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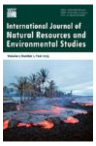

Evolution Characteristics of Coal Pore Structure with Nitrogen Hydraulic Composite Transformation

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ABSTRACT

China has abundant coalbed methane resources, comparable to conventional natural gas, with huge development potential. However, low-permeability and low-pressure coal reservoirs are widely developed, and single well production is generally low. Nitrogen hydraulic composite transformation is a potential technology to break through the efficient development of coalbed methane in low-permeability and low-pressure coal reservoirs. In order to study the influence of nitrogen hydraulic composite action on the evolution characteristics of pore and fracture structure in coal reservoirs, the Gaohe No. 3 coal in Lu'an mining area was taken as the research object. A nitrogen hydraulic composite transformation experimental device was built to carry out nitrogen transformation and nitrogen hydraulic composite transformation related experiments. The original coal samples and the coal samples modified by three methods were compared and analyzed through high-pressure mercury injection experiments to characterize the pore structure characteristics of the coal samples in the study area. The conclusion is as follows: The nitrogen hydraulic composite transformation in the Lu'an mining area has the effect of expanding and increasing the capacity of pores, causing the expansion and connectivity of transitionpores in coal to transform into large, medium, and micro cracks. The pore volume in the stage of pore size below 2nm is smaller after nitrogen and nitrogen hydraulic composite transformation, while the pore volume in the stage of pore size above 2nm increases; The improvement effect of pores after nitrogen transformation is relatively significant, and the pores are further improved after nitrogen hydraulic composite transformation.

KEYWORDS

Coalbed methane; Nitrogen hydraulic composite transformation; Pore structure characteristics.

1. INTRODUCTION

China's coalbed methane development started relatively late and has made certain progress in the exploitation of coalbed methane mainly composed of high-grade coal. The geological conditions of coal seams in China are complex, with significant regional differences and characteristics such as strong heterogeneity, low permeability, and low pressure. Although conventional hydraulic fracturing technology has increased coalbed methane production to a certain extent, it has not achieved the expected increase in production [1-5]. The gas water composite fracturing technology combines the advantages of gas fracturing and hydraulic fracturing. It not only has the advantages of promoting methane desorption, increasing formation energy, improving fracturing fluid backflow, reducing reservoir damage, and complex fracturing, but also has the advantages of good sand carrying support effect, large fracture size, and low cost in hydraulic fracturing. The principles of gas water composite fracturing and high-energy gas hydraulic composite fracturing are different. High energy gas hydraulic composite fracturing technology uses high-energy gas to impact the target reservoir before hydraulic fracturing, changing the stress state of the reservoir in the near wellbore zone. It is a local production increase measure with relatively small gas volume. Its essence is a high-pressure pulse fracturing, combined with subsequent hydraulic fracturing, with the aim of forming multiple fractures (Yang et al., 1992; Sun Qingyou, 2010); Tian Lin, 2020). Gas water composite fracturing technology is a quasi-static or a combination of two quasi-static processes in which gas and water are mixed or sequentially injected into the reservoir. With a relatively large amount of gas, it is a regional stimulation measure aimed at forming a complex fracture network and reducing reservoir damage.

The core of nitrogen hydraulic composite fracturing technology is to use the combination of nitrogen and active water to fracture and transform coal reservoirs, improve the permeability of coal seams, and increase the production of coalbed methane [6-8]. The nitrogen hydraulic composite fracturing technology not only has the advantages of gas fracturing, such as promoting methane desorption, increasing the backflow rate of fracturing fluid, reducing damage to reservoirs, and creating complex fracture networks, but also combines the strengths of hydraulic fracturing, including its excellent sand carrying support effect, the ability to form large-sized fractures, and relatively low cost. This composite application makes nitrogen hydraulic composite fracturing technology more advantageous in coalbed methane development, improving coalbed methane extraction efficiency, reducing development risks, and better adapting to complex geological conditions.

The nitrogen hydraulic composite fracturing technology has achieved significant results in the development of low-permeability and low-pressure coal reservoirs [9,10]. Tuha Oilfield Company, Lu'an Group, and others have applied nitrogen hydraulic fracturing technology in low-pressure, low-permeability, and low porosity coal reservoirs, resulting in significant improvements in flowback rate and daily gas production. Cao Yunxing et al. applied the efficient development technology of horizontal well dense multi cluster fracturing in the Daning oilfield in Shanxi Province, combined with a nitrogen pre fracturing+active water fracturing system, achieving a daily output of 20-36 times that of vertical wells in the same section and more than 10 times that of multi branch horizontal wells in the same well site. Ni Xiaoming et al. [11] compared the critical desorption pressure of hydraulic fracturing with nitrogen injection for enhanced energy fracturing and conventional active water fracturing in the Tunliu well field. The results showed that hydraulic fracturing with nitrogen injection can increase the critical desorption pressure during the initial production stage of coalbed methane wells. Under the same other conditions, it can to some extent improve the recovery rate of coalbed methane wells. Zhao Zhenfeng et al. proposed an integrated nitrogen injection gas fracturing process for deep shale gas enhancement and drainage, which formed a complex fracture network through segmented and multi cluster volume fracturing of horizontal wells. Continuous drainage was achieved through nitrogen energy enhancement and post pressure controlled drainage, resulting in a breakthrough in marine shale gas production in the Ordos Basin.

In summary, scholars at home and abroad have conducted extensive theoretical, experimental, and practical research on the impact characteristics and production mechanisms of gas fracturing on coal rock pores, as well as the fracture characteristics of gas/water fracturing. However, there is relatively little research on the evolution of nanopore structure in coal reservoirs through nitrogen hydraulic composite transformation (i.e. nitrogen hydraulic composite transformation). Based on this, this article mainly focuses on the changes in nanopore and fracture characteristics of coal reservoirs before and after nitrogen hydraulic composite transformation, and the expected results have reference significance for the production improvement of low yield coal reservoirs in China.

2. EXPERIMENTS

2.1. Introduction to the Study Area

The Gaohe Mine in Lu'an is located on the west side of the Changzhi area in the Qinshui Basin. The Lu'an mining area is located in the eastern part of the Qinshui coalfield, with the eastern boundary being the coal seam outcrop line. It borders the Qinyuan and Anze mining areas to the west, the Yangquan mining area to the north, and the Jincheng mining area to the south. Administratively under the jurisdiction of counties and districts such as Wuxiang, Xiangyuan, Lucheng, Changzhi City, Tunliu, Qinxian, Huguan, Changzi County, Yushe and Zuoquan in Jinzhong City. The length from north to south is about 120km, the width from east to west is 48-77km, and the area is 7098.00km2. The No. 3 coal seam in the middle and lower parts of the Shanxi Formation and the No. 15 coal seam in the lower part of the Taiyuan Formation are stable and mineable coal seams in the entire region, mainly consisting of lean and high calorific coal, with locally developed anthracite coal. The Shanxi Formation contains 1-6 coal layers, generally 2-3 layers, numbered from top to bottom as 1, 2, and 3. Among them, the 3 coal seam has stable occurrence and can be mined throughout the entire area. This study is based on the Gaohe Mine (GH) coal mining area in the Lu'an mining area.

2.2. Sample Preparation

The sample is taken from the No. 3 coal seam of the West Formation of Gaohe Mine in Lu'an. Fresh block coal samples will be collected and sealed according to the national standards GB/T19222-2003 and GB/T16773-2008 on the working face.

Preparation of high-pressure mercury intrusion test samples: Transport the large sample to the laboratory, and use a hammer to knock the remaining coal blocks into 1-2cm3 coal blocks to prepare several samples.

The moisture, ash content, volatile matter, and fixed carbon content of the samples from the Lu'an mining area are 0.84%, 19.70%, 13.74%, and 65.72%, respectively. The C, H, O, and N element contents are 87.40%, 3.94%, 2.27%, and 1.07%, respectively. The maximum vitrinite reflectance is 2.1%, indicating that the coal is poor.

Table 1. Basic parameters of the coal samples

Proximate analysis (%)				Elemental analysis (%)				$R_{0,\ max\ (\%)}$
M_{ad}	A_{ad}	V_{ad}	FC_{ad}	C_{daf}	H_{daf}	O_{daf}	N_{daf}	
0.84	19.70	13.74	65.72	87.40	3.94	2.27	1.07	2.1

M_{ad}: air-dry moisture content; A_{ad}: air-dry ash yield; V_{ad}: air-dry volatile matter; FC_{ad}: air-dry fixed carbon; C_{daf}: dry ash-free basis *C*; H*daf*: dry ash-free basis *H*; O_{daf}: dry ash-free basis *O*; N_{daf}: dry ash-free basis N; $R_{o,\ max}$: vitrinite maximum reflectance.

2.3. Experimental Procedure

In order to carry out nitrogen hydraulic composite transformation experiments, a nitrogen hydraulic composite transformation experimental device was built, and nitrogen and nitrogen hydraulic composite transformation experiments were conducted under static water pressure conditions, as well as hydraulic fracturing experiments of the control group.

1) Nitrogen hydraulic fracturing pore experiment: Conduct a 12MPa nitrogen-16MPa hydraulic pressurization experiment in a closed tank, with a nitrogen pressurization time of 12 hours and a hydraulic pressurization time of 2 hours.

① Take about 250g of 1-2cm coal blocks each and place them in cylinder 1 and cylinder 2. Then inject high-pressure nitrogen gas, open the valve between cylinder 1 and cylinder 2, and inject high-pressure nitrogen gas from the upper part of cylinder 1 to 12MPa for 12 hours. After the fracturing is completed, take out the coal sample from cylinder 2 and conduct high-pressure mercury injection experiments on some of the coal blocks. Three samples are taken in parallel for each experiment.

② Close the valve between cylinder 1 and cylinder 2, inject high-pressure water from the lower part of cylinder 1 to completely discharge nitrogen gas, and continue to inject water until the design pressure is 16MPa. After 2 hours, release the pressure, remove the lump coal from cylinder 1, and conduct high-pressure mercury injection experiments on some of the lump coal. Three parallel samples are taken for each experiment.

2) Control group hydraulic fracturing pore experiment: Take about 250g of 1-2cm coal blocks and put them into cylinder 2, then inject high-pressure water to 16MPa for 2 hours, take out coal samples, and conduct high-pressure mercury injection experiments on some coal blocks. Three samples are taken in parallel for each experiment. (Hydraulic fracturing experiment only for later comparative analysis)

2.4. High pressure mercury injection

High pressure mercury injection (HPMI), also known as mercury porosity method, is a commonly used method for determining the pore structure of coal. Mercury has non wettability on the surface of coal, and external force is required to allow mercury to enter the pores of coal. When the pressure is higher than the capillary pressure corresponding to the pore throat, mercury enters the pores of the coal sample. At this time, the injection pressure is equal to the capillary pressure, and the corresponding capillary radius is the pore throat radius. The volume of mercury entering the pores is the pore volume connected to the throat. The larger the injection pressure, the smaller the pore radius that mercury can enter, and the amount of mercury injected is a function of pressure. By continuously changing the injection pressure, the pore distribution curve and capillary pressure curve can be obtained. The mercury intrusion meter records the relationship between injection pressure and the amount of mercury injected into the pores of coal samples, and obtains the mercury intrusion curve. By analyzing this curve, parameters such as pore volume, pore specific surface area, and pore size distribution are obtained.

The mercury intrusion method for determining pores is based on the cylindrical capillary model, and the relationship between pressure and pore size is given by the Washburn equation. The calculation formula is:

$$P_c = -\frac{2\sigma\cos\theta}{r} \tag{1}$$

In the formula, Pc represents capillary pressure, MPa; σ is the interfacial tension between mercury and air, with a value of 0.480N/m; θ is the wetting angle between mercury and coal, with a value of 140 ^{o}, and r is the pore radius, μm。 The relationship between capillary pressure and pore radius can be obtained from formula (4-1) as follows:

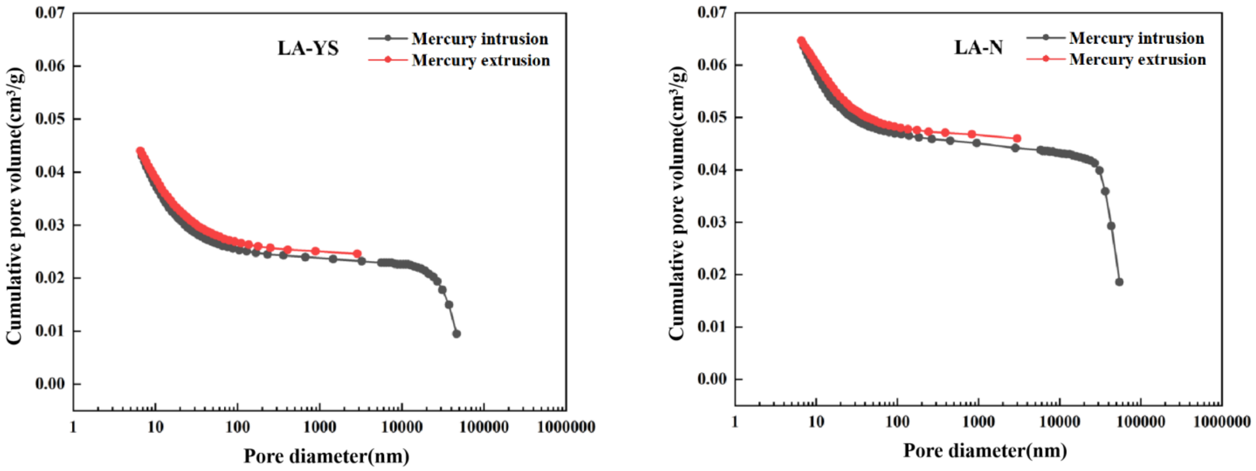
$$r = \frac{0.735}{P_c} \tag{2}$$

The mercury intrusion experiment was conducted using the AutoPore IV 9510 fully automatic mercury intrusion meter from the American company Mack (Figure 1). The experimental process followed GB/T21650.1-2008, and the coal sample size was approximately 1cm3. Dry to constant weight at 105 °C before testing. The mercury intrusion test includes two processes: pressurized mercury injection and depressurized mercury removal.

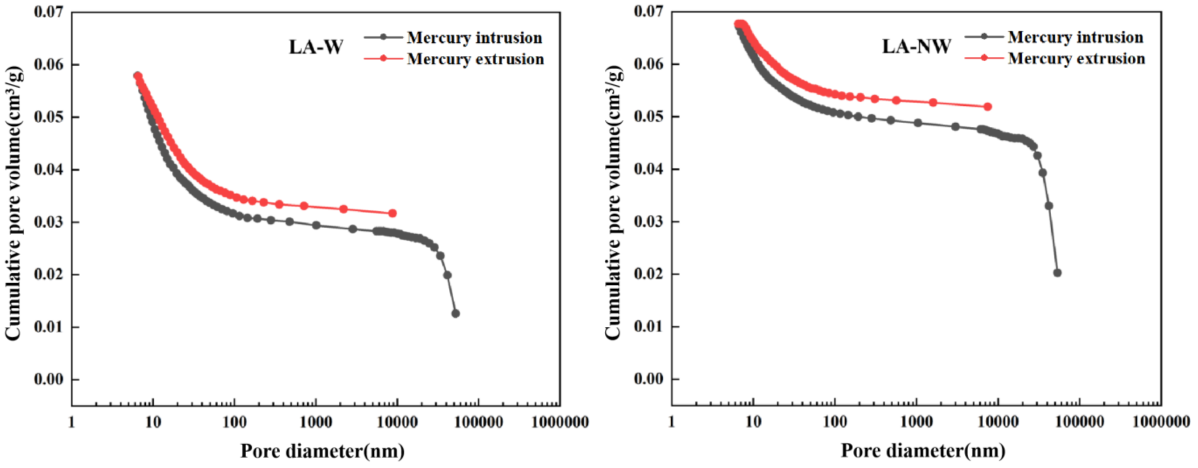
Figure 1. AutoPore IV 9510mercury porosimeter

3. ANALYSIS

3.1. Analysis of high-pressure mercury injection loop



(a)Original mercury injection loop (b) Mercury injection loop after nitrogen transformation



(c) Hydraulic transformation mercury injection loop (d) Nitrogen hydraulic composite transformation mercury injection loop

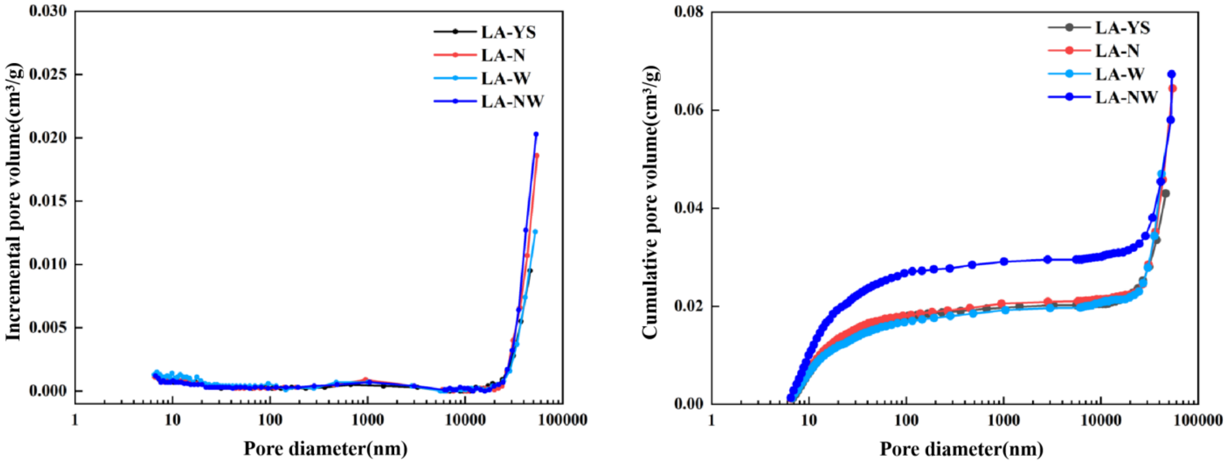
Figure 2. Mercury injection loop before and after the renovation of Lu'an mining area

The advance and retreat mercury curves of coal samples from different technological transformations in the Lu'an mining area are shown in the figure. After nitrogen transformation, the cumulative pore volume increased significantly, and there was no significant change in the hysteresis loop. After hydraulic fracturing, the cumulative increase in pore volume is relatively small, and the hysteresis loop increases, reflecting the enhancement of pore connectivity. After the nitrogen hydraulic composite transformation, the hysteresis loop significantly increased and the pore volume significantly increased, indicating an increase in the connectivity of internal pores in the coal sample, and an increase in open pores with good internal connectivity.

The advance and retreat mercury curves exhibit a phenomenon of non coincidence. The original Lu'an coal sample mercury injection curve was a non coincidence curve, but after transformation, it has been transformed into different degrees of non coincidence curves, reflecting the existence of well connected open pores inside the transformed coal sample. At the same time, this also indicates that the heterogeneity of the pore structure inside the coal body is very strong, and there are huge differences in the connectivity between pores.

3.2. Distribution characteristics of mercury intrusion pore structure

The comparison of stage pore volume and cumulative pore volume of coal samples after original, nitrogen, hydraulic fracturing, and nitrogen hydraulic composite transformation in the Lu'an mining area is shown in the figure. The distribution characteristics of pore volume in the four coal samples are basically consistent, with pores larger than 10000nm being the most developed, pores between 1-100nm being more developed, and pores between 100-1000nm being the second most developed. The total pore volume of the original coal sample is 0.043cm3/g, and after nitrogen transformation, the total pore volume is 0.064cm3/g, an increase of 0.021cm3/g. After nitrogen hydraulic composite transformation, the total pore volume is 0.067cm3/g, an increase of 0.024cm3/g, which is greater than the nitrogen transformation. The pore volume of the control group after hydraulic fracturing transformation was 0.058cm3/g, with a smaller increase compared to the nitrogen hydraulic composite transformation.



(a)Mercury injection test for stage pore volume (b) Mercury injection test for cumulative pore volume

Figure 3. Comparison of pore volume before and after the renovation of Lu'an mining area

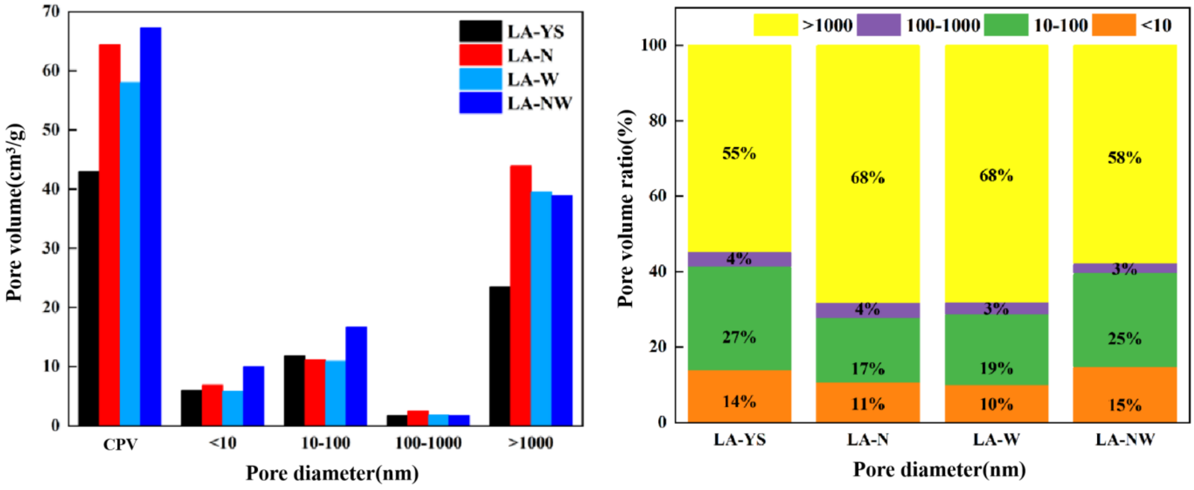
Using the Houghton decimal division method for pores, the pores measured in mercury intrusion data are divided into micropores (pore size<10nm), transitionpores (pore size 10-100nm), mesopores (pore size 100-1000nm), and macropores (pore size>1000nm). The comparison of pore volume of experimental coal samples before and after hydraulic fracturing using different techniques in Lu'an mining area is shown in the chart.

The total pore volume of the original coal sample is 43×10-4cm3/g, with a large pore volume of 23.5×10-4cm3/g, accounting for 54.65% of the total pore volume; The mesopore volume is 1.7×10-4cm3/g, accounting for 3.95% of the total volume; The transitionpores volume is 11.8×10-4cm3/g, and the proportion of transitionpores volume reaches 27.44%; The micropore volume is 6×10-4cm3/g. The proportion of micropores reaches 13.95%.

The total pore volume of the coal sample after nitrogen transformation was 64.4×10-4cm3/g, and the macropores, mesopores, transitionpores and micropores were 43.9×10-4cm3/g, 2.5×10-4cm3/g, 11.1×10-4cm3/g and 6.9×10-4cm3/g, respectively. The proportions were 68.17%, 3.88%, 17.24% and 10.71%, respectively.

The total pore volume of the coal sample after nitrogen-hydraulic composite transformation was 67.3×10-4cm3/g, and the macropores, medium pores, transitionpores and micropores were 38.9×10-4cm3/g, 1.7×10-4cm3/g, 16.7×10-4cm3/g and 10×10-4cm3/g, respectively. The proportions were 57.8%, 2.53%, 24.81% and 14.86%, respectively.

The total pore volume of the coal samples in the control group was 58×10-4cm3/g, and the macropores, medium-holes, transitionpores and micropores were 39.5×10-4cm3/g, 1.8×10-4cm3/g, 10.9×10-4cm3/g and 5.8×10-4cm3/g, respectively. The proportions were 68.1%, 3.1%, 18.79% and 10% respectively.



(a)Mercury injection test for stage pore volume (b) Mercury injection test for pore volume ratio

Figure 4. Changes in pore volume during mercury intrusion testing in Lu'an mining area

Table 2. Mercury injection test for stage pore volume

Pore diameter(nm)	Pore volume(10^{-4}cm^3/g)			
	LA-YS	LA-N	LA-W	LA-NW
<10	6.00	6.90	5.80	10.00
10-100	11.80	11.10	10.90	16.70
100-1000	1.70	2.50	1.80	1.70
>1000	23.50	43.90	39.50	38.90
CPV	43.00	64.40	58.00	67.30

Compared with the original coal sample, the pore volume of the coal sample after nitrogen hydraulic composite transformation in the Lu'an mining area is more prominent than that of nitrogen compression and hydraulic fracturing transformation. After nitrogen water composite fracturing, the total pore volume of the coal sample increased from 43×10-4cm3/g to 67.3×10-4cm3/g, with an increase ratio of 56.51%. The increase in seepage holes (macropores) is significant, with an increase ratio of 65.53%; The adsorption pores (transitionpores and micropores) showed a significant increase, with transitionpores increasing by 41.53% and micro pores increasing by 66.67%. After nitrogen hydraulic composite fracturing, the pore connectivity inside the coal body is enhanced, increasing the pore space for coalbed methane seepage and effectively improving the permeability of the coal sample.

The changes in specific surface area of each pore size segment of the original coal sample, nitrogen modified, hydraulic modified, and nitrogen hydraulic composite modified coal samples are shown in the table.

The total specific surface area of the original coal sample is 46.52m2/g. The specific surface area of macropores is 0.13m2/g, the specific surface area of mesopores is 0.12m2/g, the specific surface area of micropores is 18.35m2/g, and the specific surface area of micropores is 27.93m2/g. After nitrogen transformation, the total specific surface area of the coal sample was 48.91m2/g, with an increase of 5.13%; After hydraulic fracturing, the total specific surface area was 47.62m2/g, with an increase of 2.36%; After nitrogen hydraulic composite transformation, the total specific surface area was 64.45m2/g, with an increase of 38.54%. Micropores and transitionpores are mainly adsorption sites for coalbed methane, and the high proportion of specific surface area of these pores indicates that the coal sample has a relatively strong adsorption capacity; Before the transformation, the proportion of mesopores and macropores in the original coal sample was only 0.52%. After the nitrogen hydraulic composite transformation, the proportion decreased to 0.46%, and these types of pores are mainly channels for coalbed methane seepage.

Table 3. Mercury intrusion test specific pore area data in Lu'an mining area

Pore diameter(nm)	Pore area(m^2/g)			
	LA-YS	LA-N	LA-W	LA-NW
<10	27.9260	32.1870	28.4880	36.6670
10-100	18.3466	16.4391	18.9049	27.4821
100-1000	0.1155	0.1313	0.1021	0.1229
>1000	0.1276	0.1609	0.1221	0.1789
CPA	46.5157	48.9183	47.6171	64.4508

4. CONCLUSIONS

Low permeability and low-pressure coal reservoirs are widely developed in China, but the development effect is poor. The research and development of production increasing technology for low-permeability and low-pressure coal reservoirs must focus on efficient permeability enhancement and reservoir protection. Traditional hydraulic fracturing technology has some drawbacks. The "Nitrogen Hydraulic Composite Fracturing Technology" integrates both nitrogen and hydraulic fracturing techniques, achieving the dual goals of efficient permeability enhancement and reservoir protection simultaneously. This study takes the No. 3 coal in the West Formation of Gaohe Mine in Lu'an as the research object, and uses high-pressure mercury intrusion testing technology to study the evolution characteristics of the pore results of nitrogen hydraulic composite transformation coal samples. The main conclusions are as follows:

1) After the nitrogen hydraulic composite transformation in the Lu'an mining area, the cumulative pore volume increased from 0.043cm3/g to 0.067cm3/g, with a pore volume increase of 0.024cm3/g, the largest increase, mainly provided by 10-100nm pores, and the specific surface area increased from 46.52m2/g to 64.45m2/g, mainly provided by micropores and macropores.

2) The nitrogen hydraulic composite transformation has a better effect on enlarging and expanding coal pores than nitrogen transformation and hydraulic transformation. This study has certain reference value for the development of low-permeability and low-pressure coalbed methane in coal reservoirs.

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