Experimental study on hydration damage mechanism of shale from the Longmaxi Formation in southern Sichuan Basin, China

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Abstract
As a serious problem in drilling operation, wellbore instability restricts efficient development of shale gas. The interaction between the drilling fluid and shale with hydration swelling property would have impact on the generation and propagation mechanism of cracks in shale formation, leading to wellbore instability. In order to investigate the influence of the hydration swelling on the crack propagation, mineral components and physicochemical properties of shale from the Lower Silurian Longmaxi Formation (LF) were investigated by using the XRD analysis, cation exchange capabilities (CEC) analysis, and SEM observation, and we researched the hydration mechanism of LF shale. Results show that quartz and clay mineral are dominated in mineral composition, and illite content averaged 67% in clay mineral. Meanwhile, CEC of the LF shale are 94.4 mmol/kg. The process of water intruding inside shale along microcracks was able to be observed through high power microscope, meanwhile, the hydration swelling stress would concentrate at the crack tip. The microcracks would propagate, bifurcate and connect with each other, with increase of water immersing time, and it would ultimately develop into macro-fracture. Moreover, the macrocracks extend and coalesce along the bedding, resulting in the rock failure into blocks. Hydration swelling is one of the major causes that lead to wellbore instability of the LF shale, and therefore improving sealing capacity and inhibition of drilling fluid system is an effective measure to stabilize a borehole.

1. Introduction
The United States Energy Information Administration (EIA) released ‘Technically Recoverable Shale Oil and Shale Gas Resources: An Assessment of 137 Shale Formations in 41 Countries Outside the United States’ in which indicated that the total resource of technically recoverable shale gas is 220.73 × 1012 m3, and it suggested that shale gas has rich and huge development potential [1]. Shale gas, as a kind of relatively clean energy, has drawn wide attention from the oil companies; however, wellbore instability in a shale formation has been the technology bottleneck for developing shale gas in a safety and high efficient way [2–4]. When drilling mud intrudes into shale formation and interacts with the clay, shale would produce expansion stress, resulting in reduction of rock cohesion [5–8]. Meanwhile, because of mud filtrate invading formation along bedding plane and micro-fracture, hydration expansion of clay occurs and generates hydration stress, which concentrates on crack tip and leads to crack growth and broadening [9,10]. The internal structure damage of rock would cause rock spalling along bedding plane, and forms fracture surface, the secondary section and rock pieces in various sizes and shape [11]. Therefore, studying the mechanism of formation and propagation of cracks is significant to wellbore stability. Similar studies on the hydration damage of shale have been done by other researchers. Ma researched the micro-damage characteristics of shale hydration based on CT scanning technology, and CT images showed that the micro-damage of shale hydration mainly occurred in the early
stage of soaking, which was the initial stage and rapid evolution stage of micro-damage [12]. Wang revealed the internal micro-structure of the same sample at different steeping times, and the hydration damage process of rocks was shown dynamically [13]. Shi proposed a new approach to reveal the effect of hydration on crack development and analyzed hydration mechanism and effects on the borehole wall instability through scanning electron microscope [14]. Yang approached quantitative methods for crack propagation by using geochemical kinetics of mineral-water interactions [15].

Changning in south Sichuan is the state demonstration of shale gas exploitation, and it plays a key role in the development of shale gas. However, complex accidents of borehole such as stick and well leakage were usually happened during drilling process, which seriously affect the progress of exploration and production of shale gas in Changning. Therefore, to investigate problem of wellbore instability in Longmaxi formation, a series of experiments were carried out to analyze the physicochemical properties of LF shale, such as mineralogical compositions, cation exchange capability (CEC) and microstructure. By immersion experiment, shale damage caused by hydration was researched. At last the mechanism of formation and propagation of crack was analyzed, which can provide theoretic support for optimization of drilling fluid system and wellbore stability.

2. Samples and methods

2.1. Samples

Shale were obtained from the Lower Silurian Longmaxi Formation in the Sichuan Basin of China where is located at N28°23′52.6′′, E104°52′24.2′′. XRD analysis, CEC analysis, and SEM observation, sampling, breaking and sieving were conducted in accordance with national standards ‘Analysis method for clay minerals and ordinary non-clay minerals in sedimentary rocks by the X-ray diffraction’ and ‘Methods for testing shale physics and chemistry properties’, after which samples were analyzed with a relatively complete experimental program, including the XRD analysis, CEC analysis, cracks propagation analysis. 9 XRD results and 9 CEC results of samples from the Changning area have been analyzed in order to make the results more accurate and reliable. In addition, researchers also collected samples from the Changning area for SEM observation and hydration test.

2.2. Experimental methods

Samples were crushed to 100 mesh size grains for XRD analysis to ensure particles are smaller than 10 mm in diameter, which was conducted with an X’Pert PRO. Mineralogical composition and relative mineral percentage of the samples were estimated following the Chinese Oil and Gas Industry Standard Sy/T5983-1994 and Sy/T5163-1995.

Shale samples were cut into thin sections with a diameter of 25 mm and a thickness of 3 mm and cylinders with a diameter of 25 mm and a height of 50 mm for the purpose that we analyzed the damage mechanism of shale starting with the initiation of crack and end with failure of rock. The end surfaces of thin sections and cylinders should be polished to be even and smooth and we observes that crack initiation and formation of thin sections and how hydration makes rock broke. As shown in Fig. 1, a thin is placed on a glass slide, and then water is drip at the edge of thin. After half a minute, we observed the structural changes of thin with a polarized microscope and collected pictures about shale cracking.

3. Results and discussion

3.1. Mineralogical compositions

The XRD analysis results of the LF shale samples are shown in Tables 1 and 2. Table 1 indicates that the LF shales are mainly composed of clay, quartz and calcite; in contrast, pyrite and feldspar are minor. From Table 1, we can observe that the clay minerals contents of the LF shale samples range from 34.56% to 59.34% with an average of 44.61%; the quartz contents range from 19.89% to 30.6% with an average of 24.98%; the calcite contents range from 6.34% to 29.17% with an average of 16.53%; the dolomite contents range from 2.52% to 14.61% with an average of 6.89%. The main clay mineral in LF shale is illite with the content of more than 65%, chlorite second and it does not contain swelling montmorillonite.

The interaction between clay and water would generate hydration stress which causes rock cohesion to decrease. Hydration of illite occurs on the surfaces of particles; and illite–smectite mixed-layer mineral has expansibility ranging between illite and smectite.

3.2. CEC

The CEC of the shale is directly related to the adsorption capacity of water molecules and surface hydration, which reflects...
the hydration expansion of shale. Therefore, CEC analysis was used to investigate the hydration expansion capacity of the shale. The CEC results of the LF shale in are shown in Fig. 2. The CEC ranges from 85 mmol/kg to -105 mmol/kg with an average of 94.4 mmol/kg as shown in Fig. 2. In order to analyze the expansibility of LF shale from the CEC perspective, we compare CEC of the LF shale when contact with water. LF shale’s ability of dispersity is between the Yimin shale and the Silurian shale. Therefore, the Yimin shale has stronger dispersity than the Silurian shale’s and lower than the Yimin shale’s.

Table 1
The mineral composition of LF shale samples.

<table>
<thead>
<tr>
<th>Number</th>
<th>Clay</th>
<th>Pyrite</th>
<th>Quartz</th>
<th>Orthoclase</th>
<th>Plagioclase</th>
<th>Calcite</th>
<th>Dolomite</th>
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<tr>
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<td>19.89</td>
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<td>28.92</td>
<td>0</td>
<td>4.96</td>
<td>7.16</td>
<td>2.98</td>
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<td>3.49</td>
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<td>1.55</td>
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<td>4.15</td>
<td>6.34</td>
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<td>1.01</td>
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<td>27.95</td>
<td>8.75</td>
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<tr>
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<td>22.85</td>
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<td>1.29</td>
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<td>3.62</td>
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<td>9</td>
<td>59.34</td>
<td>1.17</td>
<td>26.48</td>
<td>0</td>
<td>3.21</td>
<td>7.16</td>
<td>2.65</td>
</tr>
</tbody>
</table>

Table 2
The clay composition of LF shale samples.

<table>
<thead>
<tr>
<th>Number</th>
<th>Clay composition/%</th>
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<tr>
<td>1</td>
<td>I 67.5, Il/S 8.4, C 24.1</td>
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<tr>
<td>2</td>
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<td>4</td>
<td>I 70.7, Il/S 5.6, C 23.7</td>
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<td>6</td>
<td>I 74.4, Il/S 2.5, C 23</td>
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<td>7</td>
<td>I 73.7, Il/S 2.5, C 23.8</td>
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<tr>
<td>8</td>
<td>I 73.6, Il/S 4.8, C 21.5</td>
</tr>
<tr>
<td>9</td>
<td>I 72.6, Il/S 6, C 21.5</td>
</tr>
</tbody>
</table>

The SEM image of the microstructure of the LF shale before and after soaked in water is shown in Figs. 3 and 4. From Fig. 3 the original SEM image indicates that the LF shale exists tiny fractures, and it is observed clearly that flaky clay mineral particles ranging tightly are aligned in the direction of bedding and tiny fracture is perpendicular to bedding plane when the shale structure is magnified to 2000 times as shown in Fig. 3-b,d,e,f. Because of the self-absorption of capillary effect, water would invade rock along those tiny fractures where hydration occurs. The SEM image of micro fracture surfaces caused by hydration is shown in Fig. 4. Compared with its original microstructures, flaky clay mineral particles after soaked in water evolved from compact to dispersive, since the edge of flake clay occurs passivation. In addition, hydration would make clay minerals expand, and the expansion amount would increase with steeping time, which would lead to flake clay minerals to thicken, and the phenomenon becomes more obvious at the edge of flake clay.

3.4. Immersion experiment

Water would intrude in rock along micro fractures, pore and beddings as rock is immersed in water which damages rock structure. We used a high-powered microscope to investigate the damage caused by hydration. The damage morphology of shale caused by hydration is shown in Fig. 5. Water initially invades into rock along the original microcracks because of hydrophilic and self-absorption. Hydration stress and would concentrate at the crack tip which would prompt cracks to propagate forwards and grow secondary cracks. Those cracks continuously propagate ahead and gradually widen until clay particles completely separate from each other, and the fractures form. Meanwhile, the edge of fractures is wetted by water and grows secondary cracks which also continuously propagate ahead and gradually widen. The cracks including secondary cracks will spread to other place. Those cracks will connect and link up until the rock become broken. From the micro morphology of fracture (Figs. 6 and 7), it can be observed that the fracture is sufficiently characterized by tension fracture. The boundary of fracture is similar with each other, and some mineral grains at the fracture boundary still connect with other. The phenomenon indicates that extensional stress concentrates at the tip of fracture, which results in growth and propagation of it. In shale rock, with increase of steeping time, a large scale of isolated fine cracks would continuously propagate and gradually widen and connect and link up with each other, eventually, impair structural integrity of rock.

After observing the process that cracks initiate, propagate, fork, merge under a microscope, we cut core sample from the direction perpendicular to the bedding in the same outcrop in order to analyze macro morphology and distribution of cracks caused by hydration when rock has been soaked in water for some time. The comparative observation of cylinder cores before and after soaked in water is shown in Fig. 8. Fig. 8-a is picture of the core sample before soaked, and Fig. 8-b, c and d are pictures of the core sample from different direction after soaked.

As shown in Fig. 8, instead of being softened or dispersed, the core generated lots of macro-cracks. With water intruding inside,
the crack initiation occurs at micro-fractures, and the cracks continuously propagate ahead and gradually widen until clay particles adhesive force which promotes macro-fractures to form. The macro-fractures connect with each other and generate complicated network of rock fractures, which results in rock broken. The most of macro-fractures are parallel to beddings approximately and the surface of macro-fractures is rough and irregular, which leads to rock break. A few macro-fractures grow along other direction instead of bedding, and it not only decreases the integrity of rock but it also arouses sloughing. Through immersion test, the characteristic of the hydration of LF shale is that fracture network caused by hydration makes shale lose structural continuity and break.

3.5. Damage mechanism analysis

Lots of microcracks and bedding are developed in LF shale, and provide paths for water intruding into shale, space for hydration. Because of the good hydrophilicity of shale, water intrudes into shale along the microcracks and bedding under the capillary force, and the hydration occurs concurrently. Microcracks extend around the weak position and become longer cracks under the driving force of hydration. It is the first damage that the original microstructure gets changed, which would promote water to intrude into shale. Hydration occurs at the surface of clay particles; however, type and content of clay play a crucial role in degree of hydration.
The XRD analysis results indicate that the clay mainly consists of illite and a small quantity of chlorite and illite-smectite mixed-layer mineral, without expanding montmorillonite. Illite is a clay-sized, micaceous mineral, and its structure is constituted by the repetition of tetrahedron – octahedron – tetrahedron (TOT) layers. Illite crystal would generate negative charges when amorphous substitution takes place, which is balanced by cation like Na\(^+\), K\(^+\), and Ca\(^{2+}\) outside the interlayer space of illite. Hydration of the cation makes water molecules aggregate around the surface of cation. Distance between lamellas was enlarged by increasing water molecules. Illite/smectite (I/S) is the mineral that smectite transfers into illite and forms into honeycomb structures, and its hydration ability is between illite and smectite.

Because illite and I/S have the characteristic of flaky, it would present two damages to shale that polar water molecule invades into layers of slices. On the one hand, thickened water membrane results in the production of swelling stress. On the other hand, water erodes and dissolves the cement between rock grains and weakens connection force, which makes cohesion decrease. So that the fracture toughness of shale would decrease after water invades into rock along microcracks and pore [14]. When the stress intensity factor on the crack tip generated by hydration expansion stress is over its fracture toughness, microcracks would stretch and widen, and communicate with other microcracks which damages partial microstructure. The hydration expansion stress provides a constant source of motivation for the formation and propagation of microcracks. Microcracks continuously propagate ahead and generate secondary fracture under the driving forces of hydration, which lead to the failure of rock.

To further analyze the damage mechanism of hydration, a propagation model of shale cracking was established with the hydration swelling stress. We suppose that there is an oval crack with 2a in the longer axis. Whether the crack stretches is determined by the in-situ stresses and hydration stress, and Fig. 9 shows the distribution of these stresses. Stress intensity factor caused by the in-situ stresses and hydration stress at the crack tip is solved using the superposition principle:

\[
K_I = K'_I + K''_I
\]

Where \(K_I\) is stress intensity factor at the crack tip, MPa m\(^{1/2}\); \(K'_I\) is stress intensity factor caused by hydration stress, MPa m\(^{1/2}\); \(K''_I\) is stress intensity factor caused by in-situ stresses, MPa m\(^{1/2}\).

Water invades shale resulting in strength reduction of shale and hydration stress concentrating at crack tip. The expansion caused by hydration is small, but the hydration stress is large and
can't be ignored. The hydration stress is uniformly and symmetrically distributed on the crack surface, and the length of the hydration stress acting on the crack surface is $b$, as shown in Fig. 10. The stress intensity factor caused by hydration stress can be expressed as follows:

$$K_0 = 2P \sqrt{\frac{a}{\pi}} \arcsin \left( \frac{b}{a} \right)$$

(3)

Where $P$ is hydration stress, MPa; $a$ is the semi-axis of the oval crack, m; $b$ is the length of the hydration stress acting on the crack surface, m.

Shale cracking is also affected by the in-situ stresses, and the normal stress on the crack surface is calculated from the Fairhurst equation [18]. The stress intensity factor caused by in-situ stresses can be expressed as follows:

$$K' = \sigma \sqrt{\pi a}$$

(4)

Where $\sigma$ is the normal stress on the crack surface, it can obtained by Fairhurst equation, MPa.

$K_0$ (stress intensity factor at the crack tip) is composed of $K'_I$ (stress intensity factor caused by hydration stress) and $K'_Y$ (stress intensity factor caused by in-situ stresses). When stress intensity factor is greater than Fracture toughness of shale, the crack would stretch and widen, and communicates with other microcracks.

4. Conclusion

The LF shale is mainly composed of quartz, clay and calcite, and contains a small quantity of pyrite and feldspar. The main clay mineral is illite, and a few of I/S exists in it. Clay minerals are directionally arranged, and bedding is well developed in the macroscopic. There are large scale of micro-crocks and porosity. Microcracks and the clay are the necessary prerequisite to hydration. The LF shale has high clay content, produces lots of bedding and porosity so that hydration is strong. The water invasion has two effects: the first aspect is that water erodes and dissolves the cement resulting in the cohesion and the fracture toughness of shale degradation; the second aspect is that hydration expansion stress and capillary pressure would concentrate at cracks tip leading to stress intensity factor at the crack tip increase. When the stress intensity factor at the crack tip is more than the fracture toughness of shale, original and secondary microcracks expand and extend, which finally result in the macroscopical perforation failure of rock. Improving sealing capacity and inhibition of drilling fluid system is an effective measure to prevent water intruding rock and keep wellbore stable.

Seal compactness materials would enter into the micro-fractures under pressure, and a layer of tight seal would be formed. As a result, the seal compactness materials prevent the liquid of drill fluid from entering into the micro-fractures. So these seal compactness materials are added into the drilling fluid to improve the sealing capacity of fluid system. Clay is a mineral composition with hydration property, but hydrate inhibitor, such as organic amine inhibitor, can keep clay from hydrating so that shale can remain intact. In conclusion, seal compactness materials and hydrate inhibitor are added into drilling fluid system, which can improve sealing capacity and inhibition of drilling fluid system to prevent shale cracking.
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References


