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Experimental and numerical study on the relationship between water imbibition and salt ion diffusion in fractured shale reservoirs

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Field observations demonstrate that shale gas wells feature a low flowback efficiency (<30%) and high-salinity flowback water (approximately 200kppm) after multistage hydraulic fracturing operations. The water recovery and salinity profile could be regarded as a critical method for volumetric and chemical analyses to characterize the reservoir properties and complexity of the fractured network. This paper aims to understand the relationship between fracturing imbibition and ion diffusion, which are responsible for inefficient water recovery and high-salinity flowback fluid, respectively. Comparative imbibition experiments are performed on different shale and sandstone samples, and an electrical conductivity meter is used to monitor the change in ion concentration change of the imbibition fluid. A mathematical model based on theoretical analysis is proposed to clarify the correlation between imbibition and ion diffusion. Both the experimental and analytical solution results show that the imbibition fluid conductivity resulting from ion diffusion is proportional to the square root of time, which is similar to the law of capillary-driven imbibition into porous media. Water imbibition into gas shale and ion diffusion into water proceed simultaneously in the opposite direction, and only the imbibition front contacting the pore wall with salt ions can cause the salt ions to dissolve and diffuse into water. The analytical solution results also indicate that the effects of the porosity, surface tension, contacting area and wetting angle on the water imbibition rate are in consistent with that of the ion diffusion rate. The permeability, however, shows a positive correlation with the imbibition rate and a negative correlation with the ion diffusion rate. The initial water saturation is negatively related to the imbibition rate, and positively related to the ion diffusion rate. In addition, smectite and I/S could enhance the imbibition and diffusion rates. It is observed that illite concentration has no relationship with the imbibition and diffusion rates, indicating that illite minerals do not significantly affect the imbibition/diffusion rate in these clay-rich shales. This research contributes to understanding the correlation between imbibition and ion diffusion, which is significant for flow-back analysis after fracturing operations.

1. Introduction

Multistage fracturing is a key technology for the economic exploration of shale gas (Novlesky et al., 2011). After a large volume of slick water is injected into a shale formation, the flowback efficiency is generally lower than 30%, and even lower than 5% in the Haynesville shale reservoir (Penny et al., 2006; King, 2012). Meanwhile, the concentration of salt ions in flowback water increases with time (Ghanbari and Dehghanpour). The salinity in shale gas wells reaches 200kppm in the Horn River Basin (Blaunch et al., 2009; Pritz and Kirby, 2011). The volumetric analysis based on flowback efficiency and chemical analysis based on the salinity of flowback water are of great significance to understanding the

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reservoir properties and evaluating the characteristics of artificial fracture networks.

Recent studies show that the fracturing fluid imbibition into the shale matrix is the key reason for low flowback efficiency. Published studies have focused on the fracturing fluid imbibition mechanism in gas shale. The driving force of imbibition in a conventional reservoir is capillary pressure, while both capillary pressure and osmotic pressure could drive the water into the shale matrix (Dehghanpour et al., 2013; Yang et al., 2016). In clay-rich shale, the osmotic pressure is more powerful than the capillary pressure, and the water volume imbibed into the matrix therefore significantly surpasses the pore volume measured by gas (Ge et al., 2015). Moreover, the intense imbibition effect in gas shale can enhance the pore pressure and then induce the generation of tensile fractures (Yang et al., 2015; Junjian and Sheik, 2015). The pore structure in shale is more complicated than that in conventional sandstone, which causes the special imbibition characteristics in gas shale. As for the dual-porosity nature of shale, shale exhibits a distinct transition from a higher imbibition rate to a lower imbibition rate (Roychaudhuri et al., 2013). The low imbibition rate suggests that the matrix reflects a relatively low pore connectivity depicted by the slope in the curve [log (cumulative imbibition) vs. log (imbibition time)]. Hu et al. (2012) proposed that the matrix in Barnett shale presents imbibition behavior of 0.26, indicating the low pore connectivity.

Some studies concluded that the salt concentration in flowback water is related to the diffusion of salt ions into the injected water (Haluszczak et al., 2013; Ghanbari and Dehghanpour, 2015). Keller and Liovando (1989) proposed that the electrical conductivity of the produced water increases with time, demonstrating that salt ions of shale dissolve and diffuse into water. The ion diffusion rate in the direction parallel to the bedding plane is faster than that vertical to the bedding plane (Ghanbari et al., 2013). In addition, clay minerals have a great influence on ion diffusion. The charged clay interlayers have the property of a semipermeable membrane, which cannot restrict water molecule movement but does restrict ion movement (Mitchell and Moench, 1993). Many researchers argued that the ion diffusion into water follows Fick's diffusion law (Treybal, 1980; Ghanbari and Dehghanpour, 2015). Nevertheless, Knudsen's diffusion law rather than Fick's diffusion law may be more reliable for ion diffusion in nanopores of gas shale as Fick's law cannot well explain the collision between the ions and the nanopore walls.

The physical and chemical flowback data could be used to characterize the reservoir and artificial fracture network. Fan et al. (2010) stated that a more complex fracture network could result in lower water recovery. As the important indicator for distinguishing the formation water and fracturing fluid, the ion content and type in flowback water should be given much attention (Asadi et al., 2008). The chemical signature in the water recovered from induced fractures is different from that recovered from reactivated secondary fractures (Bearinger, 2013). In addition, the architecture of the induced fracture network has effects on the shape of salt concentration profiles during the flowback operations, and the concentration of dissolved salt is positively related to the surface area of the fracture network.

As for the investigations into the relationship between imbibition and ion diffusion, Ballard et al. (1994) and Zolfaghari et al. (2014) proposed that the ion diffusion rate is similar to the imbibition rate, depending on permeability, porosity, clay content and contact surface area. Ghanbari et al. (2013) conducted imbibition/diffusion experiments and found that the behaviors of imbibition curves are well correlated to that of diffusion curves. In spite of these recent studies, two major questions still remain: (1) What is the reason for the similar behaviors of imbibition and diffusion curves? (2) How do we set up the mathematical model for the quantitative interpretation of the imbibition/diffusion relationship? The aim of this paper is to extend the previous investigations and answer these questions.

2. Experiments

2.1. Materials

The shale samples are selected from typical shale formations in China, and the conventional sandstone reservoir samples are collected for comparison. The geological information of different formations can be found in Table 1. It should be noted that the shale formations (i.e., Lujiaping, Longmaxi, Niutitang and Xujiahe formation) in China's Sichuan Basin have the greatest potential for shale gas production. In particular, a large number of breakthroughs along the path to commercial exploitation have been realized. The Ganchaigou formation of Qaidam Basin, known to be clay-rich, is provided as a calibration standard.

The average mineralogy content of the formations is shown in Table 2. The mineral compositions are obtained by D/MAX 2500X X-ray diffractometer following the testing standard of SY/T5163-2010. The measurement temperature and relative humility are 20 °C and 40%. The minerals of the shale formations are characterized by high content of smectite + I/S (14–33%) and quartz (36–55%). The total concentration of clay minerals ranges from 23.7% to 55%. In addition, the total concentration of clay in continental environment shale is much larger than that in marine shale (Ji et al., 2014).

Pictures of the samples are shown in Fig. 1. It is demonstrated that the shale is black and gray and composed of small particles that are obviously different from sandstone. Twelve samples are used to conduct comparative imbibition/diffusion experiments. The physical parameters of the samples are shown in Table 3. Due to the different brittleness of the core materials, not all the samples can be drilled into cylindrical plugs. Some samples must be machined into rectangular blocks by cutting. In addition, one-end-open (OEO), two-ends-open (TEO) and all-faces-open (AFO) are common boundary conditions for imbibition experiments (Kim and Kovscek, 2014). OEO means that only one end face is open for imbibition and epoxy is used to keep other faces impermeable. OEO and TEO are obtained to conduct one-dimensional imbibition experiments. The fluid used in the experiments is distilled water with an electrical conductivity of 2.3 uS/cm. The fracturing fluid adopted during the multistage fracturing operations is slick water, which contains a small amount of friction reducing agents. Therefore, the adoption of distilled water is able to meet the application requirements.

2.2. Experimental procedure

Spontaneous imbibition refers to the process by which water is imbibed into shale spontaneously under the joint actions of capillary pressure and clay osmotic pressure (Dehghanpour et al., 2013). Because of the relatively low imbibition volume in shale, a higher accurate analytical balance is adopted to measure the shale mass variation, which is the Mettler XPE205 analytical balance with a precision of 0.00001 g (Fig. 2(a)). Ion diffusion refers to the process by which salt ions in shale matrix pores diffuse into water under the influence of concentration differences. As the concentration of total dissolved solids increases, the electrical conductivity of water rises linearly (1 mg/L = 2 uS/cm). The ion diffusion can be evaluated by the conductivity meter. The Mettler Toledo S470 electrical conductivity meter is used in this study. The accuracy is 0.1 μS/cm, and the measurement range is 2000 ms/cm.

The test procedure for imbibition/diffusion experiments