

## Research Article

# Experimental Investigation on Cracking Characteristics of Dry and Saturated Shales in Nitrogen Fracturing after Liquid Nitrogen (LN<sub>2</sub>) Injection

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Cryogenic LN<sub>2</sub> fracturing is one of the environmentally friendly waterless fracturing technologies that promote the fracture complexity of shale gas reservoir. The water-ice phase transition under freezing condition causes frost heave in saturated shale. The effect of moisture in shale should be taken into account during cryogenic damage process. Therefore, the differences of cracking characteristics between dry and saturated shales were studied in this paper. A laboratory triaxial and high temperature fracturing system was developed for nitrogen fracturing dry and saturated shale after LN<sub>2</sub> injection. The influence of moisture on breakdown pressure was studied under different confining pressures (3 MPa, 6 MPa, 9 MPa, and 12 MPa) and bedding directions (parallel bedding and vertical bedding). The experimental results demonstrated that when the confining pressure increased from 3 MPa to 12 MPa, the breakdown pressure of dry parallel bedding after LN<sub>2</sub> preconditioning decreased 7.12 MPa, 6.06 MPa, 4.58 MPa, and 3.11 MPa, respectively. Therefore, LN<sub>2</sub> preconditioning could damage shale resulting in a lower breakdown pressure, but the effect of cryogenic damage decreased with the confining pressure increasing. The moisture in shale had little impact on nitrogen fracturing without LN<sub>2</sub> injection because the breakdown pressure difference between dry and saturated shales was small. However, the breakdown pressure of saturated shale after LN<sub>2</sub> preconditioning was always lower than that of dry shale. The breakdown pressure of saturated parallel bedding shale after LN<sub>2</sub> injection decreased 8.62 MPa, 7.67 MPa, 6.08 MPa, and 4.63 MPa, respectively, with the confining pressure increasing from 3 MPa to 12 MPa. The breakdown pressure difference between dry and saturated shales was impacted by the migration of unfrozen water and frost heave. In addition, the extent of cryogenic damage varied substantially between different bedding directions. When the confining pressure was 3 MPa, the breakdown pressure of saturated parallel bedding shale reduced by 69.18% after LN<sub>2</sub> preconditioning, but that of saturated vertical bedding shale only decreased by 22.49%. The tensile strength of shale had a greater influence on the breakdown pressure. According to the Brazilian disc test results, the tensile strength of matrix was much higher than that of bedding planes. As a result, it is useful to wet the shale in order to reduce the breakdown pressure. The fracturing direction of horizontal drilling should be consistent with the bedding direction for better cryogenic fracturing effect.

## 1. Introduction

Shale gas is plentiful and extensively spread throughout China, which exceeds  $2.2 \times 10^{13} \text{ m}^3$  [1]. The production of shale gas increased rapidly from  $2 \times 10^8 \text{ m}^3$  in 2013 to

$1.5 \times 10^{10} \text{ m}^3$  in 2019 [2]. The exploitation of shale gas reservoirs has always been restricted by the ultralow permeability, significant in situ horizontal stress differential, enormous burial depth, and low brittleness index [3]. Hydraulic fracturing has been used to stimulate the unconventional gas

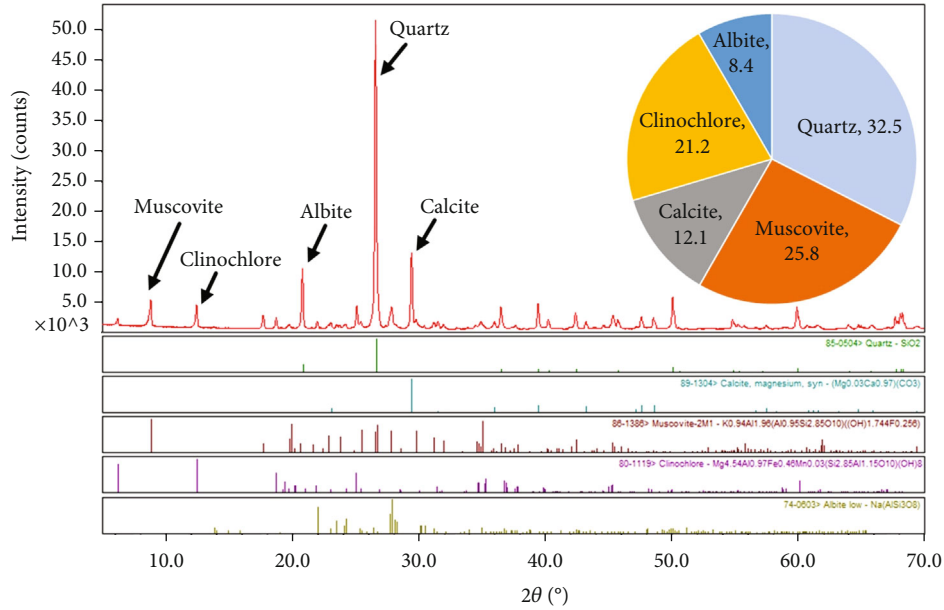


FIGURE 1: Mineralogy analysis of shale sample.

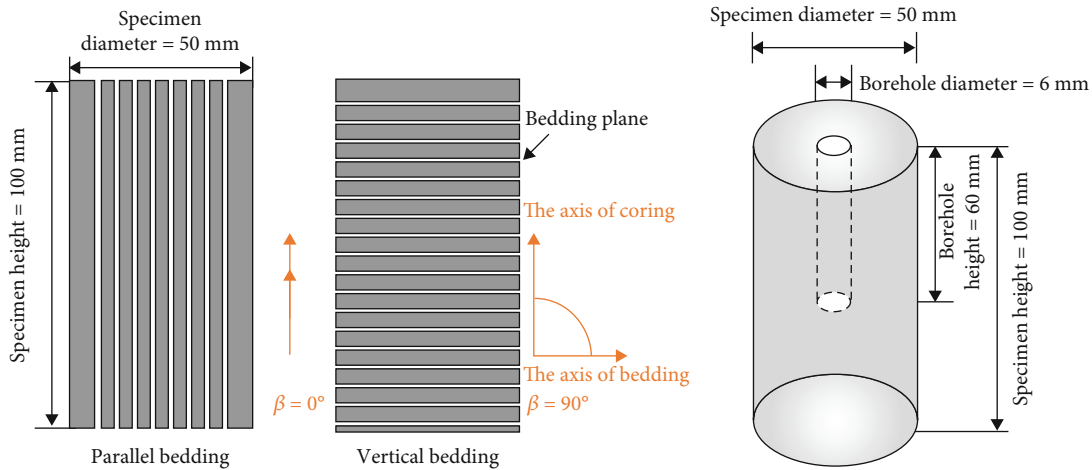
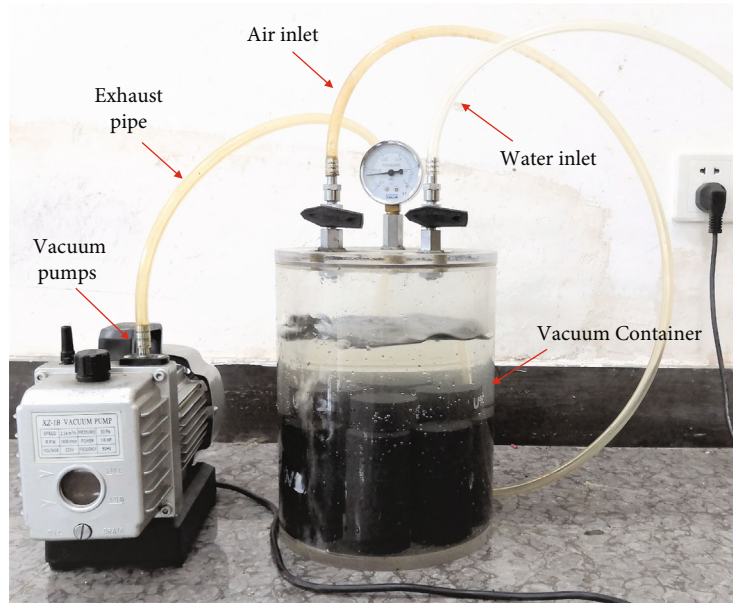


FIGURE 2: Sample preparation.

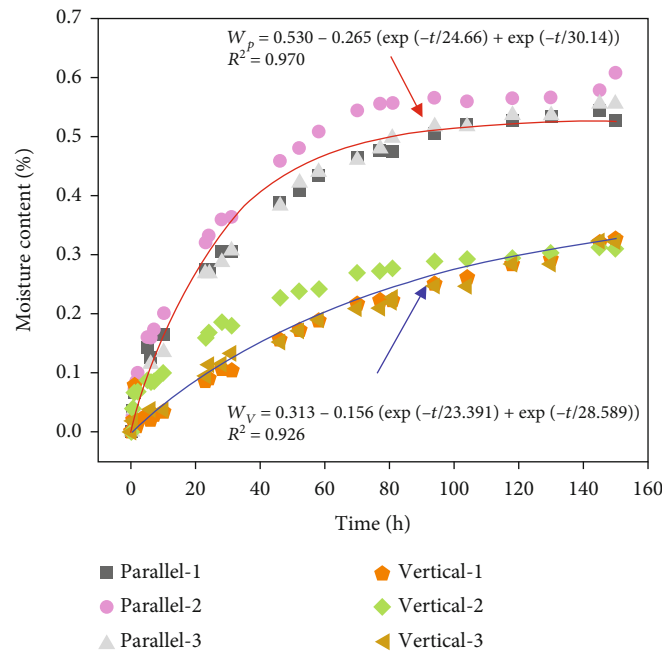
reservoir for enhancing the complexity of fracture system [4, 5]. The merits of hydraulic fracturing technology include its low cost and convenient availability [6]. However, the shortcomings are obvious in the process of fracturing. First, shale gas reservoir contains clay minerals. These minerals are prone to swelling, resulting in water lock. Meanwhile, the mineral hydration takes place, resulting in a reduction of effective permeability [7]. Second, water-based fracturing fluids contain chemical impurities and toxic compositions. These synthetics inevitably linger in gas reservoirs and threaten clean water resources [8]. Moreover, the purification treatments increase the development cost. Third, water usage for hydraulic fracturing ranges from  $1400 \text{ m}^3$  to  $33900 \text{ m}^3$  per horizontal well [9, 10]. The shale gas reservoirs are located in hilly and arid regions, such as Wuning area and Sichuan Basin in China

[11]. Therefore, novel waterless fracturing fluids have been drawn attention to solve the water-related issues.

There have been many studies on waterless fracturing fluids, such as liquefied petroleum gases [12–14], supercritical carbon dioxide or liquid carbon dioxide [15–17], high energy gas [18, 19], nitrogen foam [20, 21], and  $\text{LN}_2$  [6, 11]. These waterless fracturing technologies have been applied. The burial depth of recoverable shale gas reservoirs ranges between 1000 m and 5000 m. The shale reservoir temperature is high [22].  $\text{LN}_2$  fracturing technology has received more concentration among waterless fracturing technologies. For example, the reservoir temperature of Cooper Basin reaches  $150^\circ\text{C}$  at the burial depth of 3500 m [23]. When  $\text{LN}_2$  is delivered to the shale gas reservoir, a sharp thermal gradient of more than  $300^\circ\text{C}$  is generated in the local area of shale formation. The high local thermal stress induces primary



(a)

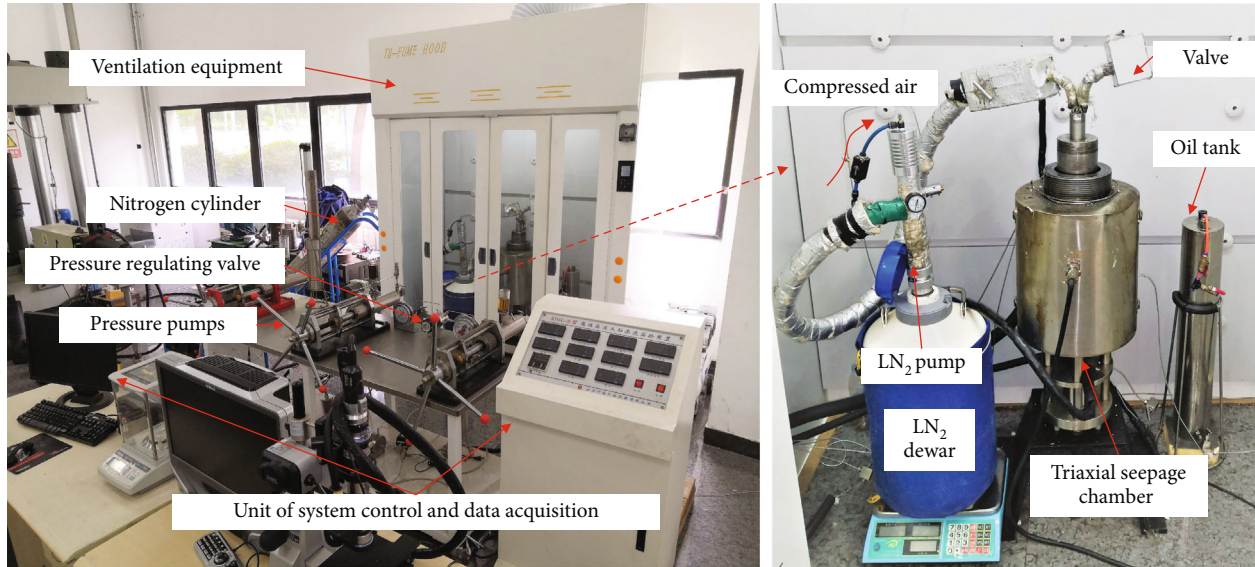


(b)

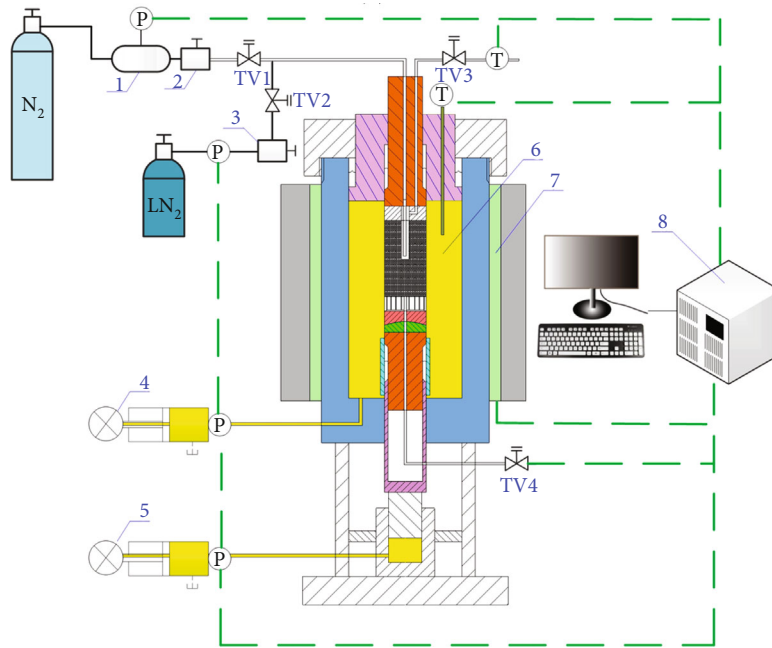
FIGURE 3: (a) Vacuum-saturated device. (b) Moisture content-time curve of shale sample.

fractures and promotes secondary development of preexisting fractures [24]. Laboratory studies demonstrated that the injection pressure decreased 40% under LN<sub>2</sub> fracturing [25, 26]. The shortage of LN<sub>2</sub> fracturing is the high equipment cost. The drilling pipelines and fracturing pumps bear the burden of adapting the high pressure and low viscosity characteristics of LN<sub>2</sub> [27]. A novel fracturing technology proposed that LN<sub>2</sub> was utilized as assist-gas fluid to cryogenic damage of the reservoir, and nitrogen was used as fracturing fluid to improve the fracture complexity [28]. Both LN<sub>2</sub> and nitrogen are easy to flowback and friendly towards the environment [29, 30].

The effect of cryogenic damage after LN<sub>2</sub> preconditioning is affected by not only the environmental factors such as reservoir temperature, ground stress, and moisture content but also the internal structure factors such as mineralogical composition, microcracks, and bedding orientation [31, 32]. The cryogenic damage extent of frost heave is controlled by initial moisture content [33]. When water freezes, its volume expands by 9%. Besides, the unfrozen water is repelled by disjoining force and moves to the growing ice. More microcracks develop under the freezing condition [34]. Numerous studies have investigated the mechanisms of frost heave and established models to explain the microcrack



(a)



- |   |                                    |
|---|------------------------------------|
| 1 High pressure gasholder                   | 5 Axial pressure pump              |
| 2 Nitrogen pressure regulating valve        | 6 Triaxial pressure chamber        |
| 3 LN <sub>2</sub> pressure regulating valve | 7 Electric heating device          |
| 4 Confining pressure pump                   | 8 Real-time data collection system |
| TV1 Nitrogen inlet valve                    | TV3 LN <sub>2</sub> outlet valve   |
| TV2 LN <sub>2</sub> inlet valve             | TV4 Nitrogen outlet valve          |
| Ⓟ Pressure sensor                           | Ⓣ Temperature sensor               |

(b)

FIGURE 4: (a) The experimental device. (b) Diagram of real triaxial seepage and fracturing device.

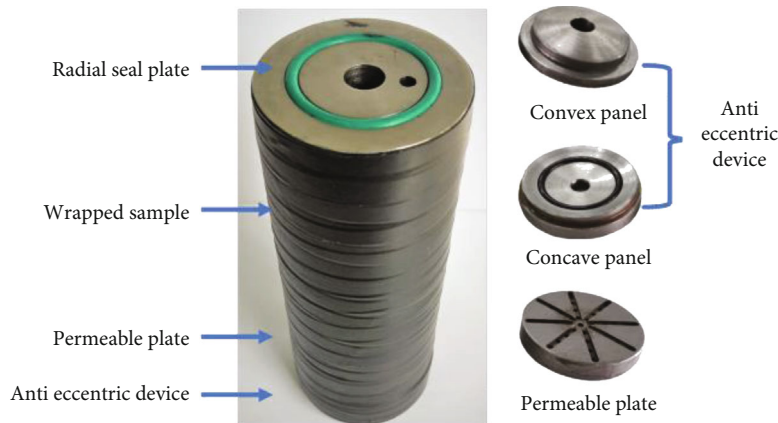
generation and the process of ice crystallization [35–37]. However, the studies on the cryogenic damage of low-permeability rocks by frost heave need to be further improved. For example, the breakdown pressure variation

of saturated shale after LN<sub>2</sub> preconditioning is neglected because of the low moisture content.

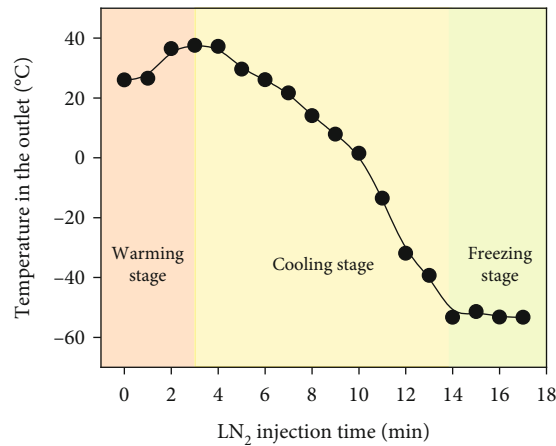
In this paper, the cracking characteristics of dry and saturated shales after LN<sub>2</sub> preconditioning were studied by

TABLE 1: Experimental scheme.

Experimental group	Bedding direction	Moisture content	Influence factors		
			LN <sub>2</sub> preconditioning time (min)	Confining pressure (MPa)	
1	Parallel	Saturated	0	3, 6, 9, 12	
2			15		
3		Dry	0		
4			15		
5	Vertical	Saturated	0, 15		3
6		Dry			



(a)



(b)

FIGURE 5: (a) The assembly of sample in the triaxial device. (b) The change of LN<sub>2</sub> outlet temperature.

performing lots of laboratory experiments. First, X-ray diffractometer (XRD) was assisted in the mineralogy analysis on the shale. Second, a laboratory triaxial and high-temperature fracturing system was developed. The nitrogen fracturing after LN<sub>2</sub> preconditioning tests was performed at confining pressure (3 MPa, 6 MPa, 9 MPa, and 12 MPa) and high temperature (100°C) to investigate the breakdown pressure characteristics between dry and saturated shales. Thirdly, the fracture surface of shale was observed by optical microscope to study its macroscopic damage fracture characteristics. Finally, the tensile strength was tested to explain the breakdown pressure difference between different bed-

ding directions. The experimental investigation evaluates the cryogenic damage effect between dry and saturated shales after LN<sub>2</sub> preconditioning. The analysis of frost heave provides insight into cryogenic LN<sub>2</sub> fracturing technology.

## 2. Experimental Preparation and Methods

2.1. *Materials.* The shale blocks were collected from the Wuning area, Jiangxi Province, China. It belongs to the Upper Ordovician Xinkailing Formation [38–40]. An X-ray diffractometer (XRD) was used to investigate the mineral composition of shale. The result is plotted in Figure 1. The



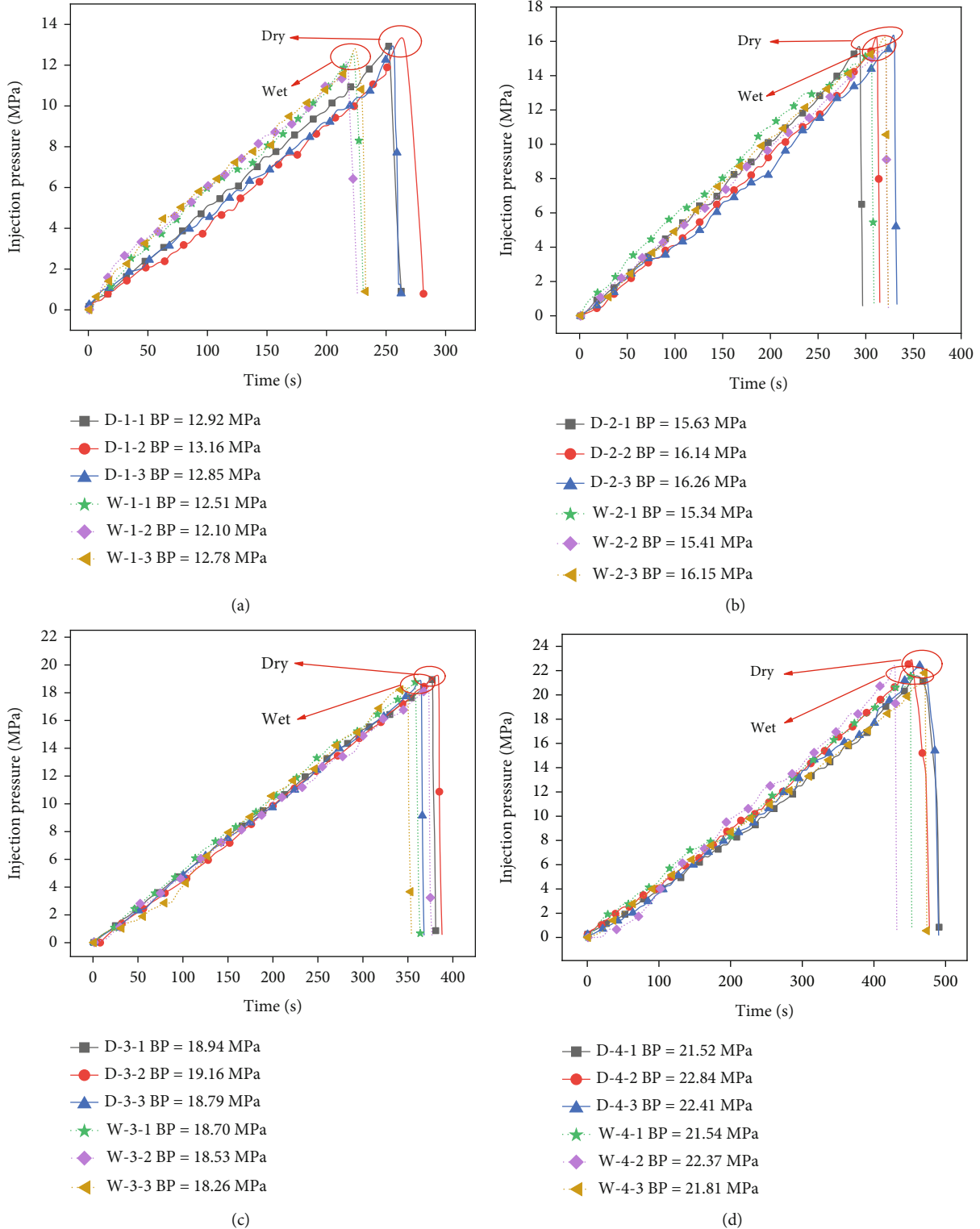


FIGURE 6: The nitrogen fracturing curves of dry and saturated parallel bedding shales without  $\text{LN}_2$  injection under different confining pressures: (a) confining pressure = 3 MPa; (b) confining pressure = 6 MPa; (c) confining pressure = 9 MPa; (d) confining pressure = 12 MPa.

primary constituents of shale were quartz, clinocllore, calcite, muscovite, and albite. The siliceous minerals of shale sample include quartz and albite, accounting for 40.9% of the main components. These minerals offer the benefits for the formation of organic-rich shale [41]. High siliceous con-

tent is also favorable for fracturing shale reservoir [42–44]. Inorganic micropores in shale are readily preserved in the process of deposition and conducive to hydrocarbon storage [45]. The formation of inorganic micropores is related to clay minerals [46]. The clay mineral of shale sample was

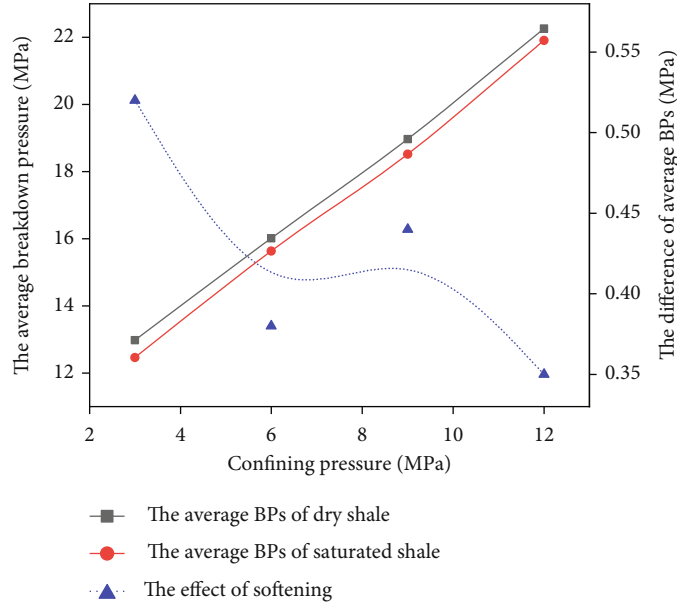


FIGURE 7: The breakdown pressure variation and the difference of average breakdown pressure between dry and saturated shales without  $\text{LN}_2$  preconditioning under different confining pressures.

clinochlore, accounting for 21.2% of the main components. Therefore, the Upper Ordovician Xinkailing Formation shale has development potential.

**2.2. Shale Sample Preparation.** Shale is one of the sedimentary rocks with many alternating beddings of thin and thick layers. These beddings greatly affect the mechanical properties of shale. Therefore, two bedding angles ( $\beta = 0^\circ, 90^\circ$ , as shown in Figure 2) were selected in the experiments. The shale block was cored as cylinders following the ASTM standard [47, 48]. A borehole was drilled for  $\text{LN}_2$  preconditioning and nitrogen fracturing in triaxial high-temperature system. The diameter of borehole was 6 mm and the length was 60 mm.

The cylinders were heated in a  $110^\circ\text{C}$  oven until the weight no longer changed. The dry shale mass was  $m_d$ . Then, the dry samples were placed into a vacuum container. The vacuum saturation method was used to prepare saturated samples. The pressure of the container was maintained at  $-0.1$  MPa for one hour. Air was expelled from the shale pores under negative pressure. Then, open the water inlet until the dry samples were submerged, as shown in Figure 3(a). The negative pressure was maintained during the process of water injection. The wet sample was weighed every two hours and recorded as  $m_s$ . The moisture content ( $\omega$ ) can be simply estimated by Equation (1). The wet sample was saturated when the moisture content no longer increased. The moisture content variation is illustrated in Figure 3(b). There were two curves of moisture content between different bedding directions. The mean moisture content of the saturated samples with parallel bedding was 0.530%, while that with vertical bedding was only 0.313%. The average shale mass of dry vertical bedding shale samples was 528.91 g, but that of dry parallel bedding shale was 520.48 g. The shale

sample with vertical bedding was denser with fewer micropores and microcracks.

$$\omega = \frac{m_s - m_d}{m_d} \times 100\%. \quad (1)$$

**2.3. Experimental Methods.** An experimental device was designed for nitrogen fracturing shale in the in situ condition as shown in Figure 4(a). The device is capable of heating sample to  $300^\circ\text{C}$  and exerting triaxial pressure (up to 60 MPa). There are four experimental units in the device: a  $\text{LN}_2$  injection unit, a high-temperature and triaxial pressure unit, a nitrogen fracturing unit, and a real-time data collection unit as shown in Figure 4(b). The detailed introduction of experimental device was published on another articles [28, 49, 50].

The axial pressure was kept at 30 MPa to prevent gas leakage during the injection of  $\text{LN}_2$  and nitrogen fracturing. The sample temperature was set to  $100^\circ\text{C}$  for simulating the in situ temperature. The experimental variables are listed in Table 1. The cracking characteristics of dry and saturated shales after  $\text{LN}_2$  injection were investigated by the six experimental groups under different confining pressures and bedding orientations.

**2.4. Experimental Procedure.** The experiment was mainly divided into two stages: the cryogenic damage stage by  $\text{LN}_2$  injection and the fracturing stage by nitrogen injection. The injection of  $\text{LN}_2$  was as follows:

- (1) The assembly sample was installed into the triaxial pressure chamber. The radical seal plate, shale sample with borehole, permeable plate, and anticentric

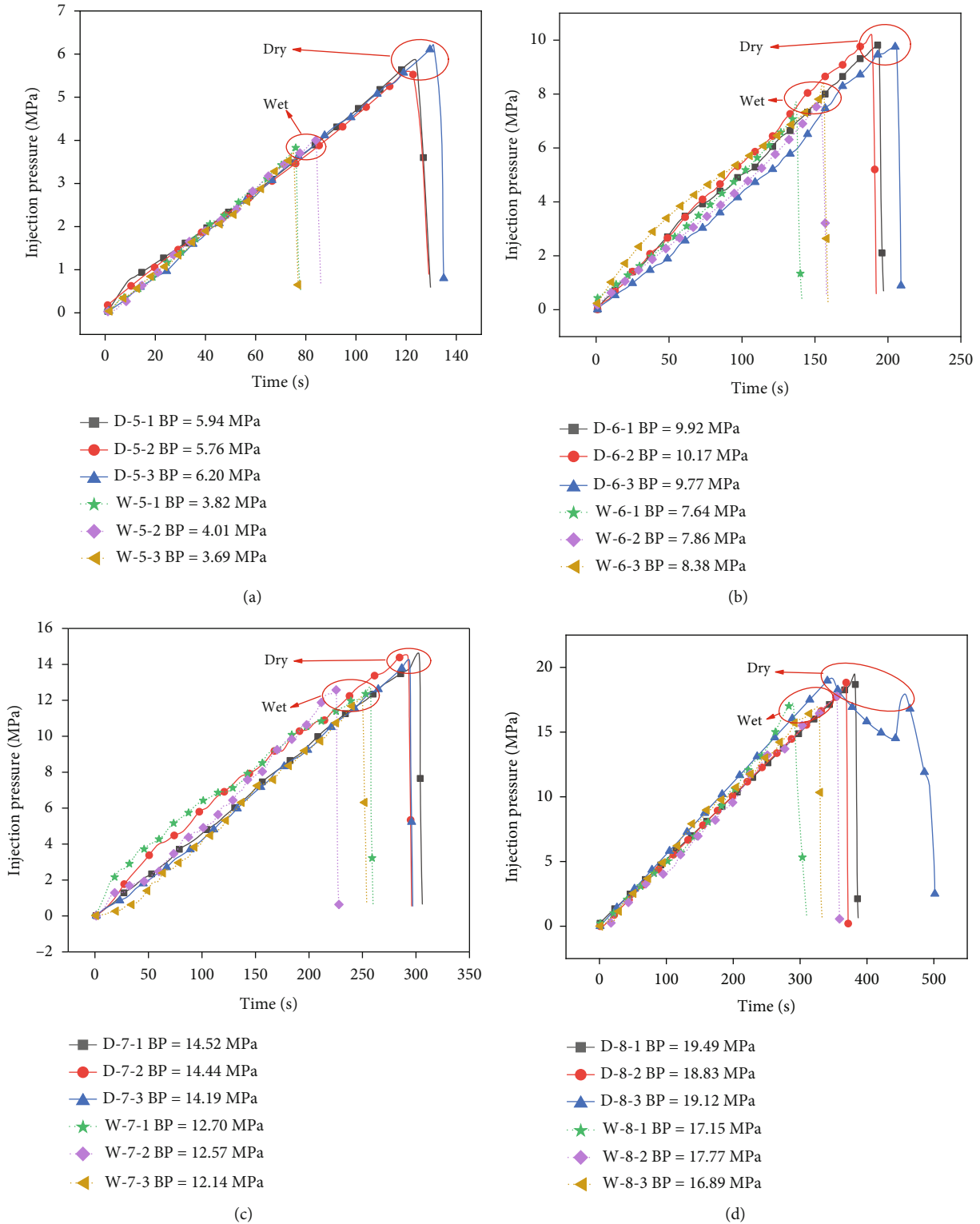


FIGURE 8: The nitrogen fracturing curves of dry and saturated parallel bedding shales after LN<sub>2</sub> injection under different confining pressures: (a) confining pressure = 3 MPa; (b) confining pressure = 6 MPa; (c) confining pressure = 9 MPa; (d) confining pressure = 12 MPa.

device were assembled together, as shown in Figure 5(a)

(2) The experimental axial pressure was applied to 30 MPa using the pressure pump. The confining

pressure was adjusted to the target values (3, 6, 9, and 12 MPa) according to the experimental scheme. The temperature of heating device was maintained at 100°C for 2 h before the injection of LN<sub>2</sub>. As a result, the shale sample could be heated to 100°C



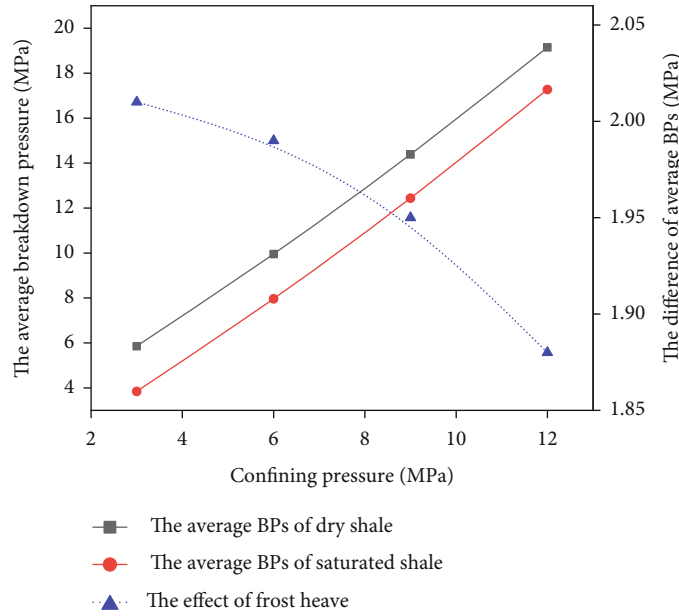


FIGURE 9: The average breakdown pressure variation and difference between dry and saturated shales after LN<sub>2</sub> preconditioning under different confining pressures.

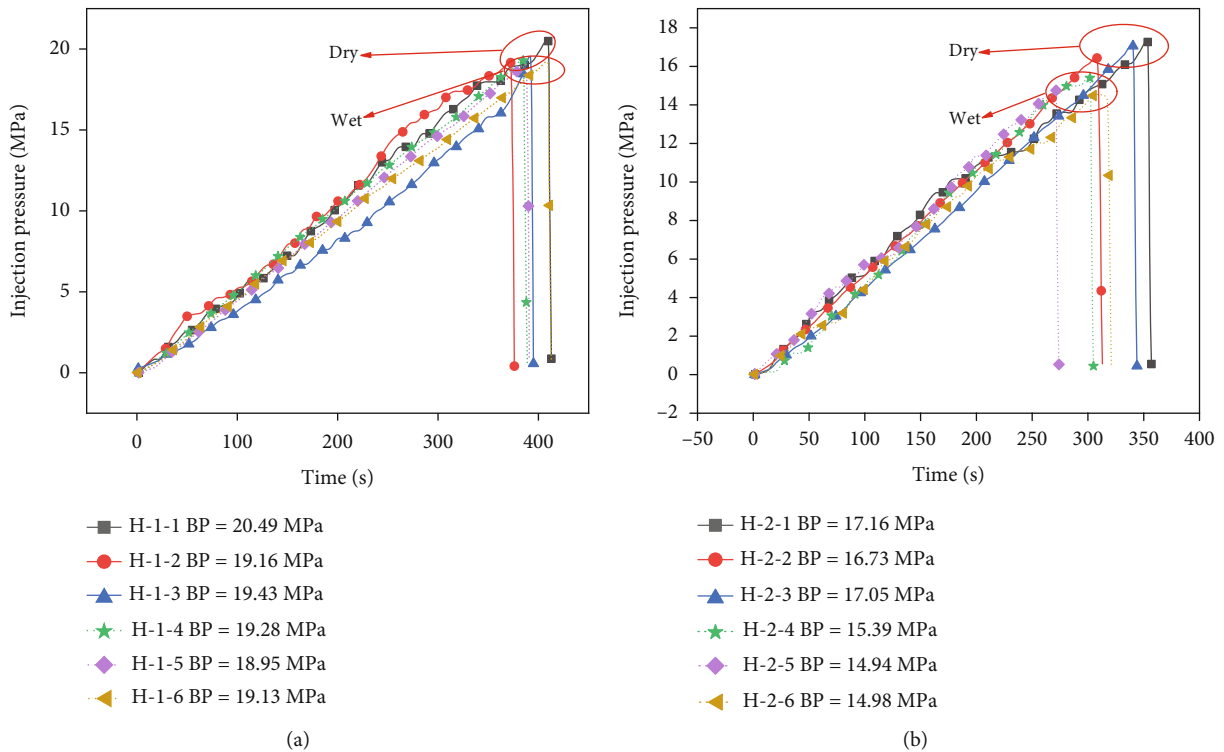


FIGURE 10: The nitrogen fracturing curves of dry and saturated vertical bedding shales under 3 MPa confining pressures with and without LN<sub>2</sub> injection: (a) without LN<sub>2</sub> injection; (b) with LN<sub>2</sub> injection.

(3) Before LN<sub>2</sub> preconditioning, the nitrogen valves (TV1 and TV4) were closed, and the LN<sub>2</sub> valves (TV2 and TV3) were turned on to facilitate the injection of LN<sub>2</sub> into the specimen for cryogenic damage

(4) LN<sub>2</sub> was delivered into the borehole of shale sample by the LN<sub>2</sub> pump. The injecting rate was controlled at 0.2 L/min. The residual LN<sub>2</sub> was discharged from the LN<sub>2</sub> outlet valve (TV3)

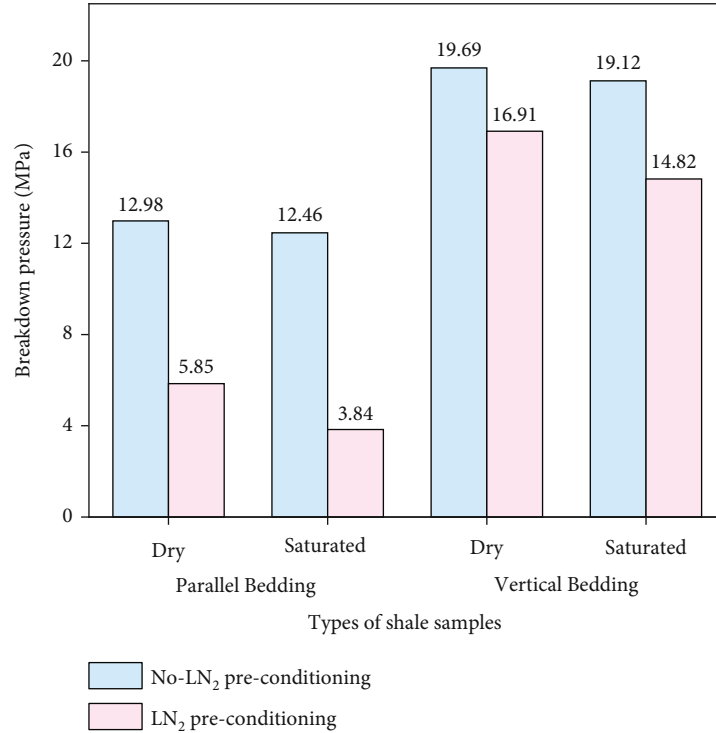


FIGURE 11: Average breakdown pressure comparison between parallel and vertical bedding shales.

- (5) The process of LN<sub>2</sub> preconditioning lasted 15 min. The injection pressure and outlet temperature were monitored by computer

The temperature variation of the LN<sub>2</sub> outlet (TV3) is illustrated in Figure 5(b). There were three stages in the process of LN<sub>2</sub> preconditioning. The outlet temperature increased from 26.0°C to 37.6°C in the warming stage. LN<sub>2</sub> was completely vaporized and extracted heat from the shale surface. The temperature of outlet decreased from 37.6°C to -53.3°C during the cooling stage. The rate of the shale temperature reduction was greater than the thermal compensation rate of heating device. The outlet temperature remained steady at -53.3°C in the freezing stage. A water-ice phase transition occurred, resulting in frost heave.

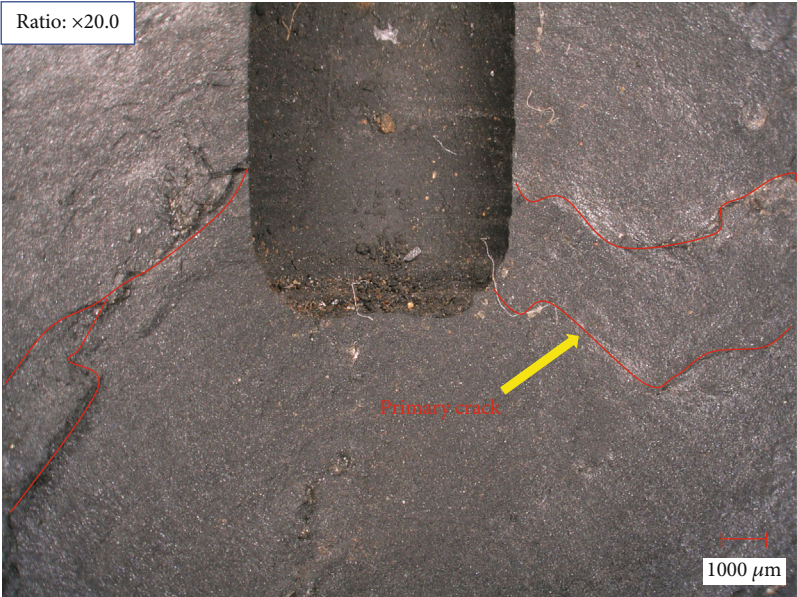
The procedures of nitrogen fracturing were as follows:

- (1) The nitrogen inlet and outlet valves (TV1 and TV4) were turned on to facilitate the injection of nitrogen into the specimen borehole, while the LN<sub>2</sub> inlet and outlet valves (TV2 and TV3) were turned off
- (2) The nitrogen pressure regulating valve was used to inject nitrogen into the shale borehole. The rate of pressurization was 50 kPa/s. The variation of injection pressure was monitored by pressure sensor. When the nitrogen flowed out of the nitrogen outlet valve (TV4), the shale specimen was split into halves. The peak pressure was the breakdown pressure of shale

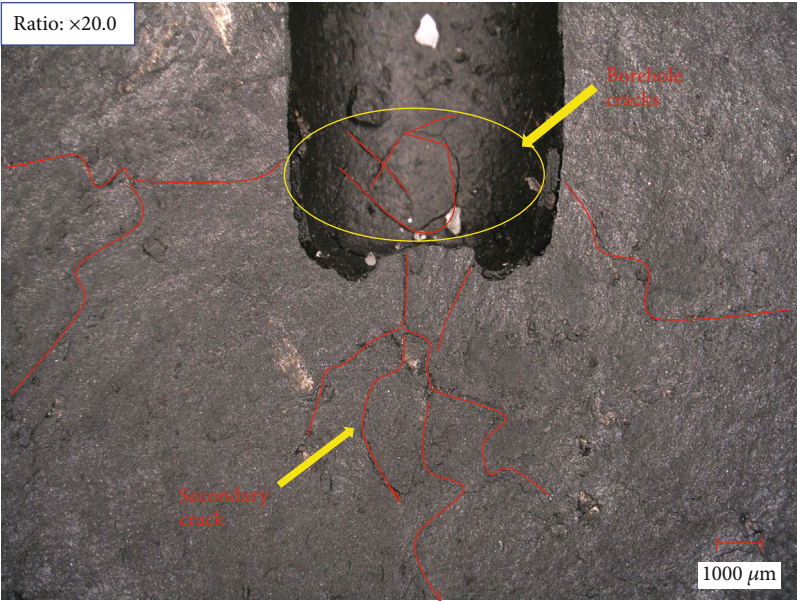
### 3. Experimental Results

*3.1. The Breakdown Pressure Characteristics of Dry and Saturated Shales before LN<sub>2</sub> Preconditioning.* Comparing the experimental results between group 1 and group 3, the nitrogen fracturing curves for dry and saturated parallel bedding shales without LN<sub>2</sub> preconditioning are plotted in Figures 6(a)–6(d). The increase of injection pressure was approximately linear by pressure regulating valve during nitrogen fracturing. The end signal of nitrogen fracturing was a sharp reduction of injection pressure. Therefore, the peak of injection pressure was the breakdown pressure (BP) of shale.

When the confining pressure increased from 3 MPa to 12 MPa, the average breakdown pressures of dry samples were 12.98 MPa, 16.01 MPa, 18.96 MPa, and 22.26 MPa, respectively, and those of saturated samples were 12.46 MPa, 15.63 MPa, 18.52 MPa, and 21.91 MPa, respectively. The breakdown pressure variation and the average breakdown pressure differences between dry and saturated shales are plotted in Figure 7. The increase in average breakdown pressure was approximately linear in relation to the increase in confining pressure. In addition, there was little variation between dry and saturated shale samples without LN<sub>2</sub> preconditioning. The differences of average breakdown pressures were 0.51 MPa, 0.38 MPa, 0.44 MPa, and 0.35 MPa with the confining pressure increasing. Therefore, the softening influence decreased with the confining pressure increasing, and the effect of softening was small on the breakdown pressure without LN<sub>2</sub> injection.



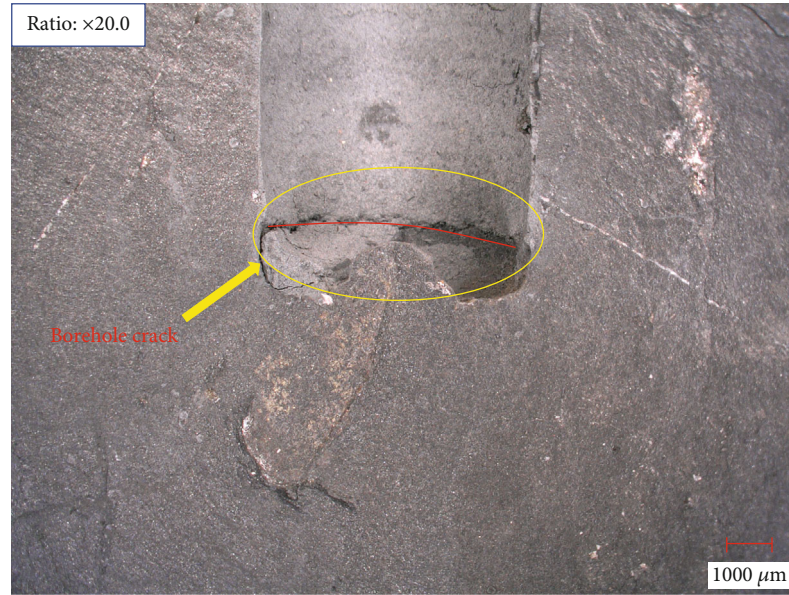
(a)



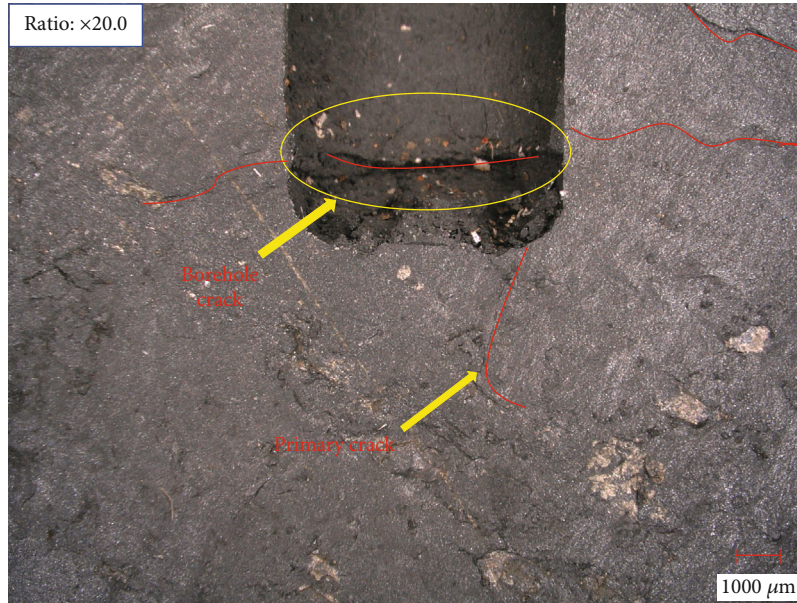
(b)

FIGURE 12: Continued.





(c)



(d)

FIGURE 12: The crack propagation around the borehole: (a) dry and parallel bedding shales; (b) saturated and parallel bedding shales; (c) dry and vertical bedding shales; (d) saturated and vertical bedding shales.

**3.2. The Breakdown Pressure Characteristics of Dry and Saturated Shales after  $\text{LN}_2$  Preconditioning.** The nitrogen fracturing curves of dry and saturated parallel bedding shales after  $\text{LN}_2$  injection are plotted in Figures 8(a)–8(d), when the experimental results between group 2 and group 4 were compared. Most curves of injection pressure were approximately linear by pressure regulating valve during nitrogen fracturing, but there was one special sample that underwent two fracturing processes. When the confining pressure was 12 MPa, the D-8-3 sample was not completely split into two halves at the first peak pressure. The injected nitrogen remained in the shale borehole and could not be exhausted

through the nitrogen outlet. The nitrogen continued to be injected into the D-8-3 sample by the nitrogen pressure regulating valve until the sample was finally fractured. The second peak pressure was lower than the first peak pressure. The fracture closed rapidly under the high confining pressure during the first fracturing stage. Therefore, the high confining pressure was not conducive to the propagation of macrocracks.

The average breakdown pressures of dry shale with  $\text{LN}_2$  preconditioning were 5.85 MPa, 9.92 MPa, 14.38 MPa, and 19.14 MPa, respectively, and those of saturated shale were 3.84 MPa, 7.96 MPa, 12.44 MPa, and 17.27 MPa as the

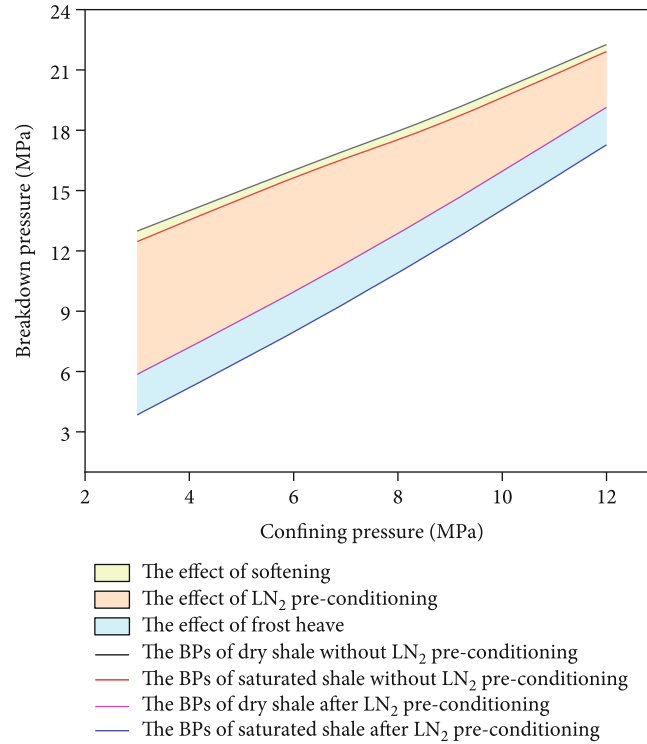


FIGURE 13: Average breakdown pressure of dry and saturated parallel bedding shales under different confining pressures.

confining pressure grew. Therefore, the cryogenic damage effect was significant by  $\text{LN}_2$  injection. The breakdown pressure of dry and saturated shales both decreased after  $\text{LN}_2$  injection compared to the experimental results of group 1 and group 3.

After  $\text{LN}_2$  preconditioning, the curves of average breakdown pressure differences between dry and saturated shales are plotted in Figure 9. The average breakdown pressures of saturated shale decreased 2.01 MPa, 1.99 MPa, 1.94 MPa, and 1.88 MPa, respectively, compared to those of dry shale. The water-ice phase transition under freezing condition causes frost heave in saturated shale. The effect of frost heave was at least 3.9 times more than that of softening. When the confining pressure increased, the average breakdown pressure difference after  $\text{LN}_2$  injection still decreased.

**3.3. The Breakdown Pressure Characteristics of Dry and Saturated Shales between Different Bedding Directions after  $\text{LN}_2$  Preconditioning.** The nitrogen fracturing curves of dry and saturated vertical bedding shales are plotted in Figures 10(a) and 10(b). The average breakdown pressure of dry vertical bedding shale without  $\text{LN}_2$  preconditioning was 19.69 MPa, and that of saturated vertical shale was 19.12 MPa. There was only 0.57 MPa reduction under the moisture content. After  $\text{LN}_2$  preconditioning, the average breakdown pressure decreased by 14.12% and 22.49% between dry and saturated shales. The difference of average breakdown pressure between dry and saturated vertical bedding shales was 2.09 MPa after  $\text{LN}_2$  preconditioning. Therefore, the moisture in shale promoted the effect of  $\text{LN}_2$  preconditioning.

The average breakdown pressures between parallel and vertical bedding shales at the confining pressure of 3 MPa are shown in Figure 11. The bedding direction had a great influence on the nitrogen fracturing. The average breakdown pressures of vertical bedding shale were all higher than those of parallel bedding shale. Moreover, the average breakdown pressure difference between parallel bedding and vertical bedding shales increased after  $\text{LN}_2$  preconditioning.

New microcracks of shale were formed during the process of  $\text{LN}_2$  preconditioning, and the cracks spread by nitrogen injection. The propagation of cracks around the borehole after  $\text{LN}_2$  preconditioning is illustrated in Figures 12(a)–12(d). The number of cracks on saturated fracture surface was more than that on dry fracture surface. There were only a few single primary cracks around the dry borehole. However, many secondary cracks were generated around the saturated borehole, forming a complex fracture network. The number of cracks on parallel bedding fracture surface was more than that of vertical bedding fracture surface. Therefore, saturated parallel bedding shale was easy to fracture after  $\text{LN}_2$  preconditioning. In addition, the cracks preferred to propagate along the bedding direction.

## 4. Discussions

According to the observations of the nitrogen fracturing dry and saturated parallel bedding shales after  $\text{LN}_2$  preconditioning, the curves of breakdown pressure under different confining pressures are depicted in Figure 13. The breakdown pressure difference between dry and saturated shales

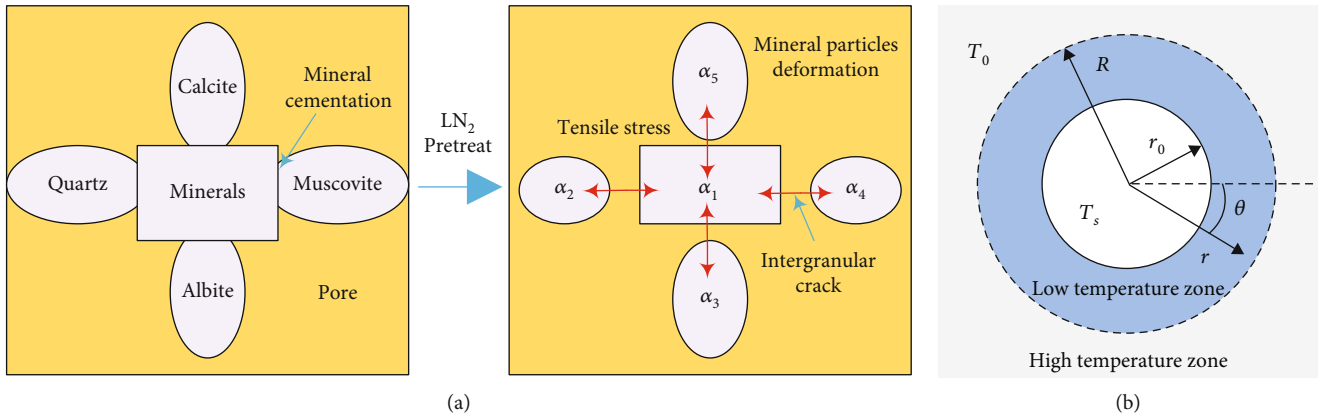


FIGURE 14: (a) Diagram of mineral particle shrinkage after LN<sub>2</sub> preconditioning. (b) Diagram of temperature field around the borehole during LN<sub>2</sub> preconditioning.

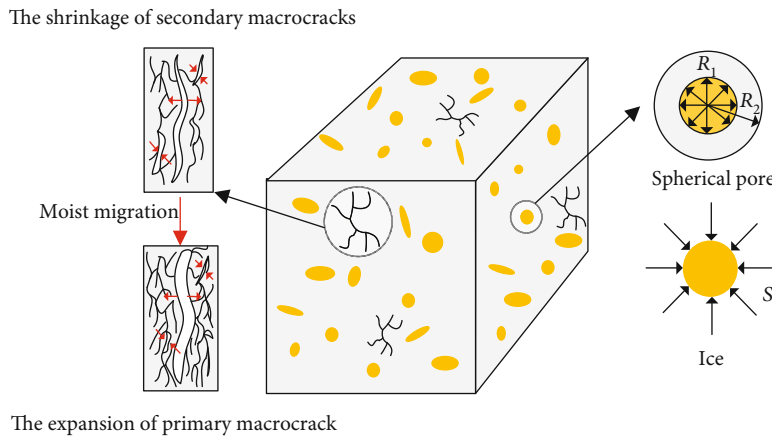


FIGURE 15: The propagation of spherical pore and primary macrocracks under frost heaving.

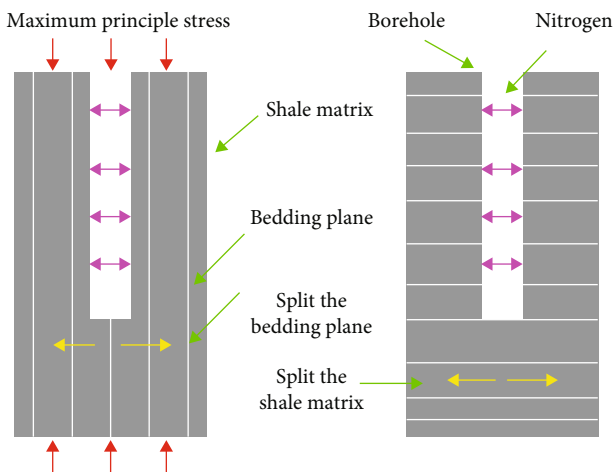


FIGURE 16: Diagram of shale in the nitrogen fracturing test.

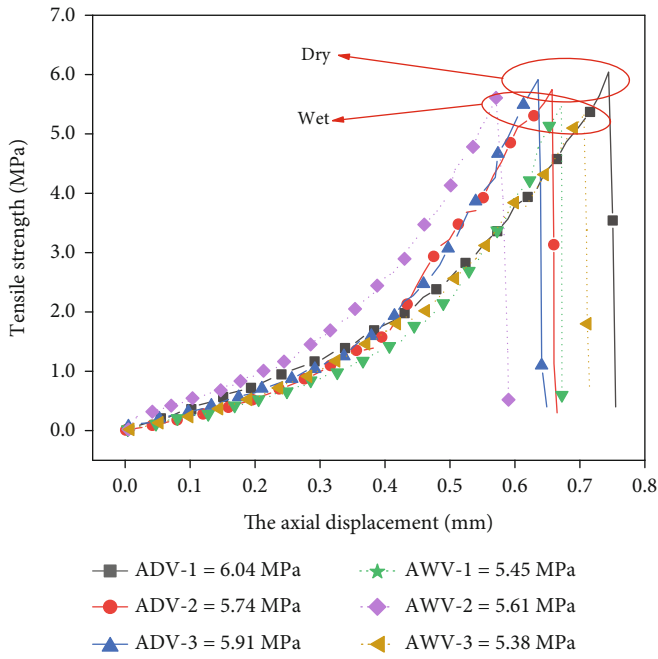
without LN<sub>2</sub> preconditioning was small. There were muscovite and clay minerals in shale. The hydration and expansion stress lead to the fragmentation and disintegration of mineral particles. Although these minerals hydrate with water

in pores and macrocracks and expand in volume, there was only 0.53% moisture content in the saturated parallel bedding shale. Therefore, the effect of softening on breakdown pressure was small.

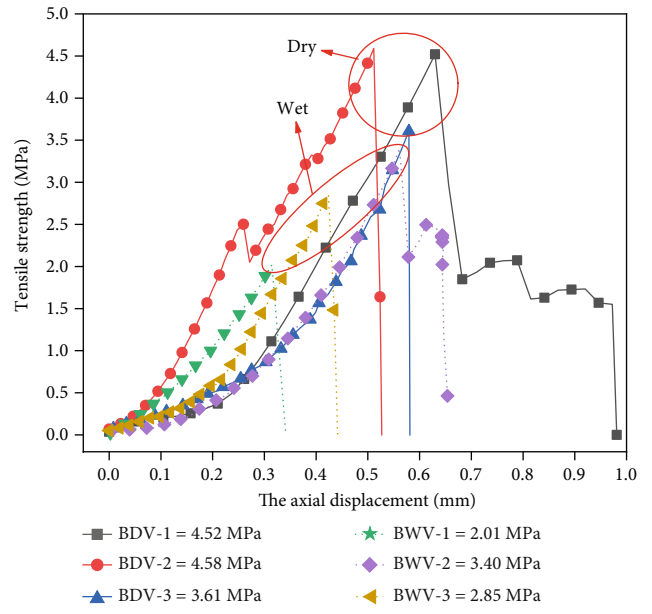
The effect of LN<sub>2</sub> preconditioning was significant, as shown in Figure 13. When LN<sub>2</sub> was delivered to the shale sample at 100°C, a sharp thermal gradient approaching 300°C was generated in the local area. The local thermal stress is the main reason of the cryogenic damage of shale, as shown in Figure 14. There are two types of crack propagation caused by the local thermal stress:

- (1) Shale consists of different mineral grains, such as quartz, albite, and muscovite. These mineral particles have different thermophysical and mechanical characteristics. The adjacent mineral particles deform to different degrees during LN<sub>2</sub> preconditioning [51], as shown in Figure 14(a). Intergranular cracks develop as the cementitious strength of minerals is lower than the tensile stress [52]
- (2) A violent heat transfer occurs around the borehole, forming a low-temperature zone with LN<sub>2</sub> injection. The zone of low temperature cannot shrink freely

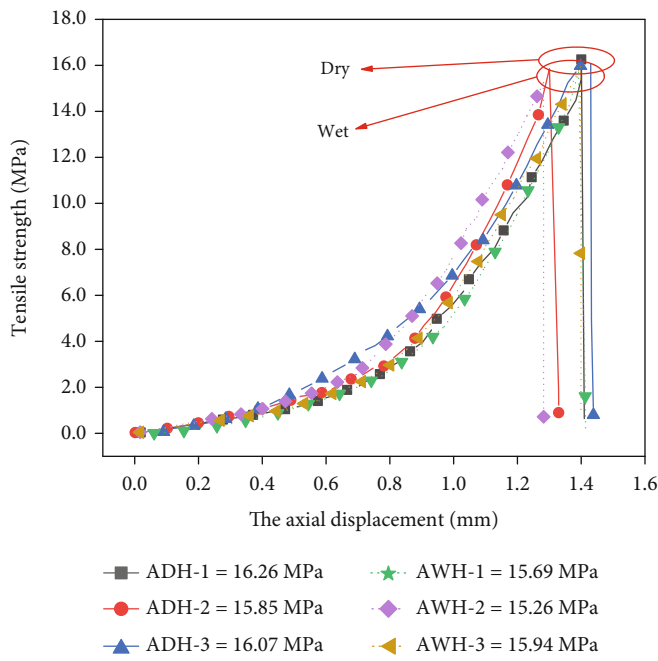




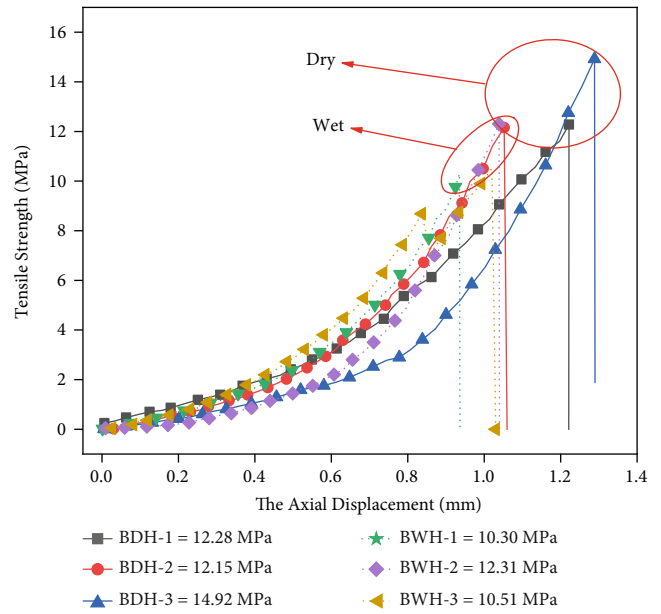
(a)



(b)



(c)



(d)

FIGURE 17: Continued.



FIGURE 17: The stress-displacement curve of dry and saturated shales: (a) parallel bedding shales without  $\text{LN}_2$  preconditioning; (b) parallel bedding shales after  $\text{LN}_2$  preconditioning; (c) vertical bedding shales without  $\text{LN}_2$  preconditioning; (d) vertical bedding shales after  $\text{LN}_2$  preconditioning. The failure pattern of shale in the Brazilian disc test: (e) parallel bedding shale; (f) vertical bedding shale.

because of the restrictions imposed by the surrounding rock, resulting in the local tensile stress, as shown in Figure 14(b). When the local tensile stress exceeds the shale matrix's tension stress, new cracks form

The damage characteristics of saturated shale samples are more complicated after  $\text{LN}_2$  preconditioning, compared to the dry shale samples. When the shale samples were pretreated by  $\text{LN}_2$ , there was a clear difference between dry and saturated shales, as shown in Figure 13. The water in pores and macrocracks was frozen during  $\text{LN}_2$  preconditioning. More cracks were produced due to the frost heave. Furthermore, the effect of frost heave would not decrease with the confining pressure increasing.

There are two mechanical mechanisms of shale fracture driven by ice growth, as shown in Figure 15:

- (1) Shale has low permeability; thus, water is hard to flow out the pores in the matrix [53]. If the spherical pores are entirely filled with water as shown in Figure 15, the water will freeze under  $\text{LN}_2$  preconditioning. The volume expansion of ice acting on the spherical pore walls causes frost heave. The microcracks develop around the pores when the expansion stress of ice exceeds the tensile stress of pore
- (2) Water exists not only in the pores but also in the cracks. There are many interconnected natural cracks. The freezing point of different volume cracks varied due to the capillary effect. The water in the larger crack is more likely to freeze [54]. Therefore, the water in the primary crack freezes under  $\text{LN}_2$  preconditioning. When water freezes, its volume expands by 9%. The volume of primary crack keeps growing with the volume expansion of ice. In the frozen fringe, water migrates continuously to ice when the pressure of crack decreases [55]. The water in the secondary cracks migrates to the unfrozen area of primary crack due to the surface-energy effect [56]. Meanwhile, the secondary cracks lose water and shrink during  $\text{LN}_2$  preconditioning. The crack deformation is restricted by shale matrix, creating a localized tensile stress. The local tensile stress around the cracks is conducive to the propagation of cracks.

Therefore, the migration of water is favorable for the expansion of primary crack

According to the observations of the cracking characteristics between different bedding directions after  $\text{LN}_2$  preconditioning, there was wide variation in the average breakdown pressure. As shown in Figure 16, the tensile stress induced by nitrogen injection was applied to the shale borehole. When the injection pressure was greater than the crack initiation pressure, the crack would occur around the shale borehole. The crack propagation tended to spread along the maximum principal stress direction. The axial pressure (30 MPa) was always higher than the confining pressure (3 MPa-12 MPa) during the nitrogen fracturing test. Therefore, the axial direction was the direction of crack. The bedding was one of the discontinuities in shale. When the bedding direction coincided with the direction of fracture propagation, the shale was split along the bedding orientation. The shale matrix was split by the injection pressure while the fracture propagation was vertical to the bedding orientation. Therefore, the reason for the variation of the average breakdown pressure was the different tensile strengths between shale bedding and matrix.

The Brazilian disc tests of dry and saturated shales were conducted to study the different tensile strengths between parallel and vertical bedding directions. The stress-displacement curves of parallel bedding shale are depicted in Figures 17(a) and 17(b). The mean tensile strength of dry parallel bedding shale without  $\text{LN}_2$  preconditioning was 5.90 MPa, and that of saturated shale was 5.48 MPa. The difference between dry and saturated shales was small because the moisture in shale had a little influence on the tensile strength. When the parallel bedding shale samples were pretreated by  $\text{LN}_2$ , the mean tensile strength of dry shale decreased 1.66 MPa and that of saturated shale decreased 2.73 MPa. Therefore, the reduction of tensile strength caused by the frost heave was 1.07 MPa. The frost heave was beneficial for the cryogenic damage. The failure of parallel bedding specimen was a typical splitting mode as shown in Figure 17(e). An approximate linear central fracture was formed along the bedding and split the specimen into halves. Therefore, the tensile strength depended on the bedding cementation.

The stress-displacement curves of vertical bedding shale are depicted in Figures 17(c) and 17(d). Compared to the

tensile strength of parallel bedding shale, that of vertical bedding shale was higher. The mean tensile strength of dry vertical bedding shale without LN<sub>2</sub> preconditioning was 16.06 MPa, which was 2.72 times greater than that of dry parallel bedding shale. The difference of tensile strength between dry and saturated shales was 0.43 MPa. Therefore, the moisture effect on the shale was minimal. After LN<sub>2</sub> preconditioning, the mean tensile strength of dry vertical bedding shale was 13.12 MPa and that of saturated shale was 11.04 MPa. Although the frost heave reduced the tensile strength by 1.65 MPa, the mean tensile strength of vertical bedding shale was still higher than that of parallel bedding shale. The failure of vertical bedding specimen included splitting failure and shear slide modes as shown in Figure 17(f). The propagation of fracture deviated from the loading direction, forming an arc. Moreover, the fractures occurred on the bedding plane, splitting the specimen into slices. Therefore, when the stress loading direction was vertical to the bedding direction, the tensile strength depended on the cementitious force of the matrix, and the bedding direction had an influence on the propagation of fracture. In conclusion, the cementation of bedding was weaker than that of the matrix, and the fracturing direction should be consistent with the bedding direction for relieving the equipment burden and forming more complex fracture network.

## 5. Conclusions

According to the characteristics of LN<sub>2</sub>, we proposed a cryogenic fracturing technology where LN<sub>2</sub> is used as an assisted fluid to cool the shale gas reservoir and nitrogen is used as a fracturing fluid to form more complex crack system. In this paper, the cracking characteristics between dry and saturated shales after LN<sub>2</sub> injection were studied. The subsequent four conclusions are as follows:

- (1) The clay minerals offer the benefits for the formation of inorganic nanopores. The inorganic nanopores are readily preserved in the process of deposition and conducive to hydrocarbon storage. The siliceous minerals are also favorable for fracturing shale reservoir. The siliceous minerals and the clay minerals account for 62.1 percent of the main components of shale. Therefore, the Upper Ordovician Xinkailing Formation should be further researched for efficient exploration and exploitation
- (2) Although there were muscovite and clay minerals in shale that hydrate with water in pores and macrocracks and expand in volume, the average breakdown pressure difference between dry and saturated shales without LN<sub>2</sub> preconditioning was small. The reason for the phenomenon was the low moisture content of shale. The moisture content of parallel bedding shale was 0.530%, and that of vertical bedding shale was only 0.313%. The average shale mass differential between two bedding directions resulted in varying moisture content. LN<sub>2</sub> preconditioning is beneficial for reducing the breakdown pressure of shale. When

the shale surface experiences violent heat transfer, local thermal stress is generated in the temperature gradient direction, causing the crack development. Meanwhile, the breakdown pressure of dry shale with LN<sub>2</sub> preconditioning decreased 7.12 MPa, 6.06 MPa, 4.58 MPa, and 3.11 MPa, respectively, as the surrounding pressure grew. Therefore, the effect of LN<sub>2</sub> preconditioning was constrained by the surrounding pressure

- (3) After LN<sub>2</sub> preconditioning, the average breakdown pressures of saturated shale decreased by 69.19%, 50.09%, 32.85%, and 21.17% with the confining pressure increasing from 3 MPa to 12 MPa. The frost heave and migration of unfrozen water are conducive to more microcracks generated and weaken the cementation between bedding planes. Therefore, the LN<sub>2</sub> preconditioning effect of saturated shale is better than that of dry shale. Wetting shale around horizontal drilling before LN<sub>2</sub> preconditioning can facilitate the cryogenic damage effect on the breakdown pressure
- (4) The cracking characteristics of vertical bedding shale are influenced by the shale matrix, whereas the bedding plane significantly determines the cracking characteristics of parallel bedding shale. After LN<sub>2</sub> preconditioning, the tensile strength of dry vertical bedding shale was 3.09 times that of dry parallel bedding shale, and the tensile strength of saturated vertical bedding shale was 4.01 times than that of saturated parallel bedding shale. The frost heave increased the tensile strength difference between dry and saturated shales. The breakdown pressure of vertical bedding shale was also higher than that of parallel bedding shale. Therefore, the fracturing direction of drilling should be parallel to the bedding direction in order to achieve better fracturing effect

## Data Availability

The experimental data are available from the corresponding author.

## Conflicts of Interest

There are no conflicts to declare.

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