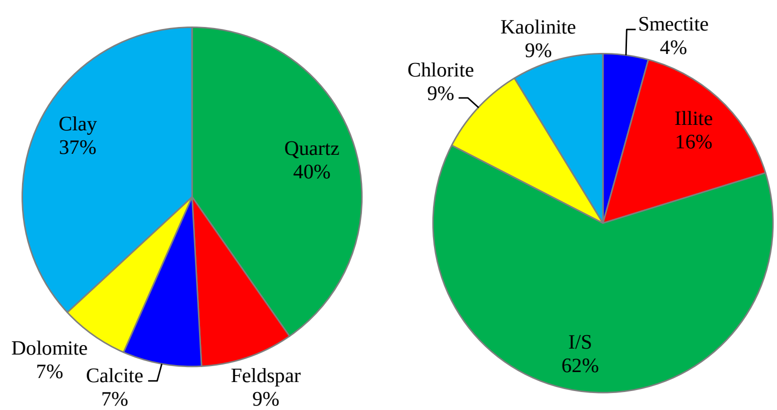

Figure 1. The mineral composition of shale sample.

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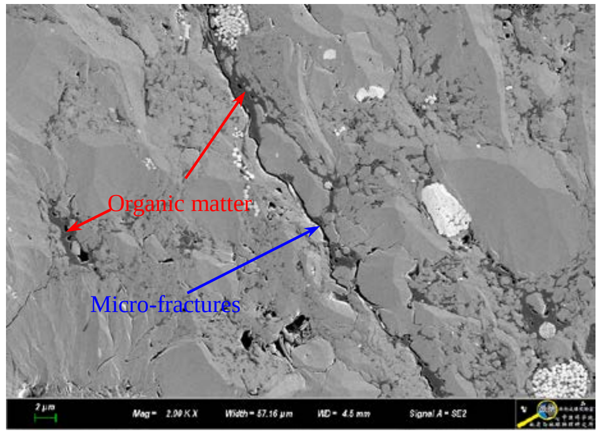

Figure 2. Scanning electron image of shale samples.

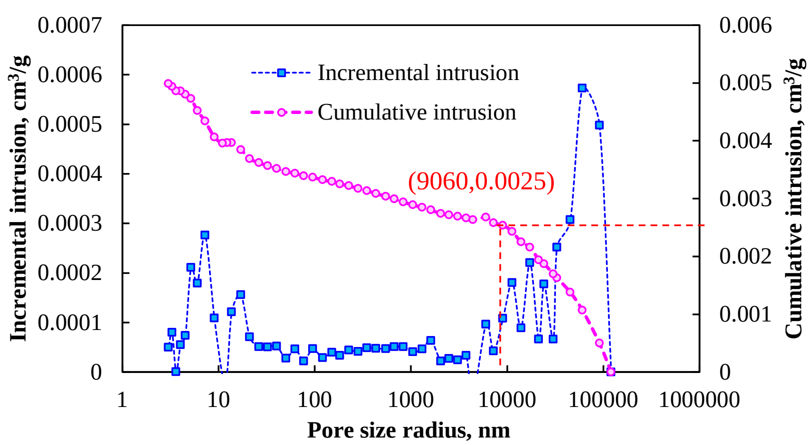

Figure 3. The pore size distribution derived from mercury injection test.

The pore size distribution is derived from mercury injection test (Figure 3). It ben seen that shale has a wide range of distribution. The macropores, mesopores and micropores are developed. In addition, the large pores (>10 μ m) are well developed which can be explained by the micro-fractures embedded in the matrix (Figure 2). It indicates that mean pore radius is about 9.06 μ m.

2.2. Apparatus

As the imbibition volume is small, high precision electronic balance should be adopted to measure the quality change of shale sample. In this experiment, the author adopts the Mettler XPE205 analytical balance with precision of 0.00001g (Figure 4a). The diffusion of saline ions in shale sample increases the water salinity. The fluid salinity can be evaluated by testing the fluid conductivity. The conductivity meter is Mettler Toledo S470 with the precision of 0.1μS/cm (Figure 4b).

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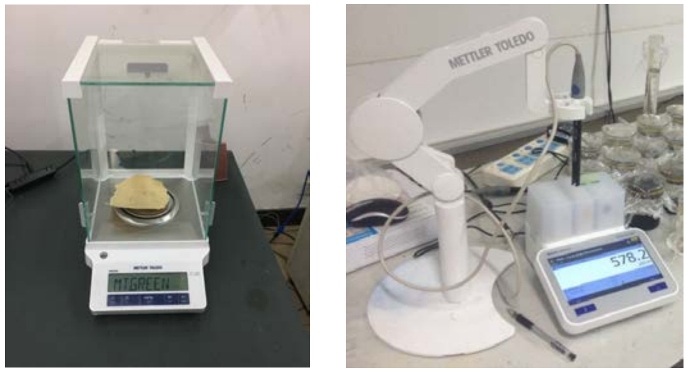

(a) *(b)*

Figure 4. The apparatus for experiments: (a) analytical balance, (b) electrical conductivity.

2.3. Experimental procedure

The experimental procedure includes the following steps:

(1) The samples are dried under the temperature of 105℃.

(2) The epoxy resin is used to seal the sample surface, with only one face contacting with water.

(3) The sample is placed in the beaker with 200ml water, and the preservative film is used to seal up the beaker to reduce evaporation (Figure 5).

(4) Test fluid conductivity after a certain time; test the quality of sample without water left on the surface (Figure 6).

(5) Repeat Step 4, the curves of conductivity and imbibed volume vs. soaking time can be obtained.

Figure 5. The shale samples in the 250ml beaker.

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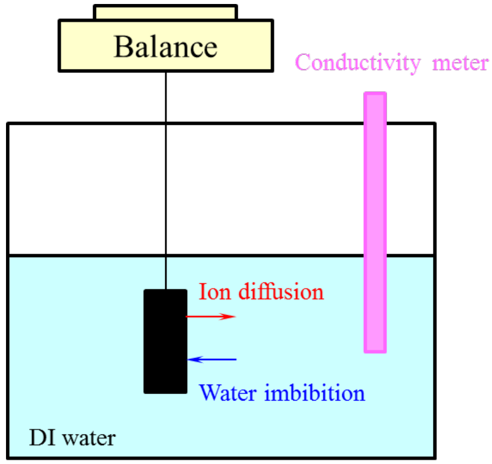
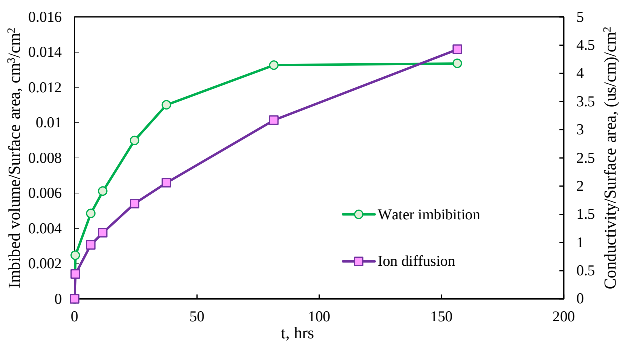


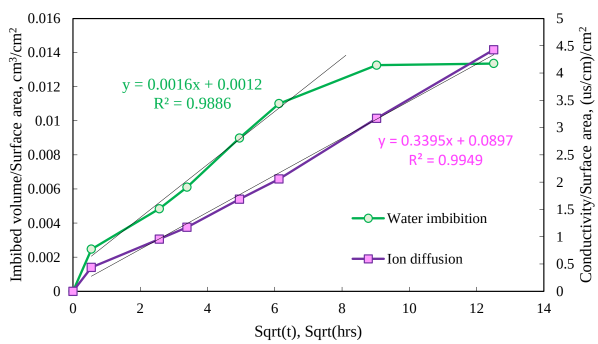
Figure 6. The schematic of experimental procedure.

3. RESULTS

The imbibition and diffusion curves are shown in Figure 7(a). The imbibed volume and fluid conductivity increase with time, illustrating that water enters into shale sample by imbibition, and the saline ions diffuse into fluid (Ballard et al., 1994). As time goes by, the curves tend to be smooth, suggesting that imbibition and ion diffusion get into equilibrium state. Besides, the imbibition and ion diffusion curves are different from each other. Compared to imbibition curve, ion diffusion has better duration (Dehghanpour et al., 2013).



(a)



(b)

Figure 7. The water imbibition and ion diffusion curves: (a) t, (b) $t^{0.5}$ (Modified from Yang et al., 2017).

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In Figure 7(b), there is a linear relationship between imbibed volume and soaking time. The conductivity curves have similar characteristics. The slopes of the curves are imbibition rate and ion diffusion rate, which are in consistent with the conclusions of Eq. 5 and Eq. 7. It is observed that the imbibition and ion diffusion happen synchronously, but they don't stop simultaneously. By the calculation, the imbibition rate of shale is 0.0016 cm/hrs0.5, while ion diffusion rate is 0.3395 (μS/cm)/(cm2· hrs$^{0.5}$). Besides, the ion adhesion amount in unit surface area C/V is 0.0897 (μS/cm)/cm2. Thus, the average pore size is

$$r = 4 \cdot \frac{A}{D} \cdot \frac{C}{V} = 4 \times \frac{0.0016}{0.3395} \times 0.0897 = 16.9 \mu m$$

This is close to the value of mercury-injection test. It proves that the new method, which is useful to evaluate the mean pore size, is developed using the experimental data of water imbibition and ion diffusion in gas-saturated shale. It is worth noting that there is deviation between new method and mercury-injection method. It can be explained by the complicated pore structure of shale. The capillary bundle model couldn't accurately describe the shale pore structure. In the future work, the authors will research the shale pore structure model to improve this method's accuracy.

4. CONCLUSIONS

The imbibition-diffusion experiment was conducted on organic-rich shale samples, and a mathematical model was built to evaluate the mean pore size of shale. The conclusions include:

- The experimental data of water imbibition and ion diffusion can be used to evaluate the mean pore size of shale. It proves to be correct.

- The experiment results demonstrate that water imbibition and ion diffusion start synchronously, while they do not stop simultaneously. The ion diffusion has longer duration time.

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