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Natural gas releasing simulation experiment of coal in process of temperature decreasing and decompression and preliminary application in Ordos Basin

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Abstract **Natural gas releasing simulation experiments were carried out in laboratory for researching the gas storage capacity in state of high temperature and high pressure and its gas releasing potential in process of temperature decreasing and decompression. The exiting phase state was studied through measuring gas adsorption of coal and PVT phase calculating of natural gas. Gas volume, gas molecular and isotope compositions in process of temperature decreasing and decompression were measured, natural-gas yields released from the Upper Paleozoic coal strata after later Cretaceous (K_3) were calculated and the formation of the reservoir was studied combining with the geological background. The results indicate that natural gas stored in coal has still bigger releasing potential after the uplift of Upper Paleozoic strata. There exists a weak gas supply-effluent equilibrium in the reservoir of Ordos Basin, which is another possible evidence that the Upper Paleozoic gas reservoir may be a deep basin gas reservoir.**

Keywords: coal, temperature decreasing, decompression, natural gas releasing, simulation experiment, Ordos Basin.

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There are two reasons to research the gas releasing in process of temperature decreasing and decompression: the first is that there are still a lot of gases releasing from coal after stopping heating in laboratory simulation[1], and the second is because of the special properties of Upper Paleozoic reservoir in Ordos Basin, that is the co-existing of organic matter high-enriching source rock (coal-bearing strata) and low-density gas reservoir. The source rocks of Upper Paleozoic reservoir in Ordos Basin are coal strata which have big potential of gas generation due to high-enriching organic matter, high maturation and good porosity and permeability, while the main reservoir rocks are lower porosity and permeability sandstone (the porosity is about 5%—10% and permeability is 0.2×10^{-3}—6×10^{-3} μm^2)[2]. The gas reservoir formed in this source-reservoir assembling is mainly high concentrating and high density one; however, the exploration indicates that gas reservoirs in Erdos Basin are almost low concentration, low pressure and low production reservoirs[3]. There are still debates on the formation of the reservoir. Some researchers thought that it is the deep basin gas reservoir[4—6]. The main evidences include lithologic trap of fluvial and delta facies sediments, lower porosity and permeability reservoir rocks, simple basin structure, fast subsidence in early stage and later uplift (esp. in the mid-eastern areas) and a regional gas-water up-down setting and water-sealing in east-north edge of the Basin, which are similar to the Alberta Basin in Western Canada[5,6]. Comparing with the Alberta Basin, the key to making the judgment is whether there exists a dynamic equilibrium of gas supply-effluent[7]. It is proven that there existed a lot of oil seepage and gas leakage in the northeastern area of the Basin[5], while the gas generation ability had decreased a lot due to the structural uplift and geothermal decreasing. Kinetic simulation indicated that there did not exist secondary hydrocarbon generation after uplift[8]. Therefore, the research of gas generation capacity of coal that is the main source rocks in the process of uplift (temperature decreasing and decompression) is getting very necessary.

The previous researches indicated that there exited an abnormal high geothermal field and the coal strata were buried deeply in 4000 m deep in the later Jurassic to the early Cretaceous. The abnormal high geothermal filed began to disappear after the early Cretaceous and the coal strata totally uplifted from the depth of 4000—2100 m (the maximum). It indicates that the coal strata experienced mainly the process of temperature decreasing and decompression due to the uplift after early Cretaceous[9]. The problem is whether there is the gas generation ability in this process? How much is the gas yield volume? What is the gas generation mechanism? And what are the gas components? It is one of the key factors to judge the existence of gas supply-effluent equilibrium. This paper try to probe these questions through studying the gas storage capacity of coal in the state of high temperature and high pressure, gas yield volume, gas composition and carbon isotopes in the process of temperature decreasing and decompression by combining experimental way and geological characteristics of the basin.

1 Experimental

The adsorption capacity of coal is studied by high pressures micro-gravity balance, which takes result by measuring the micro-scaled mass changes causing by gas adsorption[10]. Two equipments were designed for simulating the process of temperature decreasing and decompression. The first equipment is an open compressed vessel to simulate the decreasing of lithologic pressure by external stress from the lifting jack, and the vessel is a pyrolysis reactor. The detailed structure of this equipment is described in a reference literature[1]. It is called dry system because it only simulates lithologic pressure. Two depth points were selected according to the basin geology characters from 4000 to 2000 m, and the pressure and temperature conditions are P=104 MPa, T=400℃ for the first point and P=60 MPa, T=320℃ for the second point. The second equipment is a closed pressure vessel with a special designed external-controlled valve to simulate the fluid pressure. The valve can separate the gaseous phase and the liquid-solid phases in the state of high temperature and pressure. It is called wet system because it mainly simulates the fluid pressure. Two states were designed corresponding to the first system, which were: T=480℃, P_{fluid}=40—60 MPa for the first point and T=350℃, P_{fluid}= air pressure for the second point.

The experiment includes two processes: firstly, sample is fast-heated to the first state (450℃, 104 MPa) and maintained for 50 h until reaching the equilibrium. Secondly, the temperature and pressure are decreased slowly to the second state (320℃, 60 MPa), and the state is maintained for 30 h until reaching the equilibrium again. Gases in the two stages were collected separately. Gas yields in the two stages were also measured separately and analyzed through GC and GC-IRMS to determine the molecular and isotope compositions.

2 Results

(i) Gas adsorption capacity of coal. Coal is a kind of organic rocks with high enriching organic matter and very developed pore space, which make it adsorb a quantity of gases[10]. This may influence the gas storage in the high-pressure state and the gas releasing in the later stage. Four coal samples were measured by high pressure micro-gravity balance, and their isotherms (CH_4, 30℃) are shown in Fig. 1.

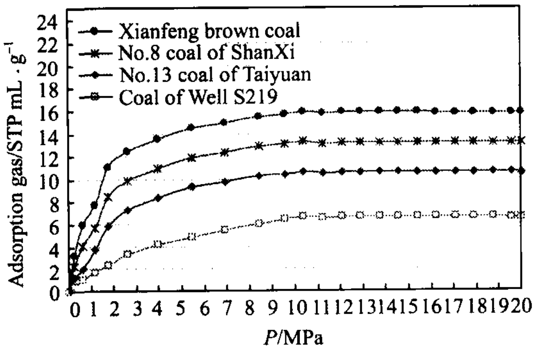

Fig. 1. Isotherms of four coal samples (CH_4, 30℃).

Many previous studies about the adsorption capacity of coal have been done[10—14]. The brown coals and coals from the Erdos Basin were measured in this experiment (Fig. 1). It could be found that the maximum gas adsorption of the Xianfeng brown coal reached 15.75 mL/g

(STP), 13.15 mL/g (STP) for No. 8 coal of Shanxi Formation, 10.55 mL/g (STP) for No. 13 coal of Shanxi formation and 6.55 mL/g (STP) for No. 13 coal of Shanxi formation. In general, the maximum gas adsorptions of coal in Erdos Basin were of about 6.55—13.15 mL/g (STP) and 10.08 mL/g (STP) in average. At the meanwhile, the adsorption capacity was also connected to coal components and maturity, which demonstrated the coincided results with the previous researches[10—15].

(ii) Gas yielding volume of coal in the process of temperature decreasing and decompression. The simulation experimental results by the above two equipments are listed in Table 1. It can be found that plentiful gases are generated in the simulation process of temperature decreasing and decompression. The gases yielding in the dry system is between 19.4 and 29.0 mL/g, and 14.55 and 79.5 mL/g in wet system. The gases generated in the process of temperature decreasing and decompression could be regarded as the releasing gases stored in coal because the equilibrium has been reached in the first point and there should be no extra gases generated in this process. The results also indicate that the coal had the higher gas storage capacity. Taking this experiment as an example, the gas storage capacity in the condition of 400—450℃, 104 MP (about 4000 m depth underground) is about 15—79.5 mL/g. If the first point could be regarded as the end the pyrolysis generation, the process between the first and the second points could be regarded as the releasing process of gases stored in coal. It could be found that 55%—75% (79% in maximum) of the total gas generation had been expelled after saturated in coal in early stages and the gases stored in coal accounted for 25%—45% (57% in maxim) um of the total gas generation.

The fluid pressure is not much higher while simulating in the wet system because the vessel is open, which could be found from Table 1 that the fluid pressure is less than 6 MPa in general. It is completely different from the actual geologic condition. From the simulation of pressure evolution history, the palaeo-pressure reaches 50 MPa or larger in Erdos Basin. For getting the higher fluid pressure, closed heating method was taken through switching off the value and increasing sample weight and compactedness. Through these methods, the simulating fluid pressure may reach 30—51 MPa (the last two samples in Table 1), which makes it possible to research the gas storage capacity and gas releasing of coal in high-fluid-pressure state. From the results of the above two samples, it could be found that the released gas volume increased a lot as the increasing of fluid pressure, which reached 136 mL/g while the fluid pressure reached 51 MPa (isothermal state).

For predicting the gas releasing volume in the process of temperature decreasing and decompression, the regression analyses between the releasing gas volume and

Table 1 Gas yields in the simulated process of the temperature decreasing and decompression

System	No.	Samples/g	Temp./℃	Pressure/MPa	Gas yield/mL	Per capita gas yield/mL • g^{-1}	Stage gas yield/ mL • g^{-1}	Stage proportion (%)
Dry	WL-3	Brown coal, Liaohe	450	104	629	29.30	20.89	71.30
		30	350	60	250		8.41	28.70
	WL-4	Brown coal, Liaohe	400	104	475	19.38	11.88	61.30
		40	320	60	300		7.50	38.70
Wet	WL-5	Brown coal, Xianfeng	450	5	112	14.55	6.27	43.10
		18	350	2	150		8.32	57.20
	WL-6	Brown coal, Xianfeng	450	6	850	73.25	42.49	58.00
		20	350	2	615		30.77	42.00
	WL-7	Brown coal, Xianfeng	450	8.2	950	79.50	47.46	59.70
		20	350	4	640		32.04	40.30
	WL-8	No.13 coal, C_{2t}	450	4.1	826	60.80	41.30	67.93
		20	350	2.4	390		19.50	32.07
	WL-9	No.8 coal, P_{2sh}	450	4.4	975	70.25	48.75	69.40
		20	350	1	430		21.50	30.60
	WL-10	Well 219 coal, C_{2t}	450	1	225	15.50	11.25	72.58
		20	350	0.4	85		4.25	27.42
	WL-15 a)	No.8 coal, P_{2sh}	580	26—13	3365	79.55	42.30	33.65
		10	300	13—0	4590		57.70	45.90
	WL-16 a)	Vitrain, Fushun	510	51—25	3845	136.0	28.30	38.45
		100	510	25—0	9764		71.70	97.64

a) Closed heating for increase temperature and pressure in first stage.

the maximum fluid pressure (excluding the sample of WL-16 due to the isothermal decompression process) were used. The regression plot is shown in Fig. 2, and the regression formula is $y = 0.0001x^3 - 0.0552x^2 + 4.4189x - 0.2467$ ($R^2 = 0.9746$), among which x is the maximum fluid pressure (MPa), and y is the gas releasing volume (mL/g). It could be found that correlation coefficient was very high and R^2 reached 0.9746. This formula may be used to predict the gas releasing volume in the process of temperature decreasing and decompression.

Moreover, the average gas releasing velocity that gas volume in pressure decreasing of 1 MPa was calculated. The average gas releasing velocity is about 6.61 mL • g^{-1} • MPa^{-1} through the result statistics of the above 8 samples. It should be noticed that the gas releasing velocity varied in different pressure stages, which would be discussed later.

(iii) Gas releasing process of coal in the process of temperature decreasing and decompression. For researching the gas releasing process of coal in the process of temperature decreasing and decompression, we precisely measured the staged and accumulated releasing gas volume and velocity (Fig. 3). The experiment was carried out in wet system with 50 MPa by increasing compactedness in closed heating mode to 400℃. Then the pressure gradually decreased isothermally and the gases were collected and analyzed in different stages (Fig. 3.)

It could be found from Fig. 3 that the gas releasing velocity is different in different pressure stages. From 50 to 20 MPa, the gas releasing volume and velocity were relatively low, the accumulated releasing gas volume was about 50 mL/g, and the accumulated gas releasing velocity was about 3 mL • g^{-1} • MPa^{-1}. And from 20 to 0 MPa, the gas releasing volume and velocity were relatively high, the accumulated releasing gas volume might reach 140 mL/g, and the accumulated gas releasing velocity was about 9 mL • g^{-1} • MPa^{-1}. The staged gas releasing volume and releasing velocity had the same trends. The above results indicate that gas releasing volume and velocity are relatively low in relative high-pressure conditions (>20 MPa), and they are relatively high in relative low pressure conditions (<20 MPa). These results may be related to the gas storage state in coal.

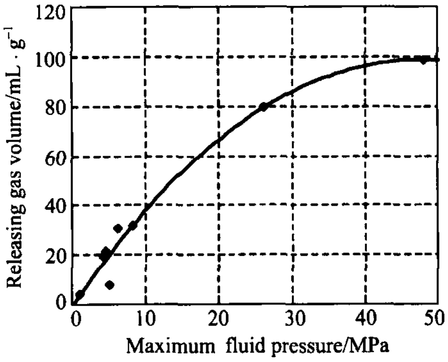

Fig. 2. Regression plot of releasing gas volume and the maximum fluid pressure.

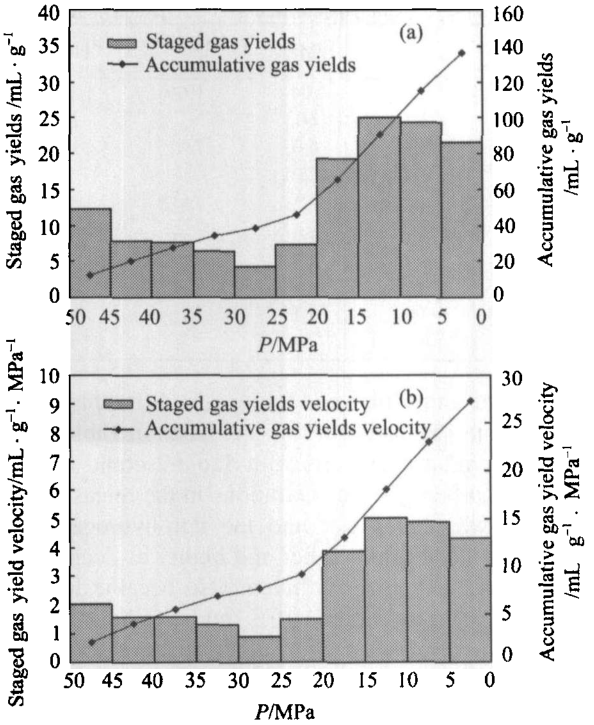

Fig. 3. Staged and accumulated releasing gas volume (up) and velocity (bottom).

(iv) Composition and isotope characters of releasing gases in the process of temperature decreasing and decompression. The molecular and isotope composition of releasing gases in the process of temperature decreasing and decompression were analyzed and the results are listed in Table 2. As mentioned above, the gases in the second stage are releasing gases. Compared with the first stage, molecular and isotope compositions of gases in the second stage have the following properties: (1) Releasing gases have high content of non-hydrocarbon gases and the content of hydrocarbon gases are 4%—6% lower than pyrolysis gases; (2) Releasing gases have lower methane, higher heavy hydrocarb (C^{2+}) and lower dryness coefficent; (3) Releasing hydrocarbon gases have lighter carbon isotope, about 3‰—4‰ lighter generally.

The above results reflect that releasing gases have lower maturation and are similar to the gases generated in earlier stages. It also indicates that this type of natural gases are releasing gases stored in coal in the earlier stage instead of the pyrolysis gases in later stage.

For researching the compositional and isotopic characteristics of releasing gases in high-pressure state, we

Table 2 Composition and isotope characters of releasing gases in the process of temperature decreasing and decompression

Sample No.	T/℃	P/MPa	Gas yield/ mL • g^{-1}	HC (%)	C_1 (%)	C^{2+} (%)	Dryness coefficient	$\delta^{13}C_1$ (‰)	$\delta^{13}C_2$ (‰)	$\delta^{13}C_3$ (‰)
WL-4	400	104	19.40	6.92	3.71	3.21	0.57	−34.54	−25.70	−27.94
	320	60		17.52	8.73	8.79	0.52	−40.63	−29.89	−28.62
WL-6	450	6.0	73.25	13.21	7.75	5.46	0.60	−35.00	−29.82	−29.98
	350	2.0		9.48	4.76	4.72	0.55	−36.76	−29.56	−27.97
WL-7	450	8.2	79.50	40.11	22.82	17.29	0.59	−31.75	−25.96	−26.51
	350	4.0		9.21	6.21	3.00	0.69	−34.51	−27.38	−27.80
WL-8	450	4.1	60.80	72.74	54.57	18.17	0.76	−30.07	−24.51	−24.35
	350	2.4		68.16	48.89	19.28	0.72	−34.55	−24.55	−25.34
WL-9	450	4.4	70.25	76. 30	61.64	14.66	0.81	−31.45	−23.02	−23.03
	350	1.0		70.99	59.40	11.59	0.84	−33.41	−23.84	−23.74

analyzed the molecular and isotope in different stages of the sample in Fig. 3. The results are listed in Table 3.

The similar characters with Table 2 could be found. The hydrocarbon gas concentrations in the releasing gases became lower and lower and the non-hydrocarbon gas concentrations became higher and higher as decreasing of pressure. The dryness coefficient also became lower and lower, the concentrations of CO_2 and CO became higher and higher, the concentration of O_2 became much lower and the carbon isotope of the hydrocarbon gases (C_1, C_2 and C_3) also became more and more light as decreasing of the pressure. The above results indicate that the releasing gases have higher maturation and the maturation of later releasing gases is relatively low. It also indicates that coal, the organic matter highly enriched and pore space developed organic rock, has very strong storing capacity of gases in high pressure state. The gases generated in earlier stage might be adsorbed and stored preferentially, and the gases generated in later stage might be stored subsequently. And the gases generated after the equilibrium might be expelled directly. And as decreasing of the pressure, the gases generated in later stage might be released firstly, that is why the gases released in later stage have relatively low maturation.

3 Preliminary application and discussion

(i) Storage phase state of gases in coal in high-pressure state. The organic matter is highly enriched in coal, which may adsorb a lot of natural gases, and the pore space is very developed in coal. The previous researches indicate that the pore size is between 0.5 and 100 nm, and the molecular size of natural gases are between 3 and 6 A[16]. That is to say the pore space is much larger relatively to the gas molecular. Therefore, there are the other existing states besides adsorption, which have been reported before[17—19]. Generally, there are three storing states of gases in coal: adsorbed, dissociated and dissolved[3]. The above results indicate that the adsorption volume of gases is about 6.55—15.75 STP mL/g and 11.5 STP mL/g in average. The volume of the releasing gases in the process of temperature decreasing and decompression is about 30—50 STP mL/g (much larger in high pressure), which is much larger than the adsorption volume. It indicates that amount of gases exist in the dissociated state. It is a vital question whether there exits the phase transition for the natural gases in coal because it will improve the gas storage capability greatly. So we researched the phase transition of natural gases in high temperature and pressure through PVTsim simulation. Fig. 4 is the phase diagram of the natural gases in Well Su-16. It can be found that there is no phase transition (i.e. gas to liquid) for pure dissociated gases in this P-T condition and it can be found there is no phase transition for most coal-deriving natural gases in this P-T condition. As for dissolved gases and whether the phase transition P-T conditions might be changes obviously could not be concluded due to the lack of study in this paper.

(ii) Preliminary application in Erdos Basin. Four types of coals including brown coal, vitrain and the coals from Taiyuan Formation and Shanxi Formation in Erdos Basin were selected in this experiment. The results indicate that the composition and maturation have definite influence to the results (Table 1). Combining with geological setting in Erdos Basin, we supposed the coal strata uplifted from 4000 to 2000 m and the corresponding pressures decreased from 40 to 20 MPa. It could be found that the gas released from Taiyuan Formation is relatively

Table 3 Composition and isotope characters of staged releasing gases in the process of temperature decreasing and decompression

Sample No.	Pressure/MPa	HC gases (%)	C_1 (%)	C^{2+} (%)	Dryness coefficient	CO_2 (%)	O_2 (%)	CO (%)	δC_1 (‰)	δC_2 (‰)	δC_3 (‰)
WL16-G-1	51—35	53.83	29.35	24.48	0.55	36.57	1.52	1.00	−28.80	−24.65	−23.85
WL16-G-2	35—15	48.04	23.26	24.78	0.48	37.20	1.43	1.59	−29.32	−25.77	−24.82
WL16-G-3	15—0	30.82	11.53	19.28	0.37	59.25	0.70	2.41	−30.81	−25.87	−25.70

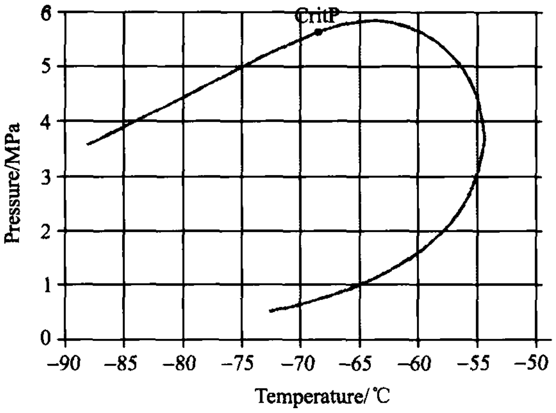

Fig. 4. The phase diagram of the natural gases in Well Su-16. The compositions (%) of natural gases include: N_2, 0.847; CO_2, 1.851; C_1, 86.964; C_2, 7.969; C_3, 1.661; iC_4, 0.277; nC_4, 0.284; iC_5, 0.106; nC_5, 0.041. Critp, critical point.

lower, which was about 4.25—19.5 and 11.88 mL/g in average, and the gas released from Shanxi Formation is relatively high, about 21.5—57.7 and 39.6 mL/g in average. The average gas released from Taiyuan Formation and Shanxi Formation is about 25.74 mL/g, and the predicted gas releasing gas volume is about 27.738 mL/g through the above regress formula. Two results are very close to each other.

From the above calculating and experimental results, it could be deduced that the released gas volume was very large from coal in Erdos Basin after the Upper Paleozoic coal uplifted from 4000 to 2000 m. Combining the releasing process (Fig. 3), it could be found that the released gas volume and velocity might be larger if the pressure decreased to lower than 20 MPa. That is to say, on one hand, there are still amounts of gases not released in the coal strata; on the other hand, the gas releasing volume might be much larger in strong uplifted area in Erdos Basin (Pressure <20 MPa).

Moreover, we can judge whether the natural gases in the west of Erdos Basin (such as Sulige Gas Field) is released from coal through the molecular and isotope composition of the gases. Actually, many researches have noticed the disagreement between the molecular and isotope composition of the gases and the maturation of source rock in this area, especially the fact that the methane carbon isotope (δC_1‰) of the gases was a little lighter and the dryness coefficient of the gases was relatively high, which were not coincident with the higher maturation of the source rocks[20]. For example, the maturation of source rocks reaches over 2.0 (R_o%) in Sulige area, while the wetness of the gas in this area is relatively higher (dryness coefficient reaches 86%) and the carbon isotope of the gas is relatively light (−29.96‰—36.45‰)[2], which could also be proved by the formula of Stahl[21]. Moreover, the concentrations of non-hydrocarbon gas in Sulige area are also relatively high compared with that of the other areas. These properties indicate that the maturation of gases is lower than the source rocks. Some previous researches regarded that it was because of the mixture of the oil-type gases from Lower Paleozoic rocks[20,22], and some previous researches regarded that it was because of the mixture of oil-type gases from Carboniferous-Permian rocks[2,23]. From the results of this experiment, the low-maturation gases might be the released gases because of the later uplift, which was originally generated in early stages and stored in the source rocks of coal. In fact, Sulige is located in the west of the basin and the uplift scale is very large, therefore the results may explain the disagreement of the maturations of gases and the source rocks of coal. If it is proved true, it can be deduced that coal may continue to provide abundant gases after the uplift and the stop of the hydrocarbon generation. Gases stored in the coal source rocks may expel the gases to the reservoir rocks, and there may exist a weak gas supply-effluent equilibrium in the basin.

Indeed, this paper mainly studies through the experimental results, so the application of above results to the geological conditions needs more deep researches because of the complexity of coal and the diversity of the pressure decreasing in actual geological conditions.

4 Conclusions

Natural gas releasing simulation experiment was carried out in this study for researching the gas storage capacity in state of high temperature and high pressure and its gas releasing potential in process of temperature decreasing and decompression. The experimental results indicate that as a kind of pore-developed organic rock, coal has a large storage capacity of gases. The adsorption volume of gases is about 6.55—15.75 and 11.5 STP mL/g in average. The volume of the released gases in the process of temperature decreasing and decompression is about 30—50 STP mL/g. Phase diagram analysis indicates that most coal-deriving gases in coal are stored in adsorption and dissociation states.

The gas releasing process of coal in the process of temperature decreasing and decompression was also researched. It was found that gas releasing volume and velocity are relatively low in relative high pressure conditions (>20 MPa), and they are relatively high in relative low pressure conditions (<20 MPa), that may be related to the gas storage state in coal. Analyzing of molecular and isotope indicated that the gases generated in the earlier stage might be adsorbed and stored preferentially, and the gases generated in the later stage might be stored subsequently. As the decreasing of pressure, the gases generated in the later stage might be released firstly. Gases released in the earlier stage have relatively high maturation and gases released in the later stage have relatively low maturation. The results might be applied to explaining the disagreement of maturations of natural gases and source rocks of coal in this area, and it was deduced that the gases in this area (i.e. Sulige Gas Field) might be the released gases from coal.

The gases are mainly released from the stored gases in coal strata while the coal strata uplifted from 4000 to 2000 m in Erdos Basin. The average gas volume released from Taiyuan Formation and Shanxi Formation is about 25.74 mL/g, and the predicted gas releasing gas volume is about 27.738 mL/g through the above regress formula. Coal might continue to provide abundant gases after the uplift and the stop of the hydrocarbon generation. Gases stored in the coal source rocks may expel the gases to reservoir rocks, and it was deduced that there might exist a weak gas supply-effluent equilibrium in the basin, which provided another evidence that the gas reservoir in Erdos Basin might be a deep basin gas reservoir.

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