Development and Application of Water Sealing Technology for Gas Drainage Boreholes

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ABSTRACT: The poor sealing effect of gas extraction boreholes causes low efficiency of gas extraction. As a consequence, the requirements of coal mine safety production are often not attained. The sealing effect of boreholes depends not only on the sealing material itself but also on the combination degree between the material and the hole wall and the structural change characteristics of the material during operation. Our theoretical analysis shows that the amount of liquid leakage increases with the gap width Δh in a cubic law, decreases with the sealing hole viscosity in a hyperbolic form, increases linearly with the diameter of the borehole, and increases with the eccentricity in a quadratic function. We have developed a PD sealing material that has good compactness and sealing effects, excellent water retention performance, and an expansion rate of 1.29. The material can generate secondary expansion through microscopic development, which is beneficial to improve the quality of the sealing hole. HV-CMC has good stability in plugging mucus. In addition, the manual pump has been redeveloped to be portable and also to overcome the blockage of the suction valve of the original device. In addition, the auxiliary device for drilling and sealing has been invented. The field application demonstrates that the gas concentration and flow pressure difference of the new sealing borehole can be maintained at a high level in a short period of time and then decrease slowly with time. The resulting improved sealing effect demonstrates that our new approach has important theoretical and practical significance for mine gas drainage.

1. INTRODUCTION

Coal mine gas accidents have been one of the most serious disasters in the process of coal mining in China that seriously affect the safety of mine production and the life safety of miners.14–16 At present, the fundamental method to prevent coal seam gas disasters is to increase the permeability of coal seam. Commonly used methods include coal seam water injection, hydraulic fracturing, hydraulic punching, hydraulic cutting, and microwave radiation. Subsequently, gas extraction can be carried out to lower the coal seam gas concentration,6–9 hence reducing the outburst risk. However, the average concentration of gas extraction in many mining areas in China is lower than 10%.10–12 The low value is interpreted to result from the following three reasons: (1) Coal has a heterogeneous structure composed of pores and fractures. The pores are the main location for gas accumulation, whereas coal-rock fractures are the main channels for gas flow.13 (2) When drilling hole drainage is implemented in the working face, the drilled hole is relatively shallow and located in the pressure relief zone of coal seam.14–16 (3) The failure of borehole seal leads to low extraction efficiency.

The destruction of the structure of the coal around the gas extraction borehole by drilling is the main reason for the failure of borehole seal, as it reduces the strength and compression capacity. Furthermore, the coal is prone to plastic deformation after compression, leading to a large number of microcracks in the coal and rock mass around the borehole.17–19 In addition, new fissures evolve through the influence of mining and other factors from the microfissures in the borehole and its surrounding channels, thus further increasing the problem of sealing gas drainage boreholes.20,21 Using conventional yellow earth, cement mortar, polyurethane grouting hole sealing process such as fixed after forming, although before the fixed shape around the drilling and the drilling of microcracks have certain effects to block, but the new fracture cannot be affected by mining derived after sealing effect, at the same time the traditional solid hole sealing material cannot with the forced deformation of the drill, it is easy to cause the failure of borehole seal.22–24 Therefore, sealing the drilling hole and the surrounding coal body to trap the gas in the coal body effectively,25,26 and extract the gas efficiently through the drilling hole has become one of the key technologies to process the co-
The mining of coal gas and the green mining of coalbed methane.

The main hole sealing methods are hole sealing devices, inorganic materials (cement, mortar, etc.), and organic materials (polyurethane). The hole sealing device can seal the borehole quickly, but in the construction process of drilling construction, the requirements are more stringent. As the soft coal seam easily collapses, the hole sealing device is difficult to be sent. Moreover, the hole sealing device needs to be matched with expansion cement, and the operation process is cumbersome. The bore is sealed by inorganic materials, simple operation, low cost, have certain block of surrounding rock around the borehole fissure, but material curing slower, hole sealing time is long, cement mortar material shrinkage resistance, especially the problem of hole sealing is lax in cement overcomes the shortage of cement material solidification, and cracking in the solidification process, which makes it difficult to guarantee the hole sealing effect. As a result, the success rate of hole sealing is low. The clay material is difficult to fill into the drilling hole, causing a limitation of the sealing hole length and thus reducing the pumping efficiency. The expansion cement overcomes the shortage of cement material solidification time, solidification shrinkage, and improves the effect of hole sealing, but its own strength is low, which makes it difficult to meet the high-pressure sealing. At present, polyurethane material, including Novartis, Marithan, and Rocsil foam, is commonly used in coal mines, as it has a high expansion ratio and light-weight quality, causing fast sealing of the hole. However, due to the polyurethane reaction and weak bonding capacity of the borehole wall, the compressive properties of the material are poor, and therefore, it is only applicable for coal seam water injection. Nevertheless, it cannot satisfy the requirement of the high-pressure hydraulic fracturing drilling hole sealing in the complex geological structure of high-gas coal seam that often comprises large cracks around the borehole, causing an easy leakage of mucus, and repeated fluid rehydration is required for the application of mucus sealing. Moreover, plugging a large amount of mucus inside the borehole is a critical problem. In addition, the sealing section of gas drainage drilling hole is generally located in the stress reduction area and the postpeak stress rise area. Due to the breakage of coal and rock mass caused by stress release, transfer, and strong unloading load, large-scale mining has repeatedly disturbed the coal and rock mass, making the coal undergo multiple deformation and destruction processes. After the cracks around the drilling hole are communicated, it is easy to form an air leakage channel, which is difficult to block. Therefore, the flexible sealing material that can adapt to borehole deformation and active penetration is an ideal choice.

Considering the current status of gas drainage borehole sealing processes, we will apply seepage flow mechanics, applied chemistry, and materials science to develop a new kind of sealing material with convenient operation and a good sealing effect that will fulfill the higher requirements of concentration of gas extraction.

2. BASIC THEORY OF DRILLING FLUID SEAL

2.1. Distribution of Pressure Relief Zone of Borehole. During borehole formation, a crushing circle develops around the borehole due to the redistribution of rock mass stress field and drilling vibration damage. The following assumptions are proposed to obtain the radius of the surrounding rock fracture zone of the borehole: (1) The borehole is circular, and the stress field of the original rock is hydrostatic, so that the surrounding area is subjected to uniform isotropic pressure. (2) The stress distribution in the elastic zone is the same as that around the circular hole in the elastomer, and the failure of rock mass in the plastic zone satisfies the Mohr–Coulomb yield criterion. (3) The distribution of the four belts around the borehole is illustrated in Figure 1 (with P: original rock stress; P_i: support resistance; c: cohesive force of rock mass; φ: internal friction angle; σ_t: tangential stress; σ_r: radial stress; a: borehole radius; R: radius of plastic zone; R_c: radius of crushing zone; I: crushing zone; II: plastic zone; III: elastic region; IV: original rock stress area).

According to the given assumptions, the plastic zone radius is obtained as

\[ R = a \left[ \frac{(P + c\cot \varphi)(1 - \sin \varphi)^{1 - \sin \varphi/2\sin \varphi}}{P_i + c\cot \varphi} \right]^{1/2} \]  

The radius of the crushing zone is

\[ R_c = a \left[ \frac{(P + c\cot \varphi)(1 - \sin \varphi)}{(1 + \sin \varphi)(P + c\cot \varphi)} \right]^{1 - \sin \varphi/2\sin \varphi} \]  

The displacement of the periphery is

\[ \mu = \frac{\sin \varphi \Delta P + c\cot \varphi}{2G} \left[ \frac{1}{(P_i + c\cot \varphi)^{1 - \sin \varphi/2\sin \varphi}} \right] \]  

where \( G \) is the shear elastic modulus of the surrounding rock.

It is evident that the stability of the borehole and the surrounding displacement mainly depend on the original stress of the rock layer \( P \), the borehole radius \( a \), the internal friction angle \( \varphi \), and the cohesion force \( c \). At the same time, it can be concluded that they obey the following relationship:

(1) The displacement around the borehole increases exponentially with the increase of the original rock stress at the borehole location, and the magnitude of the...
exponential function depends on the change of $\phi$—the smaller the $\phi$ value, the larger the exponent, and the $\mu$ value grows faster.

(2) An increase of the borehole radius increases the plastic zone radius $R$ and the peripheral displacement $\mu$.

(3) The plastic zone radius $R$ and the surrounding displacement $\mu$ of the borehole increase significantly with the decrease of the internal friction angle $\phi$ and the cohesion force $c$. Hence, the strength of the surrounding rock decreases.

It is evident that in the process of borehole sealing, the material medium is easy to diffuse through a fracture in the pressure relief zone, which directly affects the quality of the sealing hole and thus impacts the gas extraction efficiency.

2.2. Influencing Factors of Drilling and Sealing and Their Analysis. 2.2.1. Crack Width. The fluid leakage $Q$ around the borehole increases with the increase of gap width $\Delta h$ in a cubic power. (Figure 2, with $D = 50$ mm, $\mu = 4.3 \times 10^{-2}$ Pa·s, $L = 1000$ mm, and $\delta = 0$). Figure 2 shows the relationship between the liquid leakage and the gap width for a pressure difference of $P_1 - P_2 = 2.9$ MPa. It follows that the relationship between gap width and leakage quantity is

$$Q = K_1 \Delta h^3$$

with $K_1$ as a parameter related to drilling diameter, pressure difference, and other factors.

Equation 4 indicates that an increase of $\Delta h$ causes a rapid increase of the liquid leakage $Q$. Therefore, to confine the leakage of sealing fluid below the permitted value during the measurement of gas pressure, a certain limit on the gap width value $\Delta h$ should be considered.

2.2.2. Sealing Fluid Viscosity. An increase of the sealing fluid viscosity $\mu$ causes a decrease of the liquid leakage following a hyperbolic curve (Figure 3). The relation is expressed as eq 5.

$$\mu Q = K_2$$

with $K_2$ as a parameter related to the gap width, differential pressure, and other factors.

According to eq 5, when $\mu = 4.2 \times 10^{-2}$ Pa·s, $Q = 88.3$ mL/s and when $\mu = 0.1$ Pa·s, $Q = 37.9$ mL/s. Therefore, to reduce the leakage of the sealing fluid, the viscosity of the sealing fluid can be appropriately increased on the premise of ensuring pipeline transmission.

2.2.3. Diameter of Borehole. The relation of borehole diameter $D$ and leakage $Q$ is linearly proportional (Figure 4) and expressed by eq 6

$$Q = K_3 \pi D$$

with $K_3$ as a parameter related to the width of the gap, differential pressure, and other factors. It follows that an increase of the borehole diameter causes an increase of the leakage.

Therefore, drill holes with small diameters are favorable in the measurement of gas pressure, to seal the hole and improve the accuracy of the measurement of gas pressure.

2.2.4. Eccentricity. The influence of the eccentricity $\delta$ on sealing fluid leakage $Q$ is shown in Figure 5, and their relationship is expressed in eq 7.
with \( \bar{B} \) as a parameter related to gap width and borehole diameter.

\[
Q = \bar{B}(1 + 1.5\delta^2)
\]  

(7)

Equation 7 indicates that an increase of eccentricity \( \delta \) causes an acceleration of the growth rate of the leakage quantity \( Q \). However, it is difficult to keep the sealing device completely centered in the downhole gas pressure measurement and borehole gas drainage. To reduce the leakage of sealing fluid and ensure the sealing effect, a value of \( \delta \leq 0.2 \) for the eccentricity of the sealing device is appropriate.

3. SEALING MATERIALS FOR DRILLING HOLES

3.1. Sealing Drilling Fluid Materials. Two kinds of new materials are used for sealing: organic material (CA) and inorganic material expansion cement (PD). CA monomer is a white or colorless granular solid or powder. It expands after absorbing water and is able to absorb water 10–100 times of its weight to form a gel. PD organically combines expansive cement with high-water material formula and adds several kinds of thickening materials and water-retaining materials, so that the PD slurry is thin at the beginning of formation and conducive for grouting. Five to seven hours after the injection of the slurry into the borehole, the reaction between the raw materials initiates and the slurry gradually expands in volume and increases in strength. After 28 h, the slurry is transformed to a solid and can remain in this state for a long time.

3.2. Sealing Fluid Material Characteristics. 3.2.1. Expansion Properties of Materials. To study the expansion behavior of the sealing material, we used the sealing material PD and added 100 mL of main material A and auxiliary material B (additive, water, etc.) in a measuring cup with a measurable capacity of 1000 mL, stirring for 1 min to ensure uniform mixing and completion of reaction. The volume values (in mL) were read every 2 min and recorded as \( V_1, V_2, V_3, \ldots, V_n \) with \( V_n \) as the final stable volume. \((V_n - 100)/100\) is the coefficient of expansion, \((V_n - V_{n-1})/V_{n-1}\) is the instantaneous expansion rate, and \(2 \times n\) is the expansion stability time. Sealing materials with excellent expansion behavior are characterized by a short expansion stability time, large expansion coefficient, and small instantaneous expansion rate in the beginning period. The results of the volume change of analyzed PD during expansion are shown in Figure 6.

As can be seen from Figure 6, the volume of PD began to expand after 5 h and increased from 890 to 1000 mL after 6.5 h.

At this time, the expansion coefficient was \(1000/890 = 1.12\). Subsequently, the volume continued to expand slowly and stabilized at 250 mL after 26 h. The expansion rate of PD increases relatively fast in the first 8 h, as the PD slurry is relatively thin initially and the agent reaction speed is relatively high. After 8 h, the slurry thickens and starts to solidify, so the expansion rate is reduced between 8 and 26 h. After 26 h, the volume of the PD becomes stable, with a final expansion coefficient of 1.29. The expansion coefficient is slightly smaller, but the expansion in a sealing section closed at both ends should have a better sealing effect.

3.2.2. Sealing Performance of Materials. To test the sealing behavior of the material, a glass tube with an inner diameter of 75 mm and a length of 2 m was used. The three kinds of sealing material were sealed at a length of 1 m. After waiting for sealing material function stability, with the vacuum pump suction, after negative pressure when achieving the maximum closed valves on the exhaust tube, observation records of the gas column pressure recovery process. The sealing behavior of the test material is shown in Figure 7, and the relationship between the negative pressure of the air chamber and the time after sealing of the three materials is shown in Figure 8.

The curve of CA material gradually decreases during the entire process, indicating that the decrease in velocity of negative pressure is slowing down. The negative pressure of the CA material is 90 kPa, and the pressure returns to the initial value 10 min after the pumping valve was closed. The negative pressure of the PD material is 86 kPa, and the pressure returns to the initial value 90 min after the pumping valve was closed. The negative pressure decreases more slowly than that of CA. The negative pressure of the polyurethane is 37 kPa, and the pressure returned...
to the initial value 10 min after the pumping valve was closed. The steep slope of the curve during the whole process indicates that the negative pressure decreases rapidly. At the same length of the sealing section, the sealing effect of polyurethane is significantly different from that of CA and PD.

3.2.3. Water Retention Performance of Materials. The experiment of the water retention behavior is shown in Figure 9.

![Figure 9. Water retention behavior of CA.](image)

Dry crushed coal particles were rounded into a pile and flattened at the top. After digging a groove with a diameter of 40 mm, the sealing material was poured into the groove with a depth of 40 mm. The results of the experiment are summarized in Table 1.

Table 1. Water Retention Test Results of Sealing Materials

| Material | Weight (g) | Water Seepage Distance (mm) |
|----------|------------|-----------------------------|
| CA       | 190        | 32                          |
| PD       | 254        | 7                           |

The water seepage distance of CA is deeper than that of PD (Table 1). After removing the PD material, a part has been bonded with coal. After removing the bonded coal, the remaining coal particles are as dry as before, indicating a good water retention performance of the PD material that is more effective than that of the CA.

3.2.4. Meso-Structure of Sealing Materials. Characteristic images of the reaction process between PD and polyurethane are shown in Figures 10 and 11.

Due to the large mass and volume ratio of water in the newly configured PD, the mixture presents obvious suspension characteristics (Figure 10a). The content of material in the capsule wall in PD is large, so the viscosity of the suspension is high, and the liquid tension increases accordingly. The bubbles formed in the stirring process are difficult to burst (Figure 10b). PD is also solidified slowly in the process of expansion and is not uniformly solidified entirely. After 16h of configuration, major parts of PD have basically solidified and pores are sparse (Figure 10c). Subsequently, PD still contains some liquid, and the pores are relatively dense (Figure 10d). After 26h, the PD is completely solidified into a soft solid with low strength. Pores with an average interval of 500 μm occur, but communication between individual pores is not developed (Figure 10e), which indicates that PD is entirely compact.

Polyurethane expands to form a cavity array with a diameter of 700–1000 μm (Figure 11a,b). However, many adjacent cavities communicate with each other through holes, which may result in poor overall air tightness.

4. BLOCKING MUCUS

Blocking mucus is an important constituent of the fluid sealing process. The low viscosity of mucus may lead to increased permeability and unacceptable consumption of the mucus. On the other hand, the high viscosity of the mucus and low permeability may lead to a significant reduction in the plugging fracture area, resulting in a poor final sealing effect. Therefore, reasonable plugging of the mucus material is crucial for the final sealing effect of drilling.

4.1. Optimization Ratio Experiment of Plugging Chemical Additive. Six chemical additives, including HV-PAC, HV-CMC, MV-CMC, HVT-CMC, Edible-CMC1, and Edible-CMC2, were used in the experiment and dissolved in water at 40 °C. Six aqueous solutions with mass ratios of 1:20, 1:25, 1:30, 1:35, 1:40, and 1:45 were prepared using the same mass method (Figure 12).

The results of the experiment indicate a successive decrease of the viscosity of the aqueous solution with the same proportion of the six chemical additives. The aqueous solution of HV-PAC and HV-CMC with a 1:40 ratio, the aqueous solution of MV-CMC with a 1:35 ratio, and the aqueous solution of HVT-CMC with a 1:30 ratio are suitable as plugging mucus. As the concentration of Edible-CMC1 and Edible-CMC2 is high, the viscosity is low, and the hydrolysis of Edible-CMC1 and Edible-CMC2 occurs in about 5 days. Therefore, they are inappropriate as plugging mucus. Due to microhydrolysis of two proportions of HV-PAC and considering the costs, the aqueous solution of HV-CMC with a 1:40 ratio was finally selected as the plugging mucus.

4.2. Stability of Plugging Mucus. Two aqueous solutions of HV-CMC with mass ratios of 1:35 and 1:45 were prepared using the same mass method and divided into two groups. A 0.3–0.5% preservative was added to one group, and the viscosity of the two groups was observed with time until the solution was stable (Figure 13).

After 5 days, the group with no preservative started to hydrolyze, whereas the preservative-added group remains unchanged. After 1 week, the original group hydrolyzed severely and the viscosity decreased significantly. As the preservative-added group still remains unchanged, the viscosity persists intact (Figure 13). Therefore, adding an antikilling agent to the blocking mucus can conserve the mucus and maintain good stability for a long time.
5. FIELD APPLICATION RESULTS

5.1. Project Overview. The Xinjiang mine field is located 11 km away from the city center of Yangquan City. Its administrative division is under the jurisdiction of Yangquan City of Shanxi Province (Figure 14). The test point is located at the 3217 working face. The studied coal seam is the No. 3 coal seam with a thickness ranging between 0.75 and 4.80 m and an average thickness of 2.26 m. The coal seam dips gently with an angle of 3–9° and a strike length of 1552 m and an inclination

Figure 10. Microscopic characteristics of the materials. (a) Freshly configured PD (300 times magnification), (b) PD beginning to expand (150 times magnification), (c) PD during solidification (250 times magnification), (d) PD during solidification (100 times magnification), and (e) PD after solidification (250 times magnification).

Figure 11. Polyurethane structure diagram. (a) Polyurethane structure drawing (200x magnification) and (b) polyurethane structure drawing (350x magnification).
length of 234 m. The original gas content of No. 3 coal is 18.17 m³/t. According to the requirements of Basic Indicator of Coal Mine Gas Drainage, the coal seam gas content must be reduced to less than 8 m³/t before mining at the working face. The amount of gas to be extracted at the 3217 working face is 1.258 million m³.

The test boreholes are arranged in the wind roadway at working face 3127, and the boreholes are drilled at the roof of the roadway. The boreholes extend in the rock all the time. The borehole locations are more than 100 m away from the mining face, and the borehole location does not communicate with the fracture zone caused by mining. The spatial relationship of the test boreholes is shown in Figure 15.

5.2. Test Results. Three materials are used to seal the holes. Holes 1#, 2#, and 3# are sealed with high-water materials; holes 4#, 5#, and 6# are sealed with polyurethane materials; and holes 7#, 8#, and 9# are sealed with PD materials. The data for all boreholes are shown in Table 2.

The average drilling drainage parameters in the sealed borehole by the three materials were calculated and summarized in Figure 16 by comparing the drilling drainage concentration.
and flow pressure difference in the sealed borehole by the three materials.

The gas extraction concentration of drilled holes sealed by PD material is 49%, followed by polyurethane (32%) and high-water material yielded the lowest value of 20% (Figure 16). In terms of flow rate and pressure difference, the highest is 70 mm H2O for the hole sealed by PD material, followed by 25 mm H2O by high-water material and 13 mm H2O by polyurethane (Figure 16). It follows that the PD material has the best sealing effect on boreholes and is conducive to gas extraction.

The variation of the gas extraction concentration in relation to the convergence of the mining face toward the borehole in the test section is shown in Figure 17. In general, in the early stage of gas extraction, the gas concentration is at a high level and gradually decreases with time and finally converges into a stable state. In the first 5 days of observation, the gas concentration in the borehole sealed by PD material was high, with a maximum of 86.1% and an average of 82%. The drilling hole sealed by polyurethane material follows, with a maximum value of 80.4% and an average value of 46.7%. The borehole sealed by high-water material has the lowest value, with a maximum of 60.8% and an average of 35.6%. After 5 days, the gas concentration in the drilling holes sealed by the high-water material passes the curve of the polyurethane material but on a lower level compared with the data of the PD material. Moreover, the gas concentration in the borehole sealed by PD material decreases slowly compared with that of the other two materials. The test demonstrates that the PD material has the best sealing effect and ensures that the concentration of gas extracted from boreholes is at a higher level for a long time and is conducive to gas extraction.

6. DISCUSSION

6.1. Microstructure of Sealing Materials and Its Influence on Sealing Effect. The enlarged internal structure of polyurethane resembles a honeycomb network of holes (Figure 11). During gas extraction, gas can escape from the holes in the honeycomb network. In addition, the pore structure inside the polyurethane has a smooth surface and little resistance to gas flow. In the process of gas extraction, the gas in the borehole and the air in the roadway can leak and penetrate through the holes in the polyurethane material. In contrast, the PD shows no holes

Table 2. Experimental Parameters and Test Results

| hole number | hole depth (m) | sealing material | sealing section length (m) | gas concentration (%) | flow pressure difference (mmH2O) |
|-------------|---------------|-----------------|--------------------------|-----------------------|---------------------------------|
| 1#          | 33            | high-water material | 1–8                    | 9                     | 20                              |
| 2#          | 30            | high-water material | 1–9                    | 37                    | 4                               |
| 3#          | 22            | high-water material | 1–11                   | 51                    | 15                              |
| 4#          | 35            | polyurethane     | 1–9                    | 22                    | 60                              |
| 5#          | 27            | polyurethane     | 1–11                   | 23                    | 10                              |
| 6#          | 27            | polyurethane     | 1–12                   | 15                    | 5                               |
| 7#          | 38            | PD               | 1–8                    | 48                    | 100                             |
| 8#          | 31            | PD               | 1–9                    | 37                    | 60                              |
| 9#          | 32            | PD               | 1–11                   | 62                    | 50                              |

Figure 16. Comparison of drainage concentration and pressure difference in the sealed borehole by three materials.

Figure 17. Variation of gas concentration in sealing boreholes with three materials.
even in high magnification (Figure 10). The internal structure is relatively dense and without cracks and therefore effectively suppresses the gas and air in the roadway. In the borehole drilled in the body by PD materials itself in a crack in the leak and infiltration, reduce the gas extraction in the process of the effects on the gas concentration in the borehole sealing material itself.

6.2. Microscopic Combination of Sealing Material and Borehole Wall and Its Influence on Sealing Effect. Drilling in coal induces pressure relief zones around the borehole due to vibration. To improve the quality of the sealing hole, a sealing material is required with certain fluidity and adhesion, so that the sealing material can be fully combined with the coal wall. The fluidity of polyurethane in the hole sealing process is poor. Also, the combination density with the coal wall is poor. Therefore, the combination stability of polyurethane with the coal sample at the hole wall is poor. This leads to the finding that polyurethane cannot be fully combined with the coal wall in the hole sealing process, and there is a blank area in the pressure relief area of the borehole. In the process of hole sealing, the composite material has good fluidity and the combination stability with the coal at the hole wall is strong. It can fully seal the wall collapse area caused by hole collapse without a blank area at the joint. The composite material effectively prevents the air in the roadway from being pumped into the borehole due to the negative pressure in the borehole and increases the gas extraction concentration.

6.3. Microscopic Development of Sealing Materials and Its Influence on Drilling Sealing Effect. Our field experiments showed that polyurethane permeability has been fully response, after the end not around borehole fissure circle continues to develop, still has not been stopped around the drilling micro holes and cracks, gas and air in the roadway can be through the cracks around the hole leakage and residual porosity and infiltration, which affect the gas extraction efficiency. However, due to the excellent characteristics of the composite material at the end of the compound sealing material penetration, it can continue in the residual around borehole fissure and pore development (the expansion coefficient of about 1.29), with the residual cracks and holes around the borehole, have the effect of secondary seal, can effectively improve the effect of the gas extraction.

6.4. Influence of Sealing System on Gas Extraction from Boreholes. The field data indicate an initial high level of the gas concentration and flow pressure difference in the sealing borehole, as the material can penetrate into the fracture circle to a large extent, making it denser. The gas concentration decreases with time, but compared with the other two materials, the gas concentration in the borehole sealed by PD material decreases more slowly, probably due to the microscopic development of the material. After the secondary expansion, the PD material can ensure the drilling sealing effect. Although the gas concentration decreases after stabilization, it remains at a higher level compared with the drilling hole sealed by the other two materials and can fulfill the requirements of gas extraction in coal mines.

7. CONCLUSIONS
The quality of borehole sealing is an important factor to determine the effect of gas extraction. We developed a new material and conducted field experiments to test the new approach under natural conditions.

(1) Through theoretical analysis, the factors and causes of sealing material leakage and related influencing factors are obtained. They include crack width, sealing fluid viscosity, borehole diameter, and eccentricity. The amount of liquid leakage increases with the gap width $\Delta h$ in a cubic law, decreases with the sealing hole viscosity in a hyperbolic form, increases linearly with the diameter of the borehole, and increases with the eccentricity in a quadratic function.

(2) An inorganic material expanded cement jelly (PD) sealing material and plugging fluid have been developed. The expansion rate of the PD material is 1.29, and its sealing property and water retention behavior are superior to those of CA and polyurethane materials. In addition, the material not only has a compact microstructure but also has a strong permeability. Moreover, the secondary expansion of its microscopic development can effectively seal the borehole and improve the gas extraction efficiency. An aqueous solution of HV-CMC was developed as the blocking mucus with high stability.

(3) The industrial experiment was conducted in working face 3127 of the Xinjiang mine. According to the field data, the gas concentration and flow pressure difference of the sealing borehole of the new material are significantly higher than those of other materials and can be maintained at a higher level for a long term, which improves the gas extraction efficiency.

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Notes
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REFERENCES

(1) Wang, Z. L.; Liu, B.; Han, Y. H.; Li, Z. Y.; Cao, Y. J.; Qi, F. Z. Determining the layout parameters of the gas drainage roadway: A study for Sima coalmine China. Adv. Civ. Eng. 2021, 2021, 1–8.

(2) Li, D.; Cao, Q. G.; Wang, J. Q.; Xin, M. Application of integrated method of HAZOP-AHP and fuzzy comprehensive evaluation in coal mine gas explosion accident. IOP Conf. Ser.: Earth Environ. Sci. 2021, 692, No. 042103.

(3) Zhang, J. J.; Xu, K. L.; You, G.; Wang, B. B.; Zhao, L. C. Case study of gas drainage well location optimization in abandoned coal mine based on reservoir simulation model. Energy Explor. Exploit. 2021, 39, 1993–2005.

(4) Wang, B.; Liu, S. D.; Zhou, F. B.; Hao, J. L. Experimental seismic attributes of gas-bearing anthracite. Arab. J. Geosci. 2019, 12, No. 730.

(5) Li, X. L.; Cao, Z. Y.; Xu, Y. L. Characteristics and trends of coal mine safety development. Energy Sources, Part A 2020, 1–19.

(6) Zheng, G. Q.; Han, J. Y.; Sang, F. Y.; Gao, T. X.; Chen, D.; Zhang, Z. D. Case study of gas drainage well location optimization in abandoned coal mine based on reservoir simulation model. Energy Explor. Exploit. 2021, 39, 1993–2005.

(7) Liu, Z. L.; Wang, J.; Chen, Tj.; Wu, G. Q.; Li, G. Seismic time-lapse monitoring of gas drainage in an underground coal working face. Int. J. Coal Geol. 2021, 237, No. 103712.

(8) Su, X. B.; Li, F.; Su, L. N.; Wang, Q. The experimental study on integrated hydraulic fracturing of coal measures gas reservoirs. Fuel 2020, 270, No. 117527.

(9) Selrak, N.; Swolken, J. Possibilities of Capturing Methane from Hard Coal Deposits Lying at Great Depths. Energies 2021, 14, No. 3542.

(10) Cai, J. T.; Wu, J. S.; Yuan, S. Q.; Liu, Z.; Kong, D. S. Numerical analysis of multi-effects on the leakage and gas diffusion of gas drainage pipeline in underground coal mines. Process Saf. Environ. Prot. 2021, 151, 166–181.

(11) Lin, B. Q.; Dai, H. M.; Wang, C. Q.; Li, Q. Z.; Wang, K.; Zheng, Y. Z. Combustion characteristics of low concentration coal methane in divergent porous media burner. Int. J. Min. Sci. Technol. 2014, 24, 671–677.

(12) Hao, Z. Y.; Zhou, C.; Li, B. Q.; Pang, Y.; Li, Z. W. Pressure-liberal and permeability increase technology for upper corner of working face by high position directional No. 505.

(13) Du, Z. G.; Zhang, X. D.; Huang, Q.; Zhang, S.; Wang, C. L. Investigation of coal pore and fracture distributions and their contributions to coal reservoir permeability in the Changzhi block, middle-southern Qinshui Basin, North China. Arab. J. Geosci. 2019, 12, No. 505.

(14) Duan, H. J.; Hao, S. J.; Zhao, Y. Z. Differential gas drainage technology for upper corner of working face by high position directional long borehole. IOP Conf. Ser.: Earth Environ. Sci. 2021, 687, No. 012180.

(15) Huang, J. Z.; Li, J. X.; Pan, X.; Xie, T. Z.; Hua, W.; Dong, S. M. Numerical Investigation on Mixed Mode (I-II) Fracture Propagation of CCBD Specimens Under Confining Pressure. Int. J. Appl. Mech. 2020, 12, No. 2050111.

(16) Guo, Y. T.; Lei, W.; Chang, X. Study on the damage characteristics of gas-bearing shale under different loading stress paths. PLoS One 2019, 14, No. e0224654.

(17) Gao, Z. S.; Zhu, C. J.; Lu, X. M.; Ren, J. Prevention and control of abnormal gas emission caused by accidental discharge of floor fissure water: a case study. Nat. Hazards 2020, 100, 713–733.

(18) Li, X. L.; Chen, S. J.; Li, Z. H.; Wang, E. Y. Rockburst mechanism in coal rock with structural surface and the microseismic (MS) and electromagnetic radiation (EMR) response. Eng. Failure Anal. 2021, 124, No. 105396.

(19) Liu, S. M.; Li, X. L.; Wang, D. K.; Zhang, D. M. Experimental study on temperature response of different ranks of coal to liquid nitrogen soaking. Nat. Resour. Res. 2021, 30, 1467–1480.

(20) Zhang, L. L.; Wu, W. J.; Biao, Y. P.; Yang, M. D.; Luo, H. G. Influences of gas drainage pipe positions on spontaneous coal combustion in the gob: A case study of Baode coal mine in China. Combust. Sci. Technol. 2021, 37, 1–17.

(21) Zhao, Y.; Qi, Q. J.; Jia, X. L. Prediction model for spontaneous combustion of coal around boreholes using bedding gas drainage. Shock Vib. 2021, 2021, No. S533054.

(22) Guo, X.; Xue, S.; Zheng, C. H.; Li, Y. B. Experimental research on performance of new gas drainage borehole sealing material with high fluidity. Adv. Mater. Sci. Eng. 2021, No. 6645425.

(23) Zhai, C.; Hao, Z. Y.; Lin, B. Q. Research on a new composite sealing material of gas drainage borehole and its sealing performance. Procedia Eng. 2011, 26, 1406–1416.

(24) Li, B.; Zhang, J. X.; Wei, J. P.; Zhang, Q. Preparation and sealing performance of a new coal dust polymer composite sealing material. Adv. Mater. Sci. Eng. 2018, 2018, No. 640913.

(25) Zhang, J. G.; Zhai, C.; Zhong, C.; Xu, J. Z.; Sun, Y. Investigation of sealing mechanism and field application of upward borehole self-sealing technology using drill cuttings for safe mining. Saf. Sci. 2019, 115, 141–153.

(26) Zhang, Y. F.; Zhou, F. B.; Xia, T. Q. Blockage characterization of particles for gas—solid two phase flow in the broken zone around drainage borehole. Environ. Earth Sci. 2020, 79, No. 54.

(27) Li, S. G.; Wei, Z. Y.; Lin, H. F.; Zhao, P. X.; Xiao, P.; Hao, Y. Y. Research and development of 3D large-scale physical simulation experimental system for coal and gas co-extraction and its application. J. China Coal Soc. 2019, 44, 236–245.

(28) Qin, B.; Hao, J. F.; Liang, B.; Sun, W. J.; Qin, X. W.; Li, C. Z. Nonlinear constrained multivariable spatiotemporal collaborative optimization model for coal and gas co-mining. J. Nat. Gas Sci. Eng. 2019, 44, 593–600.

(29) Li, Y. Q.; Tang, Y. Z.; Tang, S.; Yu, Y. Innovation and Development of Coal and Gas Co-mining Technology in Huanain Mining Area. Saf. Coal Min. 2020, 51, 77–81.

(30) Zhang, J. X.; Zhang, Q.; Feng, J. U.; Zhou, N.; Li, M.; Zhang, W. Q. Practice and technique of green mining with integration of mining, dressing, backfilling and X in coal resources. J. China Coal Soc. 2019, 44, 64–73.

(31) Wang, L.; Cheng, Y. P.; An, F. H.; Zhou, H. X.; Kong, S. L.; Wang, W. Characteristics of gas disaster in the Huaibei coalfield and its control and development technologies. Nat. Hazards 2014, 71, 85–107.

(32) Zhou, Y. L.; Zhou, W.; Lu, X.; Jiskani, I. M.; Cai, Q. X.; Liu, P.; Li, L. Evaluation Index System of Green Surface Mining in China. Miner. Metall. Explor. 2020, 37, 1093–1103.

(33) Zhang, C.; Jin, G. H.; Liu, C.; Li, S. G.; Xue, J. H.; Cheng, R. H.; Liu, H. Sealing Performance of New Solidified Materials: Mechanical Properties and Stress Sensitivity Characterization of Pores. Adv. Polym. Technol. 2020, 2020, 1–16.

(34) Zhang, C.; Lin, B. Q.; Zhou, Y.; Zhai, C.; Zhu, C. J. Study on “fracturing-sealing” integration technology based on high-energy gas fracturing in single seam with high gas and low air permeability. Int. J. Min. Sci. Technol. 2013, 23, 841–846.

(35) Jiang, B. Y.; Gu, S. T.; Li, W. S.; Zhang, G. C.; Zhang, J. H. Case Studies of Comprehensive Gas Control Method during Fully Mechanized Caving of Low-Permeability Ultrathick Coal Seams. GeoFluids 2021, No. 5558678.

(36) Zhou, A. T.; Wang, K. A new inorganic sealing material used for gas extraction borehole. Inorg. Chem. Commun. 2019, 102, 75–82.

(37) Chao, Z.; Jie, C.; Li, S. G.; Liu, C.; Qin, L.; Bao, R. Y.; Liu, H.; Cheng, R. H. Experimental study comparing the microscopic properties of a new borehole sealing material with ordinary cement grout. Environ. Earth Sci. 2019, 78, No. 149.

(38) Li, D. Q.; Li, H. G. A new technology for the drilling of long boreholes for gas drainage in a soft coal seam. J. Pet. Sci. Eng. 2016, 137, 107–112.

(39) Zhai, C.; Xiang, X. W.; Zou, Q. L.; Yu, X.; Xu, Y. M. Influence factors analysis of a flexible gel sealing material for coal-bed methane drainage boreholes. Environ. Earth Sci. 2016, 75, No. 385.
(40) Hu, Q. T.; Liu, L.; Li, Q. G.; Wu, Y. Q.; Wang, X. G.; Jiang, Z. Z.; Yan, F. Z.; Xu, Y. C.; et al. Experimental investigation on crack competitive extension during hydraulic fracturing in coal measures strata. Fuel 2020, 265, No. 117003.

(41) Wang, S. L.; Zhao, M.; Qin, R. X. Investigation of sealing effect of gas drainage borehole and analysis of influencing factors. China Energy Environ. Prot. 2017, 39, 92–95.

(42) Li, X. L.; Chen, S. J.; Zhang, Q. M.; Gao, X.; Feng, F. Research on theory, simulation and measurement of stress behavior under regenerated roof condition. Geomech. Eng. 2021, 26, 49–61.

(43) Li, X. L.; Chen, S. J.; Liu, S. M.; Li, Z. H. AE waveform characteristics of rock mass under uniaxial loading based on Hilbert-Huang transform. J. Cent. South Univ. 2021, 28, 1843–1856.

(44) Ma, H. Y. Research on sealing technology of pre-pumping boreholes along level bed in C8 coal seam of Hexing Mine. China Coal 2017, 43, 151–158.

(45) Chen, Y. Experimental study on gas drainage effect of crossing holes with different sealing technology. Coal Eng. 2017, 49, 99–101.

(46) Ni, G. H.; Dong, K.; Li, S.; Sun, Q.; Huang, D. M.; Wang, N.; Cheng, Y. Y. Development and performance testing of the new sealing material for gas drainage drilling in coal mine. Powder Technol. 2020, 363, 152–160.

(47) Bekmezci, O. K.; Sapci-Ayas, Z.; Ucar, D. Novel gas measurement based on pressure triggered release cycles for biochemical methane potential tests Int. J. Chem. React. Eng. 2021, 196, 585–596 DOI: 10.1515/ijcre-2020-0244.

(48) Guo, J. T. Composite support technology of soft coal roadway with large loose zone. Coal Eng. 2016, 48, 35–41.