

The Hole Sealing Technology of Solid–Liquid Materials with Three Pluggings and Two Injections for Gas Extraction Hole in the Coal Mine

Shiyao Yu, Xianbo Su,* Jinxing Song, Qian Wang, and Zhenjiang You*



Cite This: *ACS Omega* 2022, 7, 43847–43855



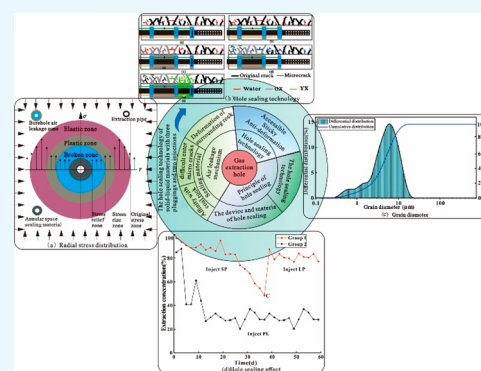
Read Online

ACCESS |

Metrics & More

Article Recommendations

ABSTRACT: The sealing quality of the gas extraction holes determines the extracted gas concentration. Based on this, the paper reveals the basic principle of hole sealing by analyzing the gas leakage mechanism of the borehole. The hole sealing technology of solid–liquid materials with three pluggings and two injections for the gas extraction hole is proposed, and the hole sealing device and material are developed. Through testing the granularity distribution of the solid material, as well as the surface tension and contact angle of the slurry, the hole sealing material that can meet the requirements of accessible, sticky, and anti-deformation is selected. The sealing material enters microcracks and bonds coal rock more easily. First, the solid material is injected for hole sealing. Second, the liquid material can be injected repeatedly to maintain a high concentration for holes with poor sealing and gas concentration attenuation in the late stage of gas extraction. Field tests show that the gas concentration of solid material is 1.3 times that of the conventional material after 30 days of sealing. The liquid material injected after the concentration decline enables the gas extraction concentration to be recovered at 85%.



1. INTRODUCTION

The efficient extraction of gas ensures the safe production of coal mines. It also contributes to clean energy and reduces methane emissions in the process of coal mining.^{1,2} In fact, it can be used as a carbon emission reduction approach for coal enterprises and a positive response to the goal of “carbon peaking and carbon neutrality”. To improve the utilization rate of gas extraction besides the increase in the amount of gas production, the key is to improve the quality of gas extraction. The higher the gas extraction concentration, the higher the utilization efficiency. Due to the low concentration of extracted gas in boreholes, the utilization rate of gas in China was only 44.8% in 2020,^{3,4} which is far below the planned target of 60%.⁵ This is mainly due to the poor sealing quality and serious leakage of gas drainage boreholes.⁶ Therefore, developing a new hole sealing technology will greatly improve the quality of hole sealing. This will reduce the noncarbon dioxide greenhouse gas emission caused by gas drainage. Moreover, improving the quality and quantity of gas extraction in coal mines is crucial for coal enterprises.

Numerous studies have been performed on the quality of hole sealing, mainly focused on three aspects: methodology, material, and technology. The hole sealing technology has experienced three stages, including passive hole sealing, active hole sealing, and compensation hole sealing. Passive sealing of the annulus gap between the extraction pipe and the borehole

can be traced back to the 19th century. This is suitable for the compact rock section without developed fractures around the borehole. The borehole with developed microfractures around the borehole lacks a sealing ability and cannot support the high concentration gas extraction at the later stage. Based on the leakage characteristics of the surrounding rock fractures, the active sealing technology of “solid sealing liquid and liquid blocking gas” (capsule-sealed liquid sealing hole) is proposed.^{7–9} In this approach, the sealing fluid with a pressure higher than the gas pressure is injected between the expansion capsules. The sealing fluid filfers into the cracks around the borehole under pressure to prevent gas leakage. However, due to the compressive strength of the gas drainage pipe and the surface tension of the slurry material, it is still difficult to seal microcracks. In recent years, in order to solve the problems of gas concentration attenuation due to borehole deformation and instability,^{10,11} secondary compensation hole sealing technologies, including secondary grouting and secondary powder sealing methods, have been proposed.^{12–16} The

Received: August 5, 2022

Accepted: October 31, 2022

Published: November 23, 2022



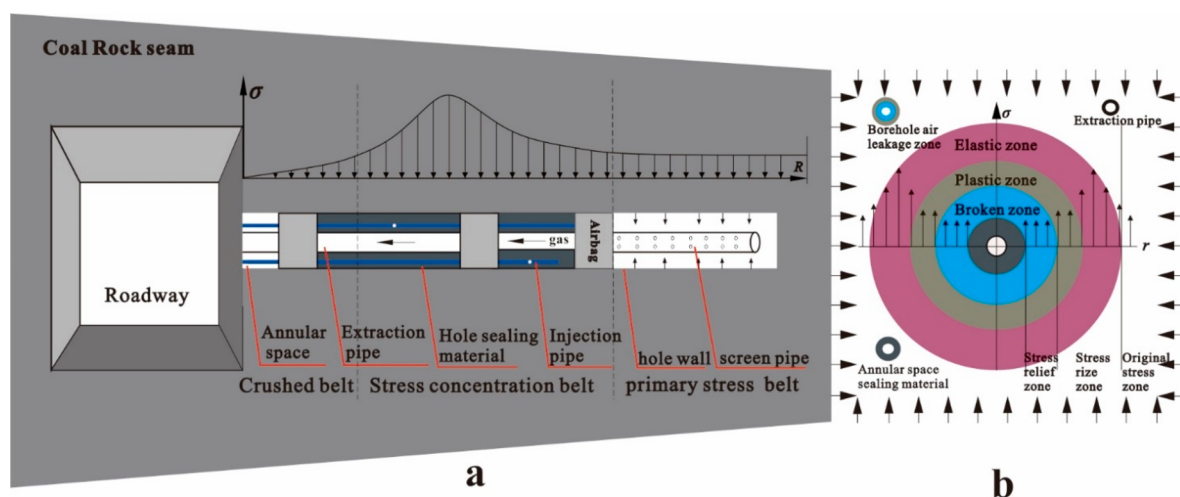


Figure 1. Schematic diagram of stress field distribution and hole-sealing structure around the borehole: (a) axial stress distribution; (b) radial stress distribution.

secondary sealing improves the sealing effect to a certain extent. However, because the deformation of surrounding rock is a dynamic process, fracture development is time-dependent, which has higher requirements for sealing materials and processes.

Currently, the widely used sealing materials mainly include cement-based (CB) and polymer organic materials.^{17–24} Cement is the first material used to replace the yellow mud sealing material. After years of continuous optimization, it efficiently solves water segregation and consolidation fissuration problems. Therefore, it has become the most widely used sealing material.²⁵ Barkman and Abrams proposed the famous one-third rule: the particles with sizes equal to or larger than 1/3 of the mean pore size can virtually plug cracks, namely the “3D principle”.²⁶ Due to the large particle size of the existing CB material, it is difficult to plug the fine cracks of coal strata. In addition, the material can easily form damage cracks under the condition of dynamic deformation of the borehole surrounding rock, which results in the decrease of the gas extraction concentration. Based on the defects of CB materials, several studies are performed on polymer materials such as polyurethane (PU). Nowadays these polymer materials are showing improved performance. However, the difficulty in entering microcracks due to large capillary force becomes the largest disadvantage of this material.²⁷ Not surprisingly, the defects of materials are the main reasons for poor sealing effect and low gas concentration.

The research and development of sealing technology is another key point. The sealing technologies mainly include single plugging, one plugging, and one injection, as well as two pluggings and one injection.^{28–30} At present, two pluggings and one injection with pressure sealing holes are the most widely used. This process has a high sealing performance, which can meet the needs of gas extraction under certain geological conditions. However, most of the mines still face the challenges of unstable extraction concentration and fast attenuation.

In the past decades, several studies tackled the sealing methodology, material, and technology of boreholes. Different sealing technology systems have been developed and applied in different mines. However, each sealing material and process has its own limitations. Therefore, a universal sealing technology does not exist. In this paper, through the analysis

of the sealing mechanism, a hole sealing technology of solid–liquid materials with three pluggings and two injections for gas drainage holes is proposed. In addition, two types of solid–liquid sealing materials with safety and environmental protection, high viscosity, and low capillary force are developed. The field test at the bottom drainage roadway 16101 of Jiaozuo Coal Mine A, Henan (China), demonstrates that the proposed technology can achieve the goal of continuous high concentration gas extraction.

2. THE BASIC PRINCIPLES OF HOLE SEALING TECHNOLOGY OF SOLID–LIQUID MATERIALS WITH THREE PLUGGINGS AND TWO INJECTIONS

2.1. Air Leakage Mechanism of Gas Extraction Borehole.

Boreholes can be approximated by small roadways. Gas extraction is performed after drilling by inserting drainage pipes and annular grouting between the drainage pipes and borehole wall in the sealing section (Figure 1). If the sealing effect of the borehole is poor, air infiltration in the extraction process will cause sealing failure. Therefore, it is necessary to analyze the mechanism of borehole leakage.

The position of the sealing hole is in the terrane of the cross-layer borehole or in the coal seam of the bedding borehole. The virgin state of stress of the surrounding coal rock is destroyed by a drilling dynamic disturbance during the construction of the roadway and borehole, while the ambient stress is redistributed after drilling. According to the distribution characteristics of the stress field and elastic–plastic theory, “three belts and four zones” are formed around the borehole.^{31–34} That is, from the coal (rock) wall of the roadway to its deep part, the axial direction of the borehole, the fracture zone, stress concentration belt, and original rock stress belt are successively presented (Figure 1a), corresponding to the fracture zone, plastic zone, elastic zone, and original rock stress zone along the radial direction of the borehole, respectively (Figure 1b). The surrounding rock in the fracture zone is highly fractured, and the internal cracks rapidly develop and connect, which forms the stress reduction zone. Although the stress concentration belt has a plastic deformation, it will produce new cracks. However, it is compacted due to the increase of stress and poor permeability, which consists of the best position for hole sealing. Cracks developed accurately in

the fracture zone and plastic zone, and the permeability is satisfactory. This is the main channel of drilling leakage, known as the “leakage circle” of the borehole.^{35,36} Due to the coarse particle size and high capillary pressure of the sealing material, it is difficult to enter the cracks in the surrounding rock, which results in insufficient sealing range. The airbag is rigid and easy to fold, the coal-friendly lithology of the sealing material is poor, and insufficient adhesion with the hole wall causes air leakage. The air in the roadway enters the drainage pipe under the action of negative pressure drainage, which results in the decrease of gas concentration.

When the borehole surrounding rock creeps occurs under the action of in situ stress, the cracks in the borehole surrounding rock continue to proliferate and penetrate, with the extrusion of the deformation pressure of the surrounding rock, and the strength of the sealing material becomes not enough to resist. Therefore, the sealing material deforms and produces new cracks due to the low deformation resistance. The deformation has a time effect, while the new cracks continue to generate and connect to form a new air leakage channel. It can be deduced that the poor deformation resistance of the sealing material is another reason for air leakage in the middle and late stages of drilling extraction. In addition, with the extraction and gas desorption, the coal shrinks and the crack opening increases, which further increases the degree of leakage.

2.2. Principle of Hole Sealing. The annulus and borehole surrounding rock fractures should be closed for hole sealing, involving rock mass mechanics, mechanics of granular media, surface physical chemistry, and other disciplines. All the sealing materials can be used for annular sealing, including the cement-based materials, while adding an appropriate amount of expansive agent and water reducing agent does not lead to dehydrating shrinkage. However, because the borehole is located in the redistribution of the mining stress field after mining, the development of cracks around the borehole caused by the dual stress of roadway and borehole hardens the hole sealing. In addition, the length of the sealing section is longer than the stress rise area of the borehole axial. The performance of the sealing materials is crucial for ensuring a high sealing quality and improving the gas extraction concentration. Therefore, higher requirements are put forward for the sealing materials.

The sealing material must have certain expansibility and compactness to avoid the pore fissure due to its contraction or looseness. Moreover, the sealing material has also the characteristics of accessibility, sticking, and anti-deformation. Note that accessibility belongs to the surface physical chemistry and powder mechanics, which indicates that the sealing “material” should simultaneously meet the low capillary pressure and 3D principles. The capillary pressure of the sealing material with low surface tension is low.³⁷ The finer the particle size, the easier it is to enter the micro cracks. Only when the sealing is under pressure can it efficiently enter the microcracks of the surrounding rock, ensure the sealing of the fissured circle, and improve the quality of the sealing hole. The “sticking” belongs to the category of surface physical chemistry, which indicates that the sealing material has a small contact angle with the coal rock surface around the borehole. That is, it has a good affinity with coal rock, and it is well bonded with the borehole wall and the fracture surface without leaving the air leakage channel. The “antideformation” belongs to the category of rock mass mechanics, which indicates that the

strength of the sealing material does not deform, crack, or leak air when subjected to the deformation and extrusion of the borehole surrounding rock, and it can play a supporting role for the borehole. However, at present, it is difficult for the deformation resistance of almost all of the solid-phase sealing materials to compete with the in situ stress. In addition, it is not sufficient to efficiently resist the deformation of the surrounding rock in the late stage of extraction. The liquid colloidal material with low capillary pressure is an invisible material that can change with deformation. It has the function of dynamic sealing cracks without the influence of drilling deformation. The liquid material with good fluidity is injected under pressure after the deformation of the surrounding rock, and the solid material produces new cracks. The water locking effect can be used to timely and efficiently seal the cracks that are difficult for solid-phase materials to enter and the new cracks, and it can also improve the concentration of gas extraction.³⁸

2.3. The Hole Sealing Technology of Solid–Liquid Materials with Three Pluggings and Two Injections.

2.3.1. The Device and Materials of Hole Sealing. Based on the mechanism of gas leakage and principles of hole sealing for the gas extraction borehole, the hole sealing technology of solid–liquid materials with three pluggings and two injections is proposed. The hole sealing device and material are developed. The hole sealing device with three pluggings and two injections consists of three airbags, four check valves, one squib valve, and two grouting tubes (Figure 1). The airbag is made of a special nylon cloth, which is thin and elastic and only water is allowed to pass through. There is no fold in the borehole, and the two ends of the sealing section are filled by efficient expansion due to the elasticity in the grouting process, which provides a good sealing environment for the middle grouting section.

The solid phase (SP) hole sealing material consists of the CB material and alkaline modifier A, made of 1250 mesh superfine cement, accelerating agent, water reducing agent, defoaming agent, and expansive agent. The liquid phase (LP) hole sealing material consists of water-soluble glue material (WSG), water retaining agent (WRA), and modifier B. Modifiers A and B have good hydrophilicity. LP is a type of viscous colloid, which has the function of dynamic sealing cracks and is not affected by borehole deformation. In addition, WRA has a large number of amide and carboxyl hydrophilic groups. It is a good water-retaining agent that can absorb water in large quantities by using the osmotic pressure generated by the difference of concentration between ions and groups in resin and the components related to the aqueous solution and the affinity between polymer electrolyte and water. Therefore, LP has a good water retention effect and a low volatilization rate. It can maintain long-term pressure sealing. Simultaneously, WRA in the slurry can absorb water from the coal seam seepage, which reduces the evaporation of slurry water to a certain extent. After evaporation and drying, the residual solid matter is very little (5% of the total liquid), which does not affect the subsequent filling slurry and continuously maintains the borehole tightness. SP can support the surrounding rock of the borehole and resist the deformation of the borehole in the process of extraction, only by keeping the extraction borehole unblocked can continuous extraction be realized. LP is designed to effectively block microcracks caused by creep deformation in the sealing section at the later stage of extraction. Whether the properties

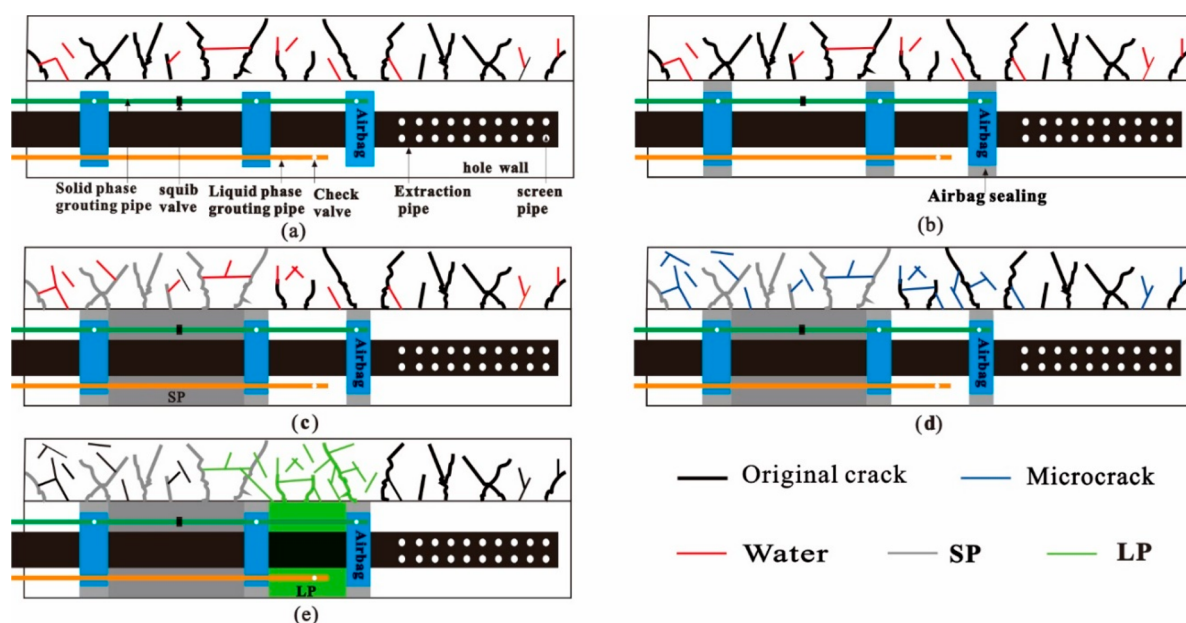


Figure 2. Schematic diagram showing the device and technology of solid–liquid materials with three pluggings and two injections: (a) before the hole sealing material is injected; (b) three airbags are filled; (c) solid-phase material is injected; (d) the water in the microcrack is drained; (e) liquid-phase material is injected.

Table 1. Basic Data of Coal Samples^a

R _o /%	Porosity/%	Pore volume /(cm ³ /g)					Pore volume ratio/%				Proximate analysis/%			
		V ₁	V ₂	V ₃	V ₄	V _t	V ₁ /V _t	V ₂ /V _t	V ₃ /V _t	V ₄ /V _t	Mad	Aad	Vad	FCad
3.3	4.732	0.019	0.008	0.001	0.009	0.037	51.23	20.98	2.72	25.07	0.63	8.30	12.22	78.85

^aV₁ micropore (3 nm < pore size < 10 nm); V₂ foraminule (10 nm < pore size < 100 nm); V₃ mesopore (100 nm < pore size < 1000 nm); V₄ macropore (pore size >1000 nm); V_t total pore volume; M ad-moisture; A ad-ash yield; V ad-volatile matter; FC ad-fixed carbon; R_o ran-vitrinite random reflectance

of sealing materials can meet the requirements of accessible, sticking, and anti-deformation needs further experimental research.

2.3.2. The Hole Sealing Technology. The hole sealing technology of solid–liquid materials with three pluggings and two injections includes four main links: the extraction pipe and hole sealing device (airbag and grouting pipe, etc.) are placed into the borehole, low-pressure injection of SP slurry into the airbag to form temporary plugging, high-pressure injection of SP into the annulus between the airbag to form a primary hole sealing, and high-pressure injection of LP to form a secondary hole sealing (Figure 2). The first consists in connecting the gas extraction pipe and the airbag and sending it to the specified position in the borehole (Figure 2a). According to the distribution of the mining pressure field, the sealing section should be selected in the area where the relatively dense and stress of the surrounding rock increases. Moreover, when the grouting pressure reaches 0.6 MPa of the opening pressure of the check valve, SP is injected into the solid grouting pipe to fill three airbags, and three “plugs” are generated with pressure at the predetermined hole sealing position (Figure 2b). The squib valve opens with the continuous increase of the grouting pressure and SP enters the surrounding rock fracture under a high pressure of 1.5 MPa to complete the primary hole sealing (Figure 2c). Finally, gas extraction is carried out, and regular detection of gas concentration, when the concentration is reduced to less than 30%³⁹ and Inject LP under 1.5 MPa high-pressure through liquid grouting pipe completes secondary

sealing (Figure 2e), further seal the air leakage cracks. Regular detection of the gas drainage concentration is performed for extraction boreholes that have completed the secondary sealing and the leakage hole several times to fill LP materials. The clearest advantage of this process is that as long as there is a flow in the gas extraction borehole, LP can be injected multiple times to maintain the gas concentration up to standard.

3. EXPERIMENTAL STUDY

3.1. Experimental Material. The CB is made up of 1250 mesh superfine cement, accelerating agent, water reducing agent, defoaming agent, and expansive agent. A viscous colloid composed of WSG and WRA can dynamically seal cracks. Alkaline modifier A and modifier B both have good affinity with coal rock and are used to reduce the surface tension and contact angle of SP and LP materials, respectively. The coal samples, mudstones, fine sandstones, carbonaceous mudstones, and silty sandstone used in the experiment are taken from the coal seam and its floor of Jiaozuo Coal Mine A. The 80–120 mesh pulverized different coal rock was pressed into a number of small diameter briquettes with a thickness of 0.5 cm and a diameter of 1.5 cm with a coal press. Some coal pillars with a diameter of 2.5 cm and a length of 5 cm were drilled from the coal sample. The samples were dried and sealed for later experiments. The pore crack development characteristics of coal have a great influence on the quality of sealing holes. According to the national standard (GB/T21650.1-2008), the porosity and pore size distribution of coal samples were

measured by AutoPoreIV9510 mercury injection apparatus, and appropriate coal samples were taken for industrial analysis according to ISO 17246–2010 (Table 1).

3.2. Test Method for Material Properties. **3.2.1. Granularity Distribution Test.** The granularity distribution of 1250 mesh superfine cement is measured using an LS-POP (9) laser granularity analyzer. The instrument uses the scattering of light by particles, and the granularity distribution is calculated according to the scattered light energy distribution. The scope of the test is 0.1–750 μm , the test time is 1–2 min, and the work environment is 5–35 $^{\circ}\text{C}$ (temperature), <85% (relative humidity). Before each test, the instrument is with distilled water to ensure the accuracy of the test.

3.2.2. Measurement of Surface Tension and Contact Angle of Sealing Slurry. A JC2000D contact angle measuring instrument is used to measure the surface tension of the sealing slurry and the contact angle of different coal rocks. The surface tension measurement range is 0–2000 $\text{mN}\cdot\text{m}^{-1}$, measurement accuracy is 0.01 $^{\circ}$, and contact angle measurement range is 0–180 $^{\circ}$. The pendant drop method was used to measure the surface tension of the sealing slurry, and the shape of a drop was measured at the moment a pendant drop fell. The image contour of the drop was fitted with a computer, and the surface tension of the sealing slurry was calculated.⁴⁰

The contact angle measuring method was used to measure the contact angle of the sealing slurry. First, the briquettes with a flat surface and uniform thickness were selected as samples. When the droplet stays on the surface of the sample for 5–10 s, the sealing slurry hanging on the sample can be captured by an optical system, and then the contact angle between the droplet and the sample can be calculated through image analysis software. In turn, the contact angle between sealing grout with (without) modifier and different coal rock was tested.⁴¹

3.2.3. Hole Sealing Quality Test. The experimental setup consists of a high-pressure helium gas cylinder, a pressure gauge, a sample holder, and a flow meter (Figure 3b). It is used

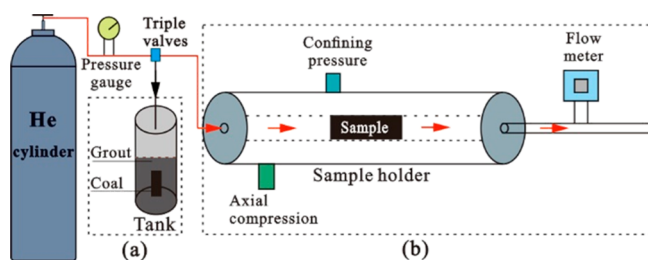


Figure 3. Experimental apparatus to test the airtightness of hole sealing material: (a) grouting device; (b) airtightness device.

to study the sealing quality of sealing material to crack of coal pillars. After assembling and checking the air tightness of the device, the coal pillar is loaded into the sample holder. Confining pressure and axial pressure of 3 MPa, respectively, are applied to the holder. The cylinder pressure is set 2 MPa, and then the valve is opened. The permeability of all of the coal pillars is measured, and two groups of coal pillars with similar permeability are selected. One group consists of coal pillars 1 and 2, the other group consists of coal pillars 3 and 4 (Figure 4). SP with or without modifier A is injected into coal pillars 1 and 2, respectively. LP with or without modifier B is injected into coal pillars 3 and 4, respectively.



Figure 4. Four coal pillars after the experiment.

Four high-pressure tanks with good air tightness (Figure 3a) are prepared, named I, II, III, and IV corresponding to coal pillars 1, 2, 3, and 4, and the coal pillars are put into the corresponding high-pressure tanks, respectively. Four kinds of grout in turn are loaded into the high-pressure tank, and all of the high-pressure tanks are tightened. 1.5 MPa high-pressure helium gas is injected. Once grouting begins, grout will enter the fracture of coal under the action of gas pressure. The filling time of SP is 30 min (the solidification time is 40 min about SP), and the filling time of LP is 3 h. After the grouting, the coal pillar is removed, and the residual grout from the surface of the coal pillar is removed and placed into the sealing bag to timely test the permeability of coal pillars 3 and 4. The permeability test was conducted for coal samples 1 and 2 after drying. The permeability of coal pillars before and after grouting is compared to evaluate the sealing property of the sealing material. Helium gas is used because it is not adsorbent, and 1.5 MPa gas pressure depends on the site grouting pressure.

3.3. Experimental Results and Analysis. **3.3.1. Granularity Distribution Test.** The laser granularity test results (Figure 5) show that the minimum granularity of 1250 mesh

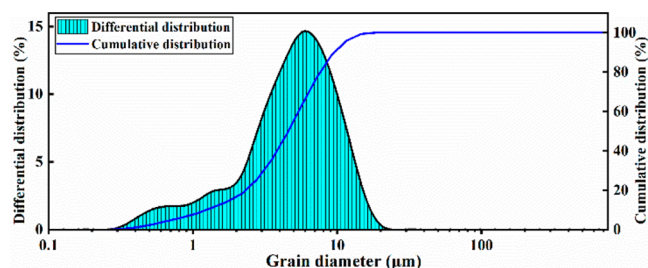


Figure 5. Particle size distribution of 1250 mesh ultrafine cement.

superfine cement is 0.325 μm , the maximum size is 18.598 μm , and the peak value is 5.671 μm . D10, D50, and D90 values are 1.236, 4.581, and 9.580 μm . The granularity of ultrafine cement that is less than 5 and 10 μm reaches 50% and 90%, respectively.

SP is composed of 1250 mesh superfine cement as well as other additives. According to the 3D principle, most of the ultrafine cement can enter the microcracks with a crack width of more than 15–30 μm , which can block the smaller microcracks compared with the ordinary cement.

3.3.2. Wettability Test Results. The wettability of SP and LP materials is also tested. After adding 0.3% alkaline modifier A, the surface tension of SP decreases from 34.25 to 26.23 $\text{mN}\cdot\text{m}^{-1}$.

Table 2. Wettability Test Results of Sealing Material

Grout type	Surface tension / (mN m ⁻¹)	contact angle / (deg)				Coal	Pore size range/nm
		Siltstone	Packsand	Mudstone	Carbonaceous mudstone		
CB	34.25	53.50	55.90	72.00	50.01	68.25	
CB+0.3% A	26.23	31.75	24.75	22.75	15.50	21.75	
WSG+WRA	39.97	60.00	66.25	63.63	61.00	45.00	42–75
WSG+WRA+0.05% B	21.58	26.34	28.50	30.50	21.75	22.75	50–54
Water	74.99					55.50	115

m⁻¹, and the contact angle with different coal rocks decreases from 50.01°–72.00° to 15.50°–31.75°. After adding 0.05% modifier B to LP, the surface tension decreases from 39.97 to 21.58 mN·m⁻¹, and the contact angle with different coal rocks decreases from 45.00°–66.25° to 21.75°–30.50° (Table 2).

The addition of the hydrophilic modifiers A and B significantly reduces the surface tension of the two-phase sealing material and the contact angle with coal rock, then reduces the capillary pressure (Laplace–Young eq 1),⁴² and enhances the affinity with coal rock of the two-phase sealing material. Although the capillary pressure of the SP is low, the influence of the particle size is given priority, according to the 3D principle. When the grouting pressure is 1.5 MPa, LP can overcome the capillary pressure into the smaller pore size range of different coals rocks (50–54 nm), which is far lower than the size (15–30 μm) of SP material that can enter coal rocks cracks. This shows that the SP and LP sealing materials have the characteristics of accessibility, sticking, and anti-deformation

$$p_c = \frac{2\sigma \cos \theta}{r} \quad (1)$$

where p_c is capillary pressure, MPa; σ is surface tension between the solution and air, mN m⁻¹; θ is contact angle between the solution and rock sample, °; and r is pore radius of the capillary tube, nm.

In the initial stage of gas extraction, the existence of water in the cracks of the surrounding rock of the borehole will prevent LP from entering the microcracks (Figure 2a). This is also the reason why the method of two pluggings and one injection is not used to directly inject LP sealing hole. First, the cracks above 15–30 μm must be sealed with SP. By this time, the microcracks will not leak air due to water sealing (Figure 2c). After a period of time, the water in the microcracks is pumped dry (Figure 2d). With the passage of extraction time, the creep of the surrounding rock of the borehole leads to the development of cracks and the increase of air leakage channels. After the concentration of extracted gas drops, the LP can smoothly enter smaller microcracks above 50–54 nm driven by grouting pressure and play a sealing role (Figure 2e). When LP is dried with extracted, the LP can be repeatedly injected in the extraction process to meet the sealing of the whole life period of the extraction borehole and realize a high concentration gas extraction.

3.3.3. Sealing Quality Test Results. The permeability of coal pillars (1,2,3,4) before and after grouting is tested, and the permeability of coal pillars after grouting is reduced by 53.85%, 29.17%, 47.37%, and 35.00%, respectively (Table 3). The results indicate that SP and LP have an obvious sealing effect on the fractures in coal rock. After grouting, coal pillars 1 and 3 with modifiers A and B, respectively, have lower permeability than those coal pillars 2 and 4 without modifier (Figure 6). It was further shown that the capillary pressure of the sealing

Table 3. Permeability Comparison of Coal Samples before and after Sealing

Coal pillar	1	2	3	4
Permeability before sealing/mD	0.26	0.24	0.19	0.20
Permeability after sealing/mD	0.12	0.17	0.1	0.13
Permeability reduction rate/%	53.85	29.17	47.37	35.00

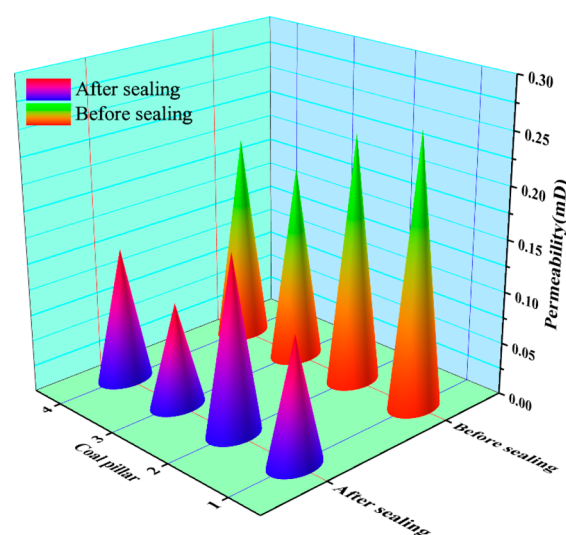


Figure 6. Permeability comparison of coal samples before and after sealing.

material is reduced by adding modifier, easier to get into the fissure, and meeting the requirements of accessibility. Furthermore, the modifier has a good affinity with coal rock, so that the sealing material can be more closely bonded with the fissure wall, reduce air leakage passages, and meet the requirements of sticking.

4. FIELD APPLICATION

4.1. Engineering Test Conditions. In order to validate the proposed sealing method of three pluggings and two injections, and the sealing effect of SP and LP materials, a field test is carried out in the bottom drainage roadway 16101 of coal mine A in Jiaozuo, China. The II₁ coal seam is mainly mined in the covered coal seam of the roadway, has a coal seam inclination of 127°, an inclination of 10°, an average thickness of 6.5 m, an original gas content of 21.46 m³/t, and a pressure of 1.62 MPa.

Two drilling sites are selected in the test area, with 6 boreholes in each group, and the spacing of the drilling fields is 6 m (Figure 7). The mine uses PU as the sealing material for plugging an injection of the whole rock sealing method. However, the sealing cost is high, the concentration of gas extraction rapidly decreases, and the sealing effect is not ideal.

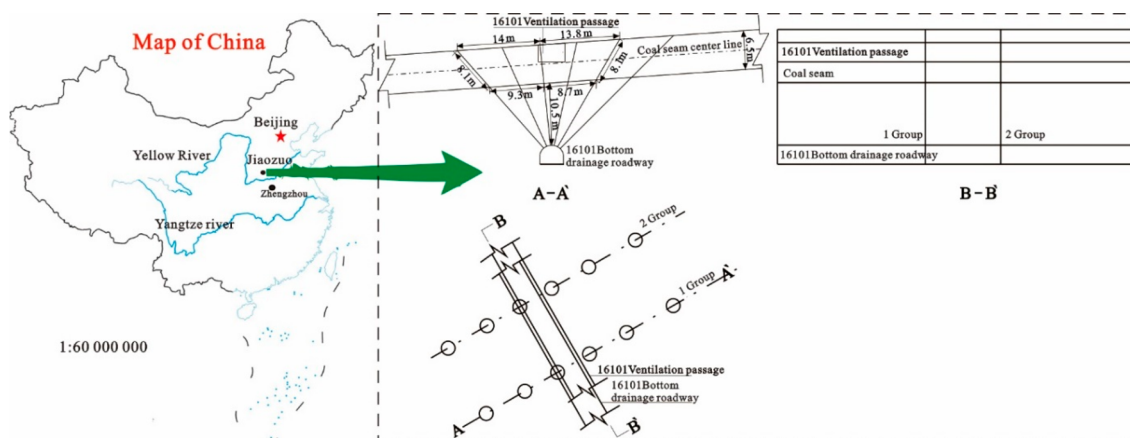


Figure 7. Schematic diagram of bottom drainage roadway 16101 drainage borehole layout.

To compare the sealing effect of different materials and technology, the first group is selected as the experimental drilling site, and the three-pluggings and two-injections solid-liquid sealing device, materials, and technology are used. The second group is the comparative drilling site, using the traditional hole sealing method and material of the mine. First, the drilling-stamping integrated gas increase production technique is used to improve the permeability of the coal seam about two groups.⁴³ Second, gas extraction is implemented after hole sealing. The sealing position is determined according to the stress field distribution and lithology of the rock section. In addition, the hole is sealed in the stress concentration zone and relatively dense lithology of the sealing section. The rock section has a length of 10–13 m. Therefore, the length of the solid sealing hole in group 1 is 5 m and that of the liquid phase is 1 m, which is close to the coal seam floor. The gas concentration is observed in real-time, and the sealing effects of SP and PU are compared. The LP is injected when the gas concentration is significantly reduced or less than 30% in group 1, and a change of gas concentration is observed.

4.2. Test Results and Analysis. After sealing, gas extraction is performed. In the initial stage (8 days), the gas concentration of the two groups of boreholes is high. This is due to the weak deformation of the borehole and the existence of water in the coal rock mass, which plays a water lock plugging role.⁴⁴ Under a capillary pressure of 0.74 MPa, water can also enter the microcracks of coal with a width of 115 nm (Table 2). Therefore, the existence of water in the microfractures can also block the fractures below a certain width and therefore inhibits gas production.⁴⁵ Afterward, the concentration of group 2 rapidly decreases to point A; this shows that the water in the microcracks is extracted by gasification and the water locking effect is invalid. Consequently, the gas migration occurs in the part of the fracture blocked by water. Therefore, the concentration increases to point B and then decreases. When the extraction time passes, the borehole deformation is gradually strengthened, and new fractures around it are continuously generated, which results in the increase of gas leakage channels and the decrease of gas concentration. The test results demonstrate that the attenuation of gas concentration in group 2 is faster than that in group 1 (Figure 8). After 30 days of sealing, the gas concentration of group 1 remains at 67.8%, while that of group 2 is only 29.4%. It can be seen that the sealing effect of the

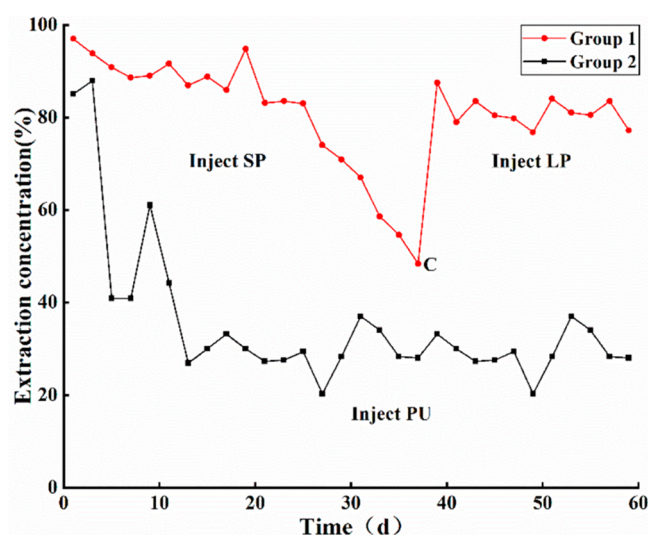


Figure 8. Comparison of gas extraction concentration between two groups of boreholes.

pure SP is better than that of PU because ultrafine particles are more likely to enter the microcracks than PU with high capillary pressure (i.e., surface tension of $60 \text{ mN}\cdot\text{m}^{-1}$). The gas concentration of group 1 decreases. After LP injection at point C (Figure 8), the gas extraction concentration is recovered to 85% of the higher concentration. LP can maintain the gas concentration for more than 20 days without significant attenuation, which proves that LP has a high sealing performance. Therefore, the sealing effect of the hole sealing technology of solid-liquid materials with three pluggings and two injections is better than that the traditional technology. Finally, the effect of gas extraction can be improved.

5. CONCLUSIONS

This paper is based on the distribution characteristics of the borehole stress field and fracture field, and the analysis shows that the annulus and surrounding cracks are the main channels of air leakage. The sealing material should meet the new requirements of accessibility, viscosity, and deformation resistance.

A hole sealing technology of solid-liquid materials with three pluggings and two injections for gas extraction borehole is proposed. The SP has a smaller granularity distribution and

can enter fractures with smaller pore sizes. The SP and LP have lower capillary pressure and a stronger affinity with coal rock; they can seal smaller microcracks and meet the new requirements of hole sealing materials.

The field test shows that SP can maintain a high concentration of gas extraction for a longer time than the traditional PU. After the secondary filling of LP, the attenuated gas concentration is increased again, and the attenuation level is maintained at a low level. It can also be seen that the proposed approach will provide support for the increment of gas extraction quality. Finally, it can highly improve the gas utilization rate and enhance non-carbon dioxide greenhouse gas emission reduction.

AUTHOR INFORMATION

Corresponding Authors

Xianbo Su – Institute of Resources and Environment, Henan Polytechnic University, Jiaozuo 454003, China; Unconventional Gas Research Institute, Henan Polytechnic University, Jiaozuo 454003, China; School of Energy Resources, China University of Geosciences, Wuhan 430074, China; Collaborative Innovation Center of Coalbed Methane and Shale Gas for Central Plains Economic Region, Jiaozuo 454003, China; orcid.org/0000-0003-2976-164X; Email: suxianbo@hpu.edu.cn

Zhenjiang You – Center for Sustainable Energy and Resources, Edith Cowan University, Joondalup, WA 6027, Australia; School of Chemical Engineering, The University of Queensland, Brisbane, QLD 4072, Australia; orcid.org/0000-0002-4843-2107; Email: zhenjiang.you@gmail.com

Authors

Shiyao Yu – Institute of Resources and Environment, Henan Polytechnic University, Jiaozuo 454003, China; Center for Sustainable Energy and Resources, Edith Cowan University, Joondalup, WA 6027, Australia

Jinxing Song – School of Energy Science and Engineering, Henan Polytechnic University, Jiaozuo 454003, China; Collaborative Innovation Center of Coalbed Methane and Shale Gas for Central Plains Economic Region, Jiaozuo 454003, China

Qian Wang – Institute of Resources and Environment, Henan Polytechnic University, Jiaozuo 454003, China; orcid.org/0000-0003-4340-7989

Complete contact information is available at: <https://pubs.acs.org/10.1021/acsomega.2c05001>

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China [grant nos. 42072193, 41872176] and the Natural Science Foundation of Henan Province [grant no. 222300420173]. We are also grateful for the constructive comments by reviewers and the editor on an earlier draft of this manuscript.

REFERENCES

(1) Wang, Z.; Hurter, S.; You, Z.; Honari, V.; Sun, Y.; Zhang, S. Influences of negative pressure on air-leakage of coal seam gas extraction: Laboratory and CFD-DEM simulations. *J. Pet. Sci. Eng.* **2021**, *196*, 107731.

(2) Wang, C.; Guo, Z.; Zhang, L.; Kang, Y.; You, Z.; Li, S.; Wang, Y.; Zhen, H. 3D Fracture Propagation Simulation and Pressure Decline Analysis Research for I-Shaped Fracture of Coalbed. *Energies* **2022**, *15* (16), 5811.

(3) Yang, F.; Xu, H. Analysis of the development path of ecological environmental protection and comprehensive utilization of resources in the coal industry in the 14th Five-Year Plan. *China Coal* **2021**, *47* (05), 73–82.

(4) Fan, W.; Yang, W. Innovative utilization and practice of coal mine gas. *Coal Process Comprehension Utilization* **2022**, *4*, 99–103.

(5) Zhang, Z.; Huo, C. Research progress of CBM utilization technology in mining areas. *Min. Saf. Environ. Prot.* **2022**, *49*, 59–64.

(6) Zhou, F.; Sun, Y.; Li, H.; Yu, G. Research on the theoretical model and engineering technology of the coal seam gas drainage hole sealing. *J. China Univ. Min. Technol.* **2016**, *45* (3), 433–439.

(7) Wu, Q.; Zhou, S. Study on the direct measurement of coal seam gas pressure in coal roadway by capsule-sealing liquid sealing technique. *Saf. Coal. Min.* **1994**, *25* (8), 7–9.

(8) Wang, H.; Wang, E.; Li, Z.; Shen, R.; Liu, X. Study and application of a new gas pressure inversion model in coal seam while drilling based on directional drilling technology. *Fuel* **2021**, *306*, 121679.

(9) Zhou, S. A new method for measuring gas pressure in coal seam. *Saf. Coal. Min.* **1983**, No. 9, 5–8.

(10) Wang, Z.; Sun, Y.; Wang, Y.; Zhang, J.; Sun, Z. A coupled model of air leakage in gas drainage and an active support sealing method for improving drainage performance. *Fuel* **2019**, *237*, 1217–1227.

(11) Duan, K.; Wu, W.; Kwok, C. Y. Discrete element modeling of stress-induced instability of directional drilling boreholes in anisotropic rock. *Tunn. Undergr. Space Technol.* **2018**, *81*, 55–67.

(12) Gao, B.; Ren, C.; Kang, W.; Chen, L.; Dong, Q. Study on secondary grouting technology for coal seam gas extraction failure in borehole pipe bottom. *Coal Sci. Technol.* **2021**, *49* (5), 151–158.

(13) Li, H.; Wang, W.; Liu, Y.; Ma, J.; Gao, H. An integrated drilling, protection and sealing technology for improving the gas drainage effect in soft coal seams. *Energy Reports* **2020**, *6*, 2030–2043.

(14) Wang, H.; Wang, E.; Li, Z.; Wang, X.; Ali, M. Study on sealing effect of pre-drainage gas borehole in coal seam based on air-gas mixed flow coupling model. *Process Saf Environ. Prot.* **2020**, *136*, 15–27.

(15) Wang, Z.; Zhou, Y.; Sun, Y.; Wang, Y. Novel gas extraction borehole grouting sealing method and sealing mechanism. *J. China Coal. Soc.* **2015**, *40* (3), 588–595.

(16) Zhang, C.; Lin, B.; Zhou, Y.; Zhai, C.; Wu, H.; Hao, Z. Strong-weak-strong borehole pressurized sealing technology for horizontal gas drainage borehole in mining seam. *J. Min. Saf. Eng.* **2013**, *30* (6), 935.

(17) Cheng, J.; Zhao, G.; Liu, Y.; Tang, H. Study on the properties of non-solidified materials and intelligent hole sealing device for gas drainage drilling. *Coal Sci. Technol.* **2020**, *48* (2), 131–135.

(18) Liu, J.; Tang, T.; Lu, T.; Ji, X.; Qian, L. Activation effect of the fly ash with nano silicon nitride on the early strength properties of the mining-left cement sealing materials. *J. Saf. Environ.* **2020**, *20* (5), 1754–1757.

(19) Wu, Z.; Fan, D.; Jiang, S.; Shao, H.; Wang, K.; Zhang, W.; Guo, C. A wireless communication based measurement of gas pressure with capsule-slime sealing device. *Int. J. Min. Sci. Technol.* **2019**, *29* (6), 917–923.

(20) Xu, C.; Zhang, H.; Kang, Y.; Zhang, J.; Bai, Y.; Zhang, J.; You, Z. Physical plugging of lost circulation fractures at microscopic level. *Fuel* **2022**, *317*, 123477.

(21) Yang, W.; Jia, R.; Li, X.; Guo, M.; Lin, B.; Lin, M. Theory and technology of “blasting injection” integrated outburst prevention in coal face. *J. China Univ. Min. Technol.* **2021**, *50*, 764–775.

(22) Zhang, T.; Bao, R.; Li, S.; Zhang, C.; Zhang, L.; Jiang, X. Experimental study on expansion and creep characteristics of new CF sealing material. *J. Min. Saf. Eng.* **2019**, *36* (1), 175–183.

- (23) Zhai, C.; Xu, J.; Xiang, X.; Zhong, C. Flexible gel (FG) for gas-drainage drilling sealing material based on orthogonal design. *Int. J. Min. Sci. Technol.* **2015**, *25* (6), 1031–1036.
- (24) Zhou, F.; Xia, T.; Liu, Y.; Hu, D.; Liu, C. Study of gas solid coupling model on transport properties of secondary sealing's powder particles. *J. China Coal. Soc.* **2011**, *36* (6), 953–958.
- (25) Fu, J.; Wang, D.; Li, X.; Wang, Z.; Shang, Z.; Jiang, Z.; Wang, X.; Gao, X. Experimental Study on the Cement-Based Materials Used in Coal Mine Gas Extraction for Hole Sealing. *ACS omega* **2021**, *6* (32), 21094–21103.
- (26) Yang, Y.; Cheng, T.; Liu, H.; You, Z.; Hou, J. Oil Displacement Performance Using Bilayer-Coating Microspheres. *Ind. Eng. Chem. Res.* **2021**, *60* (5), 2300–2313.
- (27) Zhou, A.; Wang, K. A new inorganic sealing material used for gas extraction borehole. *Inorg. Chem. Commun.* **2019**, *102*, 75–82.
- (28) Li, H.; Guo, S.; Chen, H. Application of coal-powder borehole-sealing material in borehole-sealing engineering. *EMERG MATER RES* **2019**, *8* (2), 290–296.
- (29) Yang, H.; Ren, F.; Wang, Z.; Chen, S. Quality inspection and quantitative evaluation method for borehole sealing in gas drainage. *J. China Coal. Soc.* **2019**, *44* (S1), 164–170.
- (30) Zhang, J.; Liu, Y.; Ren, P.; Han, H.; Zhang, S. A fully multifield coupling model of gas extraction and air leakage for in-seam borehole. *Energy Reports* **2021**, *7*, 1293–1305.
- (31) Fu, J.; Li, X.; Wang, Z. A novel sealing material and a bag-grouting sealing method for underground CBM drainage in China. *Constr Build Mater.* **2021**, *299*, 124016.
- (32) Hao, Z.; Lin, B.; Gao, Y.; Cheng, Y. Establishment and application of drilling sealing model in the spherical grouting mode based on the loosening-circle theory. *Int. J. Min. Sci. Technol.* **2012**, *22* (6), 895–898.
- (33) Wang, Z.; Sun, Y.; Li, Z.; Wang, Y.; You, Z. Multiphysics responses of coal seam gas extraction with borehole sealed by active support sealing method and its applications. *J. Nat. Gas Sci. Eng.* **2022**, *100*, 104466.
- (34) Zhang, Y.; Zou, Q.; Guo, L. Air-leakage Model and sealing technique with sealing-isolation integration for gas-drainage boreholes in coal mines. *Process Saf Environ. Prot* **2020**, *140*, 258–272.
- (35) Xiang, X.; Zhai, C.; Xu, Y.; Yu, X.; Xu, J. A flexible gel sealing material and a novel active sealing method for coal-bed methane drainage boreholes. *J. Nat. Gas Sci. Eng.* **2015**, *26*, 1187–1199.
- (36) Zhang, B.; Sun, H.; Liang, Y.; Wang, K.; Zou, Q. Characterization and quantification of mining-induced fractures in overlying strata: implications for coalbed methane drainage. *NAT RESOUR RES* **2020**, *29* (4), 2467–2480.
- (37) Huang, F.; Dong, C.; Shang, X.; You, Z. Effects of proppant wettability and size on transport and retention of coal fines in saturated proppant packs: experimental and theoretical studies. *Energy Fuels* **2021**, *35* (15), 11976–11991.
- (38) Su, X.; Wang, Q.; Song, J.; Chen, P.; Yao, S.; Hong, J.; Zhou, F. Experimental study of water blocking damage on coal. *J. Pet. Sci. Eng.* **2017**, *156*, 654–661.
- (39) Zhou, Y.; Yang, Y. Development Direction of Coal Mine Gas Utilization Technology to Realize Carbon Peak and Carbon Neutrality in China. *J. Coal Technol.* **2022**, *41*, 146–149.
- (40) Liu, D.; Zhang, R.; Liu, X.; Chen, W.; Tian, X.; Yuan, J.; Huang, B. Superhydrophobic Surface Fabrication for Strengthened Selective Water Shut-off Technology. *Energy Fuels* **2020**, *34* (5), 6501–6509.
- (41) Haeri, F.; Tapriyal, D.; Sanguinito, S.; Shi, F.; Fuchs, S. J.; Dalton, L. E.; Baltrus, J.; Howard, B.; Crandall, D.; Matranga, C. Co₂-brine contact angle measurements on navajo, nugget, bentheimer, bandera brown, berea, and mt. simon sandstones. *Energy Fuels* **2020**, *34* (5), 6085–6100.
- (42) Su, X.; Wang, Q.; Lin, H.; Song, J.; Guo, H. A combined stimulation technology for coalbed methane wells: Part 2. Application. *Fuel* **2018**, *233*, 539–551.
- (43) Su, X.; Song, J.; Guo, H.; Lin, H.; Liu, X.; Han, Y.; Zhang, S.; Li, X.; Yu, S. Increasing production mechanism and key technology of gas extraction in coal mines. *Coal Sci. Technol.* **2020**, *48* (12), 1–30.
- (44) Liu, Q.; Guo, Y.; An, F.; Lin, L.; Lai, Y. Water blocking effect caused by the use of hydraulic methods for permeability enhancement in coal seams and methods for its removal. *Int. J. Min. Sci. Technol.* **2016**, *26* (4), 615–621.
- (45) Guo, H.; Su, X. Research on the mechanism of gas emission inhibition in water-flooding coal seam. *J. China Coal. Soc.* **2010**, *35* (6), 928–931.