

Underground Coal Mine Methane Displacement by Injecting Low-pressure Gas into the Meta-anthracite Seam: Laboratory and Field Tests

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Received 13 April 2014; Accepted 18 September 2013

Abstract

Because of the strong adsorption capacity of meta-anthracite, the gas content of a meta-anthracite seam can be as high as 10 m³/t, with a gas pressure lower than 0.74 MPa; this results in low efficiency of gas extraction in underground mines. To enhance low-pressure methane extraction efficiency in meta-anthracite seams, a new approach – methane displacement by gas injection – has been developed, investigated in the laboratory, and then applied in the field in the Fuyanshan coal mine. Laboratory results show that when the gas content of the coal seam is high, methane displacement by nitrogen injection is difficult. The volume of methane displaced is directly related to the pressure difference between the coal seam gas pressure and the injection gas pressure. If the total gas pressure is greater than 0.5 MPa after nitrogen injection, then the methane displacement efficiency will be greatly enhanced. It is also confirmed that the displacement efficiency can be improved by injecting inert gas to change the partial pressure of the methane. Field test data show quite good methane displacement efficiency.

Keywords: meta-anthracite, extraction, substitution, adsorption, gas injection

1. Introduction

Gas extraction technology has advanced considerably in recent years, with extensive theoretical and technological achievements. However, owing to the complicated geological conditions of coal beds and their low permeability, ranging from 10⁻³ to 10 m²/(MPa²·d)[1][2], it is still difficult to meet the requirements of gas extraction regulations in China, especially for the meta-anthracite coal beds that generally have a strong gas adsorption capacity. The gas content in meta-anthracite can reach up to 10 m³/t, even when the gas pressure is lower than 0.74 MPa[3]. To improve the permeability of the coal seam and to aid gas extraction and pressure relief, mechanical techniques are applied to increase the fissures and cracks. However, the rapid closures of these fissures and cracks results in a decrease in permeability as the mining depth and the *in situ* stress increase. For a meta-anthracite seam, the residual gas pressure is 0.3 MPa after gas extraction[4][5]. Under these conditions, methane can hardly be desorbed by the low negative extraction pressure in the drilling hole. Furthermore, the gas desorption efficiency induced by the mechanical techniques is also weak, so it is an urgent task to enhance the extraction efficiency of methane in low-pressure meta-anthracite seams.

Carbon dioxide was successfully injected into the coal bed to displace methane so as to increase the coal extraction rate in the San Juan Basin at the end of the twentieth century, and the extraction of methane from coal beds was also greatly improved by the injection of inert gases in Canada, Europe, Japan and China[6][7]. Studies on the adsorption capabilities of coal beds of all kinds of gases have been conducted in China by Tang Shuheng et al. (2002), Cui Yongjun (2003), Fu Xuehai and Wu Shiyue (2011). The capacities of coal for adsorbing carbon dioxide, methane and nitrogen can be ranked thus: carbon dioxide > methane > nitrogen, although this may vary depending on the characteristics of the coal. Methane displacement by nitrogen and air injection have been studied by Wang Zhaofeng (2010) and Yang Hongmin (2013), who investigated the adsorption of multi-component gases (carbon dioxide, nitrogen, methane) and the displacement and desorption effects induced by injecting carbon dioxide and nitrogen. However, previous studies focused on the adsorption and desorption laws of high-pressure gases, based on the adsorption potential theory and desorption and substitution gas theories [8]. It has been shown that low-pressure nitrogen can be injected to displace higher pressure adsorbed methane, but few studies have been conducted to investigate the desorption and displacement of methane by injection of low-pressure inert gases. To improve the desorption and adsorption theories of multi-component gases, and to elucidate the desorption and substitution

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mechanism of methane by nitrogen, the inert nitrogen is injected at low pressure (<0.74 MPa) into the meta-anthracite seam to desorb and displace the methane in a low-pressure adsorption state. This may shed light on methods to decrease the gas content in meta-anthracite, and provide a theoretical basis for improving the extraction efficiency of methane.

2. Theoretical study and the selection of gas sources

According to the adsorption capacities of carbon dioxide, methane and nitrogen ($\text{CO}_2 > \text{CH}_4 > \text{N}_2$) proposed in a previous study, carbon dioxide is adsorbed more easily by coal than methane, and so can be preserved deep underground for a long time, and can contribute to controlling the greenhouse effect; thus, injection of carbon dioxide can be used as an auxiliary technique in the methane extraction process in coal beds [9] [10]. However, owing to the potential outburst hazard in coal layers, as well as the suffocation hazard of desorbed carbon dioxide, its injection into underground coal mines should be kept to a minimum. Therefore, taking safety and environment issues into consideration, nitrogen (or air: 79.8% N_2) are preferable in the injection process. It has been shown in previous studies that injection pressures in the methane extraction process in coal beds range from 5.5 MPa to 8.0 MPa, whereas the

pressures vary much more widely – from 30 MPa to 52 MPa – in the oil extraction process [11]. The above difference in the injection pressure results from the nonexistence of a revealed line, which may prevent the high-pressured injected gas from penetrating up to the earth's surface above the coal and oil layers [12]. However, the depth of injection bores is limited, and this limitation will result in lesser resistance to preventing the injected air from penetrating through the coal layer. However, in line with the regulations for coal and gas outbursts, the pressure in the coal bed (and hence of the injected gas) should be kept below 0.74 MPa. The mechanisms and characteristics of gas injection techniques, desorption and substitution vary greatly according to the injection pressure. The present work focuses on the substitution relationship between nitrogen and methane at pressures lower than 0.74 MPa, and may provide some theoretical and experimental background for field applications.

3. Experimental set-up and procedures

3.1 Selection and preparation of coal specimens

The meta-anthracite coal specimens, which were obtained from Nan Washi Mine in Jin Cheng, Fu Yanshan Mine and Xiao Xi Mine, are characterized by a strong adsorption capability. The basic parameters are shown in Table 1.

Table 1. Basic adsorption parameters of gas in the coal samples

Specimen number	Source	Adsorption constant		Ash content (%)	Water content (%)	Porosity (%)	Density (t/m^3)
		a ($\text{m}^3/\text{t.r.}$)	b (MPa^{-1})				
1	Nan Washi Mine	56.552	1.092	11.88	1.62	4.685	1.43
2	Fu Yanshan Mine	51.361	1.116	13.03	1.19	4.172	1.44
3	Xiao Xi Mine	55.156	1.203	12.2	1.86	4.432	1.43

Based on the test results of the gas content obtained from the coal specimens and the indirect calculation equations proposed by the provisional rules of coal mine gas extraction, when the gas pressure is 0.74 MPa, the gas content in the coal bed reaches up to 13.21–13.71 m^3/t , which is much higher than the 8 m^3/t that is the maximum value specified in Prevention and control of coal and gas outburst. The content decreases to 8.5–8.68 m^3/t when the gas pressure decreases to 0.3 MPa. The corresponding gas contents at different gas pressures in the coal specimens are shown in Table 2.

The original coal samples were ground to particles, and then the particles were sieved twice, through 60 mesh and 80 mesh, giving dry coal specimens in line with the requirements of the national sampling regulations for coal seams (GB/T 482-2008).

3.2 Experimental set-up

The experimental set-up used to investigate the adsorption and desorption of the multi-components gases was designed

and manufactured by this research team. The experimental system is composed of the adsorption and desorption system at constant temperature, a high-pressure gas distribution system, a vacuum pumping system, a gas quantitative system, an isolated sampling system and a chromatographic analysis system for the gas components (shown in Fig. 1).

Table 2. Corresponding gas contents at different gas pressures

Specimen number	Source	Gas pressure (Mpa)	Gas content	
			$\text{m}^3/\text{t.r}$	m^3/t
1	Nan Washi Mine	0.3	10.03	8.68
		0.74	15.85	13.71
		0.3	10.05	8.62
2	Yanshan Mine	0.74	15.82	13.57
		0.3	9.89	8.5
3	Xiao Xi Mine	0.74	15.37	13.21

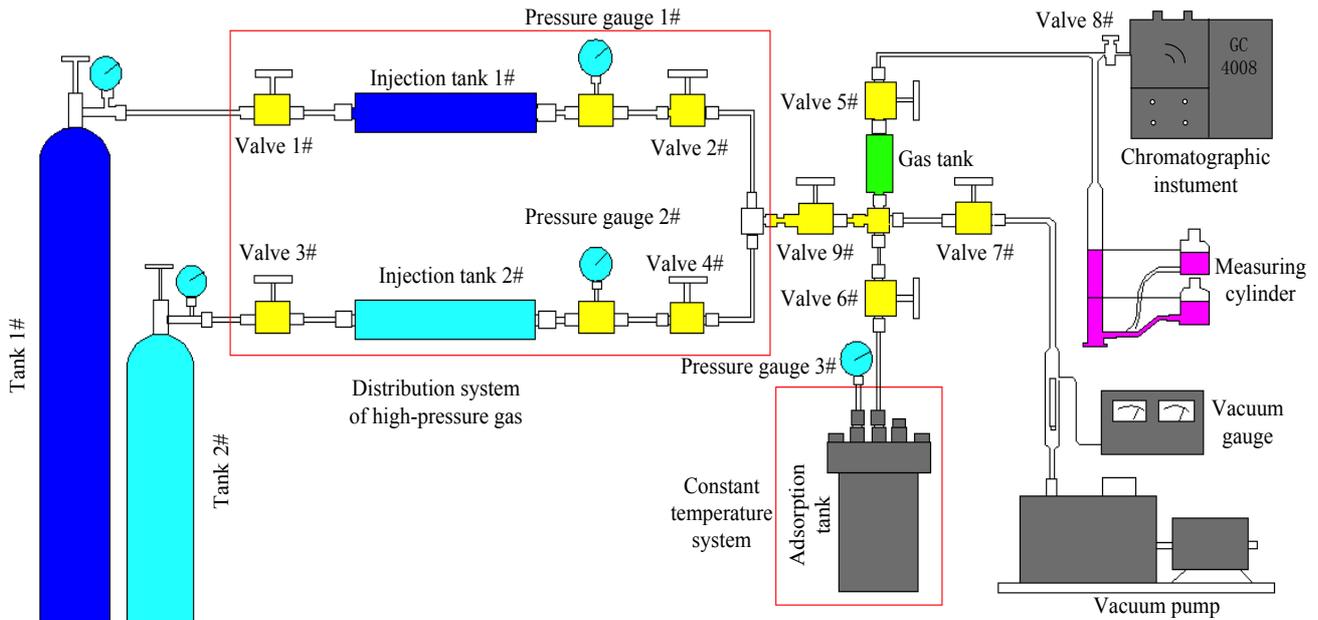


Fig.1. Experimental set-up

3.3 Test procedures

Nitrogen was injected into the coal bed after the adsorption and desorption of the methane reached an equilibrium at a system temperature of 23 °C.

Specific test procedures were as follows. (1) Assuming gas tightness, the methane (concentration 99.99%) in Tank 1 was injected into the coal specimens in the adsorption tank. After allowing 7 hours for the adsorption and desorption equilibrium to be achieved, nitrogen in Tank 2 was injected into the adsorption tank, at a pressure a little higher than the methane. The pressure and the injected gas volume in the adsorption tank were recorded after 7 hours of adsorption/desorption equilibrium. (2) To keep the pressure in the adsorption tank free from external influences, the mixed free gases at equilibrium pressure were isolated by the preserving tank. A gas chromatograph was used to analyze and record the concentrations of the released methane and nitrogen. (3) An adsorption/desorption equilibrium at higher pressure was obtained by continuous nitrogen injection from Tank 2. The concentrations of the freed methane and nitrogen were measured as in step 2. After several repetitions of the above, the data for the gases composed of five elements when the pressure of methane was in equilibrium were recorded and plotted. (4) When the adsorption/desorption equilibrium reached 0.3 MPa, 0.4 MPa and 0.5 MPa, nitrogen was injected and the corresponding data recorded, and then the entire curve of substitution and adsorption was plotted for different pressures.

3.4 Analysis of tests results and the substitution law

Nitrogen at a pressure lower than 0.74 MPa was injected into the three specimens when the adsorption equilibrium was reached at 0.3 MPa, 0.4 MPa and 0.5 MPa. The substitution volumes at five different equilibrium pressures for these three types of specimens were calculated, and the results are shown in Figs 2, 3, 4 and 5.

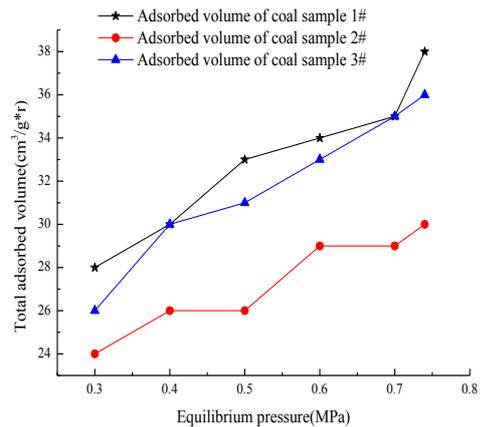


Fig. 2. Total gas adsorption at different equilibrium pressure conditions for the three coal samples

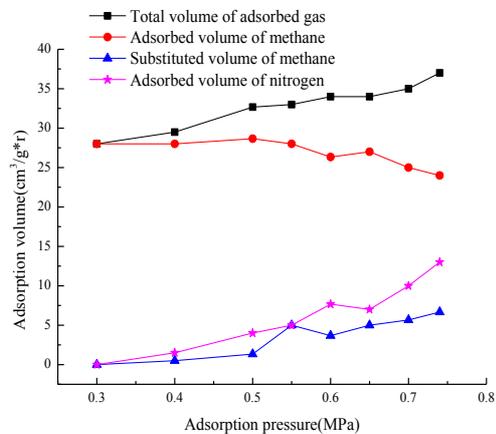


Fig. 3. Volume of gas adsorption and substitution of coal sample #1 at different pressures

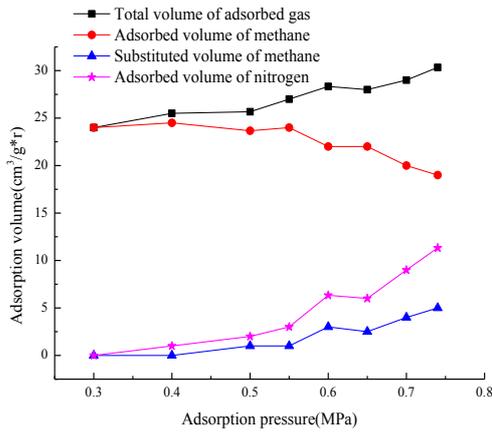


Fig. 4. Volume of gas adsorption and substitution of coal sample #2 at different pressures

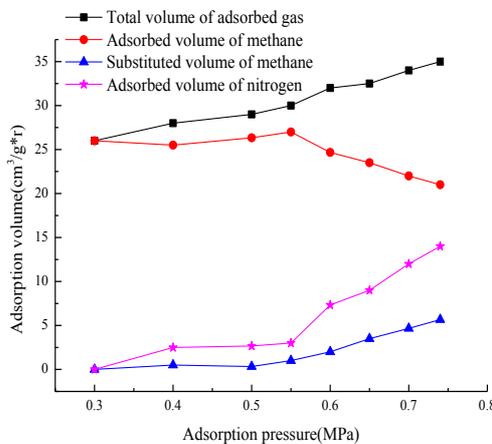


Fig. 5. Volume of gas adsorption and substitution of coal sample #3 at different pressures

It can be concluded from Fig. 2 that the gas adsorption volume increases as the pressure increases, and this tendency is in accordance with the isothermal adsorption law. The following results are obtained from Figs 3–5.

(1) It becomes a little more difficult to substitute nitrogen for methane as the pressure of the methane adsorbed in coal increases. For example, the substitution rate of methane in the three groups of coals is 19.31% when the equilibrium pressure of methane is 0.3 MPa; when the equilibrium pressure increases to 0.4 MPa, the substitution rate decreases to 18.63%; and when the pressure increases to 0.5 MPa, the substitution rate decreases to 17.64%.

(2) It can be seen from tests that the substitution volume of methane increases when the total pressure of the gas is higher than 0.5 MPa. Thus, the efficiency of injecting inert gas to eliminate the outburst of methane is high when the residual gas pressure is 0.5 MPa, and it will be improved further when the injection pressure is greater than 0.5 MPa. Overall, the substitution volume has a linear relationship with the pressure difference between the injected inert gas and the original gas; that is, the substitution volume increases as the pressure of the injected inert gas increases. Based on the relationships between the pressure of the injected inert gas, the difference between the pressure of adsorption equilibrium (ΔP) and the substitution volume (ΔV), a linear equation can be written as:

$$\Delta V = 9.4523 \Delta P + 1.2875 \quad (1)$$

where ΔV is the substitution volume ($\text{cm}^3/\text{g.r.}$) and ΔP is the pressure difference between the pressure of the injected inert gas and the original gas (MPa).

As the amount of data increases, so the optimum injection pressure can be obtained for more and more adsorption pressures. This conclusion can provide a theoretical reference for eliminating the outburst of methane by injecting inert gas.

(3) Because of the unsaturated state of methane at low-pressure adsorption equilibrium, the vacant adsorption locations will be the first to be occupied by the injected nitrogen, and then the pressure difference between the methane and the nitrogen will force the nitrogen to replace the methane at the occupied locations. Although the coal specimens are not saturated, the substitution of methane proceeds. Thus, the substitution volume is rarely affected by the adsorption limit when the adsorption pressure equilibrium is the same in meta-anthracite. Above all, this injection and substitution method can be used to decrease the gas content when the gas pressure is low. While it is clear that nitrogen's capacity to be adsorbed is poorer than that of methane, the injection of nitrogen will nevertheless lower the pressure of methane, which will then be partially desorbed, and the desorbed locations will be occupied by nitrogen. The substitution of methane is achieved when the dynamic equilibrium between nitrogen and methane is reached.

4. Application in a coal mine

The field tests were conducted at #3 coal bed in the Fu Yanshan mine, which averages 4.96 m in thickness. The immediate roof, false roof and seam floor are, respectively, silty mudstone, carbon mudstone and silty mudstone. The test field was located in the return airway of the 3210 workface, and the residual gas content was $10.2 \text{ m}^3/\text{t}$, with an average extraction concentration of 5.8%. The distance between the extraction boreholes was 3 m, and the injection boreholes were displaced at the middle of the extraction boreholes. The injection pressure of the nitrogen (79.8% concentration) was 0.5 MPa. The concentration and the extraction volume of the gas were monitored successively, and the results are shown in Fig. 6.

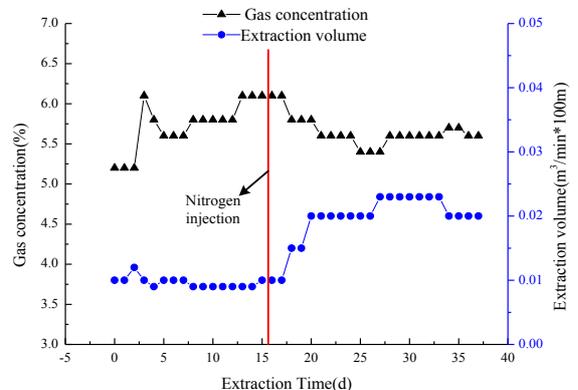


Fig.6. Gas concentration and extraction volume before and after gas injection

It can be concluded from the field tests that the average extraction concentration of gas before injection was 5.8%,

decreasing slightly to 5.6%. The average extraction volume per 100 m increased significantly from 0.01 m³/min to 0.02 m³/min.

The above results confirm the improvement in the extraction of methane by injecting nitrogen. Furthermore, it was shown that the improved level of extraction in the field tests was greater than in the laboratory tests. This increased improvement in efficiency may result from the fact that the adsorption process is located in a confined space in the laboratory tests, and methane is only substituted by nitrogen, whereas in the field tests, the methane was not only substituted but also dispersed by the nitrogen.

5. Theoretical analysis of the injection of nitrogen and the substitution of methane

Based on the experimental results and the corresponding theoretical studies conducted by previous researchers, the current authors propose that the methane, which is not saturated in the meta-anthracite, can be substituted by the injected nitrogen, even though the nitrogen has a lower adsorption capacity than methane. Corresponding analysis can be conducted on the following two aspects.

1) The total gas pressure, which is increased by the injection of nitrogen, will result in a decrease of the methane pressure while the total volume of the methane is stable. According to the extended Langmuir equation, to meet the requirement of pressure equilibrium, methane will be desorbed from the coal.

Furthermore, the adsorbed volume of methane will be decreased by the injection of external gas, even if the pressure of the methane is stable. This can be concluded from Eq. (2), in which $a_1b_1p_1$ is a constant when the pressure of methane (p_1), is stable and the pressure of the injected N₂ (p_2), is higher than zero. However, the increase of the denominator, $1+b_1p_1+b_2p_2$, will result in a decrease of methane adsorption:

$$V_1 = \frac{a_1b_1p_1}{1+b_1p_1+b_2p_2} \quad (2)$$

Where V_1 is the adsorption volume of methane, a_1 and b_1 are adsorption constants, b_2 is the adsorption constant of nitrogen, and p_1 and p_2 are the pressures of methane and nitrogen, respectively.

2) Based on the analysis on molecular kinematics, a dynamic equilibrium is reached between adsorption and desorption. Generally, the degree of adsorption saturation can be written as Eq. (3) when the methane is not saturated:

$$C = \frac{V}{a} = \frac{bp}{1+bp} \quad (3)$$

Where C is the degree of adsorption saturation, V is the adsorbed volume, P is the adsorption pressure, and a and b are adsorption constants.

When methane is not at saturation, some adsorption locations exist in the coal, which are not occupied by methane. Desorption of methane at the occupied locations and the adsorption of methane at adsorption locations that are not occupied will result in the transfer of methane among the adsorption locations that are not occupied. With the injection of nitrogen, some of these adsorption locations will be occupied, and the movement of the methane between the adsorption locations that are not occupied will be restrained, and the methane will be replaced, even though the adsorption capacity of methane is higher than that of nitrogen. All of the adsorption locations will not be completely occupied by nitrogen straight away, but the nitrogen will decrease the adsorption possibilities for methane, until finally its substitution is achieved.

6. Conclusions

To enhance underground coal mine methane extraction efficiency in meta-anthracite seams with low gas pressure, a new approach, methane displacement by gas injection, has been studied, first in the laboratory and then in a field trial in the Fuyanshan coal mine. The following preliminary results have been obtained.

1) It is feasible to inject nitrogen to displace residual methane in coal seams, and this method improves the extraction rate at low pressure.

2) The injection of nitrogen weakens the concentration and pressure of methane. The methane is desorbed partially, and the desorbed locations are occupied by nitrogen. When the dynamic equilibrium between nitrogen and methane is reached, the displacement of methane is achieved.

3) It is difficult to displace methane by nitrogen injection when the pressure of methane in the coal is high. The displacement volume is linearly related to the difference between the pressure of the injected inert gas and the pressure of the original gas. Thus, it is advantageous to increase the injection pressure to increase the extraction volume when the residual pressure of gas is reduced.

4) The displacement effect of nitrogen in the field tests gave greater efficiency in gas extraction when compared to the laboratory tests, which may be attributable to additional gas flow interaction.

5) Methane displacement by gas injection depends not only on the gas adsorption property, but also on the partial pressure of each gas.

Acknowledgements

This work was funded by projects 5097056 and 51174081, supported by the National Natural Science Foundation of China, and project 201304, supported by the Open Research Fund Program of Hunan Province Key Laboratory of Safe Mining Techniques of Coal Mines in Hunan University of Science and Technology.

References

1. X.F. WANG, D.S. ZHANG, G.J. LI, X.D. WANG, "Boreholes layout of coal mine methane drainage for high gassy and low permeability coal seams in Tiefa coal-field". *Journal of China Coal Society*, 36(8), 2011, pp. 1296–1330
2. Z.J. HAN, S.X. SANG, Z.Z. CHENG, H.Z. HUANG, "Exploitation technology of pressure relief coalbed methane in vertical surface wells in the Huainan coal mining area". *Mining Science and Technology*, 19(1), 2009, pp. 25–30.

3. L.R. WU, Z.G. JINANG, W.M. CHENG, X.W. ZUO, D.W. LV, Y.J. YAO, "Major accident analysis and prevention of coal mines in China from the year of 1949 to 2009". *Mining Science and Technology*, 21(5), 2011, pp. 693–699.
4. Mjewska Z., Zietek J, "Acoustic emission and volumetric strain induced in coal by the displacement sorption of methane and carbon dioxide". *Acta Geophysica*, 56(2), 2008, pp. 372–390.
5. Jessen K, Guo-Qing T, Kovscek AR, "Laboratory and simulation investigation of enhanced coalbed methane recovery by gas injection". *Transport in Porous Media*, 73(2), 2007, pp. 141–159.
6. Pini R, Storti G, Mazzotti M. "A model for enhanced coal bed methane recovery aimed at carbon dioxide storage". *Adsorption*, 17(5), 2011, pp. 889–900.
7. Clarkson C R, Bustin R M. "Binary gas adsorption /desorption isotherms: effect of moisture and coal composition upon component selectivity". *International Journal of Coal Geology*, 42(4), 2000, pp. 241–272.
8. Éttinger I L. "Methane saturation of coal strata as methane–coal solid solution". *Soviet Mining*, 26(2), 1990, pp. 159–164.
9. Yavitt J B. "Methane and carbon dioxide dynamics in Typha Latifolia (L.) wetlands in central New York state". *Wetlands*, 17(3), 1997, pp. 394–406.
10. Kuilenya M V, Serdyukov S V. "Methane desorption and migration in thermodynamic in equilibrium coal beds". *Journal of Mining Science*, 46(1), 2010, pp. 50–56.
11. Papp H, Schuler P, Zhuang Q. "CO₂ reforming and partial oxidation of methane". *Topics in Catalysis*, 3(3-4), 1996, pp. 299–311.
12. H.G. YU. "Study of Characteristics and Prediction of CH₄, CO₂, N₂ and Binary Gas Adsorption On Coals and CO₂/CH₄ Replacement" PhD thesis of Shandong University of Science and Technology, Qingdao, China. 2005.