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Research Article

# Reasons for the low flowback rates of fracturing fluids in marine shale☆

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## Abstract

In this paper, marine shale cores taken from Zhaotong, Changning and Weiyuan Blocks in South China were used as samples to investigate the interaction between fracturing fluids and shale and the retention mechanisms. Firstly, adsorption, swelling, dissolution pore, dissolution fluid mineralization degree and ionic composition were experimentally studied to reveal the occurrence of water in shale and the reason for a high mineralization degree. Then, the mechanisms of water retention and mineralization degree increase were simulated and calculated. The scanning electron microscopy (SEM) analysis shows that there are a large number of micro fractures originated from clay minerals in the shale. Mineral dissolution rates of shale immersed in ultrasonic is around 0.5–0.7%. The ionic composition is in accordance with that of formation water. The clay minerals in core samples are mainly composed of chlorites and illites with a small amount of illites/smectites, but no montmorillonites (SS), and its content is between 18% and 20%. It is verified by XRD and infrared spectroscopy that the fracturing fluid doesn't flow into the space between clay mineral layers, so it can't lead to shale swelling. Thus, the retention of fracturing fluids is mainly caused by the adsorption at the surface of the newly fractured micro fractures in shale in a mode of successive permeation, and its adsorptive saturation rates is proportional to the pore diameters. It is concluded that the step-by-step extraction of fracturing fluids to shale and the repulsion of nano-cracks to ion are the main reasons for the abrupt increase of mineralization degree in the late stage of flowing back. In addition, the liquid carrying effect of methane during the formation of a gas reservoir is also a possible reason. Based on the experimental and field data, fracturing fluid flowback rates and gas production rates of 9 wells were analyzed. It is indicated that the same block follows an overall trend, namely, the lower the flowback rates, the more developed the micro fractures, the better the volume simulation effect and the higher the gas production rates.



*Keywords:* Shale; Fracturing fluid; Core; Adsorption; Flowback rate; Mineralization degree; Origin; Marine shale in south China; Gas production rate

Fracturing is a conventional method applied to volumetrically stimulate a shale reservoir which is tight and highly heterogeneous. However, it delivers a low flowback rate – generally 10–60% [1–3], while a large amount of fracturing fluid is retained in the reservoir. Chen et al. [4], based on an analysis of the Longmaxi Fm shale in the Zhaotong block, suggested that reservoir over-pressure in the area is attributed to the pressure increase resulted from hydrocarbon generation and deep burial of organic-rich shale, basin-scale tectonic compression, erosional unloading, and tight sealing from surrounding rocks. Pu [5] analyzed the accumulation conditions of shale gas in the Sichuan Basin and believed that organic matters in this basin are dominated by Type II kerogen, and *TOC* ranges from 0.80% to 2.79%, representing moderate to good source rocks. Core analysis reveals a common presence of fractures in the Longmaxi Fm shale and abundant interlamellar nano-cracks. Feng et al. [6] suggested that methane is soluble in water in a nano-scale environment to form water-soluble gas, the interaction between methane and water causes decrease in hydrocarbon bond and, as a result, the gas diffusion coefficient would be increased

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significantly, favoring the diffusion of methane. An [7] suggested that, basically, there is no resistance to the migration of water-soluble gas. Therefore, investigating the occurrence state of operating fluid in shale and the mechanism of its interaction with shale therein, and discussing and determining the reasons for low flowback rates and gradual increase in the mineralization degree of backflow fluids provide significant guidance to ensuring high-efficiency shale gas production and selecting the proper type of fracturing fluids. The authors presented a study of shale in the southern Sichuan Basin with cores taken from the Weiyuan, Changning and Zhaotong blocks.

## 1. Instruments and research methods

### *1.1. Water adsorption and dissolution test*

Samples are hammered into pieces with an equivalent mass. Put a sample with an accurate weight of 10.0000 g ($m_1$) sinto a 100 mL penicillin bottle and add it with 60 mL liquid. The bottle is sealed and soaked in water under 80 °C for 1−7 days. Remove the surface moisture centrifugally, weigh the weight of shale ($m_2$) precisely, and then put the sample in an oven and dry for 5 h (at 105 °C) to obtain a constant weight ($m_3$).

$$A\ dissolution\ rate = \frac{m_1 - m_3}{m_1} \times 100 \quad (1)$$

$$A\ water\ adsorption\ rate = \frac{m_2 - m_3}{m_1} \times 100 \quad (2)$$

### *1.2. Clay-swelling XRD method*

After being soaked for 1, 3, 5, and 7 days, the samples are taken out and dried for 2 h at 105 °C. In the process, 1 nm peak change of the samples is observed with scanning angle of 7° to 10°.

### *1.3. SEM observation of shale features*

The cores are sliced and split apart, and the natural fracture surface is taken for observation.

## 2. Results and discussion

### *2.1. Clay composition and shale features*

Clay composition was measured in accordance with SY/T 5163-1995 and the measurement results are shown in Table 1. Samples were prepared with the method introduced in Section 1.3. Features of cores were observed under a 8000× microscope. The results are shown in Fig. 1.

A large number of clay interlamellar nano-cracks are present in cores taken from the Changning and Zhaotong blocks, which provide ideal sites for shale gas adsorption and free gas storage. In addition, some natural micro-fractures are also visible, which may serve as the main sites for free gas storage and migration. XRD test denies the presence of SS structure of smectites in cores.

### *2.2. Influences of soaking in different operating fluids on shale swelling*

Clay swelling is one of the concerns in stimulation with water-based fracturing fluid. Cores taken from Well ZT104 were used as samples and treated in accordance with the method introduced in Section 1.2. A low-angle XRD scanning (5°−10°) was conducted for shale soaked for 1, 3, 5, and 7 days. Figs. 2 and 3 illustrate the resulted XRD correlation diagrams. There is no evident adsorption peak at 1.7 nm within the 5°−6° range. It can be therefore determined that, there is no SS interlamellar pair of montmorillonite in shale. I/S correlation is made only at 1 nm in the figure.

Figs. 2 and 3 indicate that, after 7 days of soaking in discharge-aiding agent (i.e., negative ion surfactant) and slickwater (i.e., organic polymer), there is no regular change in 1 nm peak width or peak height of clay minerals. The curves are basically overlapped with each other, which means that there is no hydrous swelling occurred.

### *2.3. Adsorption and dissolution of operating fluids to shale*

Shale, in general, is hydrophilic. Water adsorption at the contact surface will inevitably occur when fractures are created with water-based fracturing fluids. The adsorption speed and rate have a great influence on flowback. Since the shale in the southern Sichuan Basin was marine-deposited, a relatively high degree of mineralization can be expected [8]. In case shale gas migrates together with water during the diagenesis and gas-generating process [9], is it possible for soluble salt crystals to exist in shale? In case shale contains a large quantity of soluble salts, fracturing fluids will lead to the increase in mineralization degree and porosity of shale. The mechanism of microscopic interaction between the operating fluids and the shale was studied by observing the concentration of the dissolved ions of shale samples (i.e., cores taken from Well CH201), soaked in a variety of solutions: distilled water,

Table 1
Clay composition of shale.

| Sample no. | Source well | Clay content | Remarks |
|---|---|---|---|
| 1 | ZT103 | 35.8% | Predominately illites, partially illites/smectites |
| 2 | ZT104 | 35.4% | 39.06% chlorite, 46.50% illites, 14.44% illites/smectites, 15−20% interlamination rate |
| 3 | CH201 | 22.5% | Predominately illites, partially illites/smectites |