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Research on a New Composite Sealing Material of Gas Drainage Borehole and Its Sealing Performance

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Abstract

Aiming at the shortcomings of the traditional sealing materials of borehole for gas suction such as yellow mud and cement-sand grout, which indicated that the traditional sealing materials of borehole are easy to shrink after sealing, the sealing length of borehole is short and the sealing quality is not satisfactory, and the sealing price of the high-water material and polyurethane are also high. In this study, taking cement as a base material, a new composite sealing material mixed by expansion admixture, additive, fibrin and coupling agent was developed and the sealing performance and expansion performance of the material were also studied and analyzed. At the same time, the microscopic image handling system was used to investigate the microcosmic structure of material. The studying results showed that the new composite sealing material possessed a good expansion performance and a definite fluidity convenient for grouting. The material after solidification could combine closely with the borehole wall, and there was certain strength and it was not easy to shrink. Additionally, the suction gas concentration by borehole sealing exceeded 40%. It can be seen that the sealing performance of the composite sealing material is advantageous.

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1. Introduction

Coal is one of main energy resources in China. The gas as an associated mineral of coal is not only a cause of the fateful disaster in coal mine and a cause of the atmosphere pollution, but also a valuable non-renewable energy resource. In China, the geological conditions of coal mines are complex, high gassy mines account for 50% - 70%^[1]. More effective measures of gas prevention is to carry out the gas suction through borehole in coal seam, so as to achieve the requirement of suction index and remove the gas disaster in coal seam. The borehole of gas suction should be sealed to carry out the gas suction. Currently, materials of borehole sealing include mainly yellow mud, cement-sand grout, high-water material and polyurethane. However, yellow mud and cement-sand grout after borehole sealing are easy to shrink, the length of borehole sealing is short and the sealing quality also is not satisfactory. At present, yellow mud and cement-sand grout are still used for sealing borehole of gas suction in about 2/3 coal mines. Owing to high price of high-water material and polyurethane, the cost of borehole sealing is very high^{[2][3]}.

The interior fractured net of underground borehole in coal mine is developed and the stress field of surrounding rocks of underground roadway in coal mine impacts on it, thus, it is very difficult to seal the boreholes and to be resolved in long time. Therefore, it would directly result in low gas concentration and bad suction results. The pre-suction gas concentration in about 65% of working faces in China now is lower than 30%. It reflected fully present state of bad quality of borehole sealing. The suction negative pressure of borehole bottom has an effect to drainage gas and constraint gas desorption, thus, the quality of borehole sealing is directly relevant to the effect of gas suction^[4].

Institute of Gas Utilization and Prevention of China University of Mining and Technology carried out a great number of experimental studies, and taking cement and resin as base materials, a new composite sealing material for gas suction mixed by a little of expansion admixture, additive, fibrin and coupling agent was developed. The cost of this material with good expansion performance is lower, and the material may combine closely with borehole wall and possesses a definite fluidity convenient for grouting. The material after solidification has certain strength uneasy to shrink. Therefore, this material will have good application prospects for borehole sealing of gas suction and pore space plugging. In this study, from the point of view of the microcosmic structure and industrial testing, the sealing characteristics of borehole by using a new material will be analyzed. (10 pt) Here introduce the paper, and put a nomenclature if necessary, in a box with the same font size as the rest of the paper. The paragraphs continue from here and are only separated by headings, subheadings, images and formulae. The section headings are arranged by numbers, bold and 10 pt. Here follows further instructions for authors.

2. Material composition

According to demand for sealing microfractures of the gas suction boreholes and combined with advantages of polymer and cement, designing and developing the composite sealing material of cementbased polymer should follow several principles as follows: (1) The sealing material should have integral density, aging resistance and dry cracking resistance; (2) The sealing material should have a definite expansion performance to seal microfractures around borehole; (3) Taking cement as base material, the polymer should be used to change the cement characteristics; (4) The sealing material should be used convenient for underground and the difficulty of distributing ratio and operation is low; (5) Different raw materials of the sealing material are all the daily chemical industrial materials; and (6) The polymer in use for changing characteristics should be innoxious.

Main raw materials of the sealing material consist of cement, water, expansion agent, additive, resin, fibrin and coupling agent. Cement is a main base material of the sealing materials to increase the compressive strength and the setting time of materials. The coupling agent is a plastic additive in use for

improving the synthesized resin and inorganic filling agent or enhancing the interface performance of materials, and its dosage usually is $0.5 \sim 2\%$. Currently, the coupling agent consists of two proportions, such as the amphiphilic inorganic base agglomerate which may act with the inorganic filling agent or enhancing material and the amphiphilic organic base agglomerate which may act with the synthesized resin. Expansion agent is a chemical applied agent added in cement. Along with coagulation and cementation of cement, the volume of expansion agent may be expanded to compensate the compressive and tensile prestressing forces as well as to plenty fill clearances of cement. The action of fibrin may be to maintain the moisture, bonding and consistency of materials, so that the moisture in grout of the composite sealing material could be reduced to permeate into coal rib. The actions of additive are as follows:

- First point, increase in grout suspension may be easy to flow the grout of the composite sealing material in transportation pipeline;
- Second point, the viscosity of grout may be increased;
- Third point, the initial setting time may be shortened;
- And so on, Composite grout may be infiltrated into fissures and fractures to plug the capillary channels, as well as to improve density and strength of materials. There is a lively epoxy group contained in the molecule structure of resin, so that it may cause the cross linking reactivity with several kinds of solidifying agents to form the high polymer with three dimensional mesh structure^{[5][6]}.

Based on the characteristic requirements of borehole sealing, expansion and strength of the sealing materials for gas suction boreholes, components of the composite sealing material are finally determined by the orthogonal testing, sealing testing of borehole for underground grouting and the repeated contrasts of the suction results. Meanwhile, the quality ratio of each component needed by preparation of the composite sealing material is also determined, i.e., cement, water, expansion agent, additive, resin, fibrin and coupling agent are 1:1.6: 0.03: 0.05:0.02: 0.04: 0.01.

3. Study on sealing characteristics of materials

3.1. Sealing principles

Usually, the sealed medium would be often leaked to the outside side of sealing connection point in form of diffusion or leakage, but for the borehole, its leaking pattern takes especially the leakage form as priority. Through experiments and field actual observations, it can be seen that a leakage reason is caused mostly by the clearances existed in the sealing connection point and cranny around borehole, and the sealing medium may result in leakage by clearances and cranny under action of the pressure difference. Thus, to increase the sealing performance of borehole needs to reduce or remove the clearances of the sealing connection point. Generally, a handling method is that the binding force of the sealing material is used to table, contact and close the sealing surfaces each other.

Sealing ability is a key factor, that is, sealing ability of sealing material could remove the clearances of the sealing connection point under the condition of the least contacting pressure, and result in a definite friction force to prevent the thrust caused by inside gas pressure. In process of the engineering practice, the rule for evaluating the sealing capability of material is the parameter of deformation-strength performances, especially the least contacting pressure value of sealing performance should be ensured. Under the same conditions the contacting pressure of material with higher sealing ability is lower and the sealing reliability is higher. Therefore, size of the contacting pressure should take the sealing contacting surface resulting in elastic deformation and the sealing structure with rigidity as the best of all. Under the condition of allowing contacting pressure, the elastic deformation of contacting surface may fill the clearances on surface, moreover, the rigid sealing structure may guarantee the sealing element, and the

contacting pressure needed by borehole sealing could be maintained. Connecting sealing is able to breakage and result in leakage if the sealing pressure is smaller than the critical value. Additionally, the stress relaxation and creep deformation are also main factors to determine the contacting sealing results^[7]. In course of borehole sealing, because coal is the fractured medium, the leakage of sealing grout not only may occur in the clearances along the axial end, but also in fractures along the radial coals. In order to discuss the leakage of the sealing fluid in fractures of coals, it supposes that the leakage of sealing grout in fractures of coals is regarded as the liquid movement in the pure fractures. According to seepage mechanics, the equation of the radial leakage quantity q_0 of sealing liquid along borehole may be deduced as Eq. 1.

$$q0 = 2\pi L \left[\frac{\ln\left(\frac{R_0}{R_e}\right) + \sqrt{\left(\ln\left(\frac{R_0}{R_e}\right)\right)^2 + \frac{4\beta K_1}{\mu} \left(\frac{1}{R_e} - \frac{1}{R_0}\right) \Delta p}}{2\beta \left(\frac{1}{R_e} - \frac{1}{R_0}\right)} \right]$$
(1)

Where, K_1 is permeability coefficient of fracture; R_0 is radius of borehole; $R_{e is}$ boundary radius of leakage; p_0 is pressure of sealing fluid; p_e is boundary pressure of leakage; L is length of the sealing section of borehole fluid; Δp is pressure difference of sealing fluid; β is constant; μ is viscosity of sealing fluid.

From Eq. 1, it can be seen that for a definite borehole the size of leakage quantity q_0 of sealing fluid is relevant to the length of borehole sealing section, permeability coefficient of fracture, fluid viscosity and pressure difference of sealing fluid. In order to reduce the leakage of sealing fluid and ensure the sealing results of sealing section of borehole as soon as possible, viscosity of the sealing fluid should be increased and the pressure difference of the sealing fluid should be properly reduced^[8].

As analyzed above, the material performance of borehole sealing is a key factor to impact on the gas suction results in coal seam. The composite sealing material possesses such characteristics as integral density, aging resistance and dry cracking resistance, because polymer is used to carry out the interface modification of cement material, the raw materials with expansion function are added into composites system, and then the micro-capsule technology is applied to handle the expansion medicament. A course of capsule wall reaction entering into the sol system is to diffuse and control the chemical reaction, meanwhile, the whole sol system is also in flocculating and finally bonded with borehole wall and fractured wall surface into an integration, thereby the sealing performance of material would be improved.

3.2. Testing of expansion performance

A method for observing the expansion performance of sealing material is as follows: The 890ml sealing material according to distributing ratio should be put in measuring cup with 1500ml capacity, and then the initial volume V_0 is registered and even admixture and entire reaction are achieved by mixing for 1min, the volume values marked by $V_{1,}V_{2,}V_{3}...V_{n}$ should be read every 2min. V_n is the final stable value, i.e. V_n/V_0 is its expansion coefficient. The results of expansion performance of the composite sealing materials are shown in table 1.

Table 1 Parameters of expansion course of composite sealing material

Raw material volume/ml	Volume of resultant/ml	Expansion
890	1150	1.29

Concrete expansion course						
time/h	volume/ml	Time /h	volume/ml	time/h	volume/ml	
0	890	6	1000	16	1150	
1	900	7	1030	18	1150	
2	920	8	1090	20	1150	
3	945	10	1110	22	1150	
4	970	12	1130	24	1150	
5	990	14	1150	26	1150	

The volume of the composite sealing material after 1h starts expanding, after 6h the volume increases from primary 890ml to 1000ml. However, the expansion coefficient is 1000/890 = 1.12, and then the volume continues to expand slowly. After 26h the volume is stable by 1150ml. Expansion velocity of the composite sealing material is rather rapid within former 8h because here the grout of the composite sealing material is rather rare, the reacting velocity of medicament is also violent, and after 8h the grout changes into density and starts solidifying. Its expansion velocity between 8h and 26h is rather slow, but after 26h the volume of the composite sealing material is stable and the final free expansion coefficient is 1.29. Thus, the composite sealing material has satisfied such characteristics as the small instantaneous expansion velocity at beginning time, the big expansion coefficient and the short stable time of expansion. The composite sealing material is shown in Fig. 1.





3.3. Observation of microcosmic structure

Observing the microcosmic structure of material may understand exactly the microcosmic modality of sealing material in process of reaction. In this study, the microcosmic image handling system developed by Occident Land Instrument Co. is used. This system can amplify the image of sample up to 50 - 7000 times by adjusting the optics system as shown in Fig.2.



Fig.2 Handling System of microscopic image



Fig.3 Configured composite sealing material (300 times amplifying)



Fig.4 Expanding composite sealing material (150 times amplifying)

The characteristic structure length of sealing material focuses on 50 - 500 micron. The proper amplifying multiple of microscopic system is 50 - 300 times. Author has observed the characteristic images of the composite sealing material in course of reaction within a range of 50 - 300 times. The reacting course is divided into configuration, expansion and solidification, and there are 4 stages after solidification as shown in Fig.3 ~ Fig.6.



(a) (250 times amplifying)

(b) (100 times amplifying)





Fig.6 Composite sealing material after solidification (250 times amplifying)

The admixture of configured composite sealing material presents an obvious characteristic of suspended turbid fluid due to a bigger proportion of water. The viscosity of suspended turbid fluid is big because of bigger content of capsule wall matter in the composite sealing material, the liquid tensile force will be also increased and the air bubbles formed in course of mixing are difficult to rupture. In Fig.3 there is such an air bubble with diameter of 750 micron. After about 7h, the expanding medicament has permeated the capsule wall to reaction and to create air viscosity in the material, so that the volume of material increases. The microcosmic structure of the composite sealing material when expanding is shown in Fig.4. The densely covered macula in Fig.4 presents the created air holes, and there are several rupture air holes with diameter of 200 - 450 micron at left bottom of diagram. In process of expansion the composite sealing material is also slowly solidified, but there is no integrated even solidification as shown in Fig.5. After configuration for 16h, a part of the composite sealing material in (a) diagram has been basically solidified and the air holes are comparatively sparseness. However, in (b) diagram there exist a part of liquids, and the air holes are comparatively dense. After 26h the composite sealing material is entirely solidified into a soft solid, and then the soft solid is smeared on the glass slide to observe its structure. Fig.6 shows that on an average an air hole in material occurs every 500 micron, but there is no linkage among air holes. It shows that the composite sealing material is entirely dense.

The microscopic structures of polyurethane as borehole sealing material currently used underground are contrasted and observed in this study as shown in Fig.7 and Fig.8.



Fig.7 Structure of polyurethane (200 times amplifying)



Fig.8 Structure of polyurethane (350 times amplifying)

In Fig.7, the contrast of image system is not clear duo to the strong light irradiation, but in Fig.8 the contrast of material by coloration is distinct. The polyurethane after expansion is formed into a cavity array, a diameter of the cavity is 700 - 1000 micron, only there is the pore space linkage among many adjacent cavities, so that the whole air tightness is not satisfactory.

3.4. Testing of sealing performance

The testing method of the sealing performance is to take a glass reinforced pipe with length of 2m and inner diameter of 75mm to seal its middle by 1m long, if the sealing material is solidified after 5h, its left side at 0.5m results in an air column, the initial pressure is atmospheric pressure, i.e. 0.1 MPa. If the vacuum pump is used to exhaust air, the maximum pressure could achieve negative pressure. The valve of the air exhaust pipe should be closed after the negative pressure achieves the maximum value, and then the rising again course of pressure on the vacuum meter is registered. Thus, the better is the air tightness of sealing material, the greater is the air exhaust negative pressure, and the slower is the rising again velocity of pressure. In process of testing, such three kinds of sealing materials as composite sealing material, high-water material and polyurethane are used to carry out the comparative test. The device used for testing the sealing performance of sealing materials is shown in Fig.9.



1-gas suction pipe;2-deformation rubber;3-grouting pipe;4-hand grout injector; 5-negative pressure meter; 6-vacuum machine;7-glass reinforced pipe

Fig.9 Testing device for sealing performance of materials



Fig.10 Changing curve of negative pressure when sealing of three kinds of materials

A relation curve between time and negative pressure of air room when sealing by polyurethane, composite sealing material and high-water material is shown in Fig.10. From Fig.10, it can be seen that the maximum laboratory negative pressure of sealing air room of high-water material is 90 kPa, after the air exhaust valve is closed for 50min, the air pressure is reverted to the initial value and the attenuation velocity of negative pressure is slower. The maximum laboratory negative pressure of sealing air room of composite sealing material is 96 kPa, after the air exhaust valve is closed for 90min, the air pressure is reverted to the initial value, and the curve slope in a whole course is reduced gradually. The velocity of negative pressure reduction is slower than that of high-water material. Therefore, the air tightness has achieved a demand for gas suction, the sealing performance is better than that of high-water material.

The maximum laboratory negative pressure of sealing air room of polyurethane is 37kPa, after the air exhaust valve is closed for 10min, the air pressure is reverted to the initial value. and the curve slope in a whole course is bigger and the velocity of negative pressure reduction is very rapid. It indicated that if the length of sealed section is identical, the sealed result of the composite sealing material is better than that of polyurethane.

The testing results of the sealing performance showed that there is a big negative pressure in sealed air room of the composite sealing material, the velocity of negative pressure reduction is slower, and its sealing performance is better than that of high-water material and polyurethane.

4. Industrial testing

Field boreholes are regarded as the boreholes of gas suction in current coal seam. The No. F_{16} -23070 working face of fourth coal mine in Pingdingshan Mining Group is selected as the testing site. The gas content of the coal seam is $11\text{m}^3/\text{t}$, the dip angle of the coal seam is $6^\circ \sim 8^\circ$, the direction of drilling is from air roadway to bedding of the coal seam, because the stress state of coal in this place is even not influenced by mining activity, and the rigidity of the coal sample is proper and drilling is provided with representation. In process of the field testing, 9 boreholes are arranged aggregately. Every three boreholes belong to a group and are sealed by polyurethane, high-water material and composite sealing material respectively, and after net-connecting suction the attenuation change of the gas concentration should be registered. The arrangement of boreholes is shown in Fig.11

The average gas concentration of every group of boreholes is shown in Fig.12. The results of industrial testing indicated that after using three kinds of the sealing materials for borehole sealing, the higher suction concentration at the initial stages of suction could be all maintained, the initial concentration of borehole sealed by the composite sealing material is 88%, the initial concentration of borehole sealed by the high-water material is 62%, and the initial concentration of borehole sealed by polyurethane is 81.5%. The gas concentration of borehole sealed by polyurethane after suction for 4d is rapidly attenuated below 30%, and then the gas concentration is lowered gradually, ultimately, the gas concentration for 5d is rapidly attenuated below 30%, and then the gas concentration is lowered gradually, ultimately, the gas concentration for 5d is rapidly attenuated below 30%, and then the gas concentration is lowered gradually, ultimately, ultimately, the gas concentration for 5d starts attenuating slowly, and finally the gas concentration is kept by $10 \sim 12\%$. The gas concentration of borehole sealed by about 30% for two months. It showed that the suction concentration of borehole sealed by polyurethane and high-water material is obviously increased better than that of boreholes sealed by polyurethane and high-water material. So, the composite sealing material has possessed a favorable sealing performance^[9].



Fig.11 Diagram of borehole in current coal seam



Fig.12 Diagram of suction concentration change when using different materials of borehole sealing

5. Conclusions

- First point, taking cement as base material mixed by expansion agent, additive, fibrin and coupling, a new composite sealing material has been developed. This material is able to meet the requirements of strength, expansion performance, sealing performance and so on.
- Second point, the expansion coefficient of the composite sealing material is 1.29 and the stable time of expansion is short; the observing results of microcosmic structure indicated that the composite sealing material presents whole density, however, polyurethane after expansion is formed into a cavity array, and there exist the pore space linkages among cavities. Therefore, the whole air tightness of the sealing material through pore space linkage is not satisfactory.
- And so on, the initial concentration of borehole sealed by composite sealing material after borehole sealing is 88%, the attenuation velocity is slower, finally, and the gas concentration is maintained by about 30%. It showed that the composite sealing material is provided with favorable sealing performance suitable for the borehole sealing of gas suction in coal seam, as well as may provide the technological guarantee for high-effect gas suction.

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