ORIGINAL PAPER - PRODUCTION ENGINEERING

Sorption capacity of Indian coal and its variation with rank parameters

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Abstract

The study of gas sorption characteristics is important for practical assessment of coal bed methane (CBM) production and CO_2 sequestration in coal seam. Adsorption isotherm is one of the critical parameters for the establishment of production as well as injection well. Adsorption isotherm provides information about the reservoir conditions and critical desorption pressure as well as volume of gas that can be sequestered in deep coal seam. Alteration in sorption isotherm reflects the increase or decrease of the gas production as well as CO_2 sequestration. Therefore, in this paper, experimental investigation was carried out to determine the CO_2/CH_4 sorption capacity of five different coal samples taken from different locations of Jharia coalfield (Moonidih area) of Gondwana basin. Gas sorption capacity was determined at 27 °C temperature and up to 7.5 MPa pressure. CO_2 adsorption was observed to be higher than that of the CH_4. The sorption ratio of CO_2/CH_4 varied from 1.6:1 to 1.2:1 for all coal samples. Furthermore, the experimental results were correlated using established Langmuir, Freundlich, Temkin and D–R isotherm models. Experimentally obtained values satisfactorily fitted to the Langmuir and Freundlich model with comparable accuracy. The excess adsorption capacity of coal was also compared with different rank parameters to understand the variation of sorption capacity with rank of coal.

Keywords Coal bed methane · Proximate and ultimate analyses · CO_2/CH_4 sorption capacity · Statistical analysis

Introduction

At present, climate change and global warming are the most debated topics. CO_2 is one of the dominant causes of greenhouse gas effect (GHG) and global warming due to its higher concentration in the atmosphere. Thermal generation capacity of India will be 290 GW in 2047, out of which 253 GW will be coal based and 37 GW gas based (1). Increasing demand of energy also increases CO_2 emission. Concentration of CO_2 in the atmosphere was found to be 396 ppmv in 2013 which was 40% higher than that in mid-1800s (2). The rate of emission was 2 ppmv/year for the last 10 years (2) alarming the world for advancement towards technology for the reduction of CO_2 concentration in atmosphere. The CO_2 concentration can be controlled by two ways: one to reduce CO_2 emissions into the atmosphere and the other to reduce

the present concentration of CO_2 in the atmosphere to safe levels through application of various engineering principles. The second way is possible by CO_2 storage in un-minable coal seams, depleted oil and gas reservoirs, abandoned and sealed mines, saline aquifers, oceans, etc. (Ishaq et al. 2009; Shi and Durucan 2005; Robertson 2010). Sequestration of CO_2 in deep coal seam is one of the techniques to reduce CO_2 concentration in the atmosphere and mitigation of global warming. Coal bed reservoir has the ability to store and retain large quantity of CO_2 gas for very long time due to its porous nature (Shi and Durucan 2005; Robertson 2010). Injection of CO_2 in coal seam also facilitates beneficial effect on enhanced coal bed methane recovery (ECBM) (Stevens et al. 1998; Seidle 2000; Pagnier et al. 2005). Estimation of CO_2/CH_4 sorption in deep coal seam requires critical experimental investigation (Hernandez et al. 2006; Vishal et al. 2015; Buscha and Gensterblumb 2011). Investigative effort has been undertaken elsewhere to estimate excess sorption capacity at different temperature and increasing pressure values (13; Prusty 2008; Chen et al. 2011). CO_2/CH_4 sorption ratio was reported to be 10:1 for low-rank coal and less than 2:1 for low- and medium-volatile bituminous coals (Deng et al. 2015). Variation in gas sorption capacity of coal based

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on proximate analysis, ultimate analysis and maceral contents has been reported elsewhere (Saikia and Sarkar 2013; Ghosh et al. 2014). The sorption isotherm gives estimated retention capacity of gas at in situ temperature and pressure of reservoir. Detailed sorption isotherm of CO_2 and CH_4 is required as input for production simulation in CBM and ECBM operations (Ghosh et al. 2014). The investigation suggests the variation of rate of sorption depends upon the pore size and porosity of the coal, molecular size of the gas and critical temperature and pressure (19; Suman and Harpalani 2019; Ali et al. 2018).

In this investigation, the sorption capacity of CO_2 and methane on five different coal samples (namely JH-MD-S_1, JH-MD-S_2, JH-MD-S_3, JH-MD-S_4 and JH-MD-S_5) from Jharia coalfield (Moonidih area) was carried out experimentally. Excess CO_2/CH_4 sorption capacity was obtained at **27 °C** with incremental gas pressure from 0 to 7.5 MPa at varying depth from 400 to 580 m. Sorption isotherms were plotted using experimentally obtained values. The experimentally obtained data were compared with four different established isotherm models, namely (Langmuir, Freundlich, Temkin and D–R isotherm models) to obtain the best-fit isotherm model for the study area. The rank parameters such as moisture content, ash content, volatile matter (daf basis), fixed carbon (daf basis) and vitrinite reflectance were correlated with CO_2 and CH_4 excess sorption amount to determine the effect of these parameters on sorption amount of coal.

Geological setting of Jharia coal bed basin

Coal sample for the sorption study was collected from the underground mine of Jharia coal bed basin (Moonidih Area) (Fig. 1) which is located in Dhanbad district of Jharkhand, India. It is bounded between 23°37′N and 23°52′N and longitudes 86°05′E and 86°30′E. Mostly these mines are full of gassy environment with elevated strata temperature (Daniel and Bustin 2007) (Guo 2003).

Sample preparation and experimentation

The sorption study was carried out on coal samples obtained from freshly exposed coal surface at depth varying from 400 to 580 m. The lump of samples was crushed and passed through a sieve of size 72 BSS mesh (212 μm). The moisture content of the samples was estimated as per standard [ASTM D 3173–11]. Proximate analysis of these samples was estimated as per the standard method [ASTMD 3172–07a] and ultimate analysis using CHNS Analyzer [Make Evisa Vario EL III- CHNS analyzer] as per the standard methods [ASTM D 3176–09].
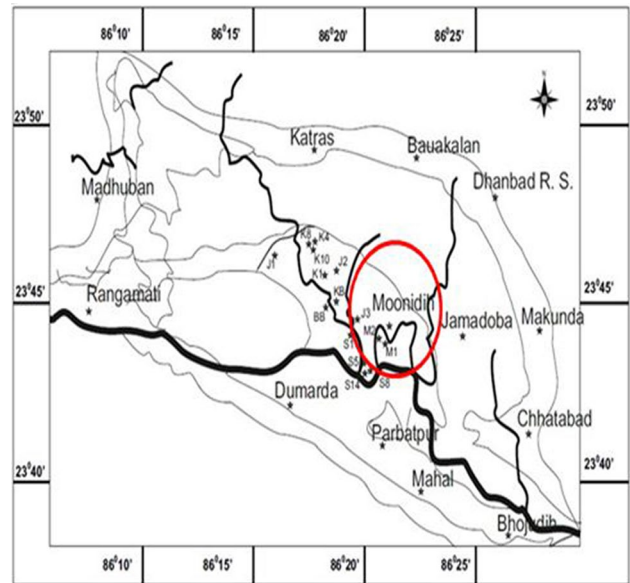
Fig. 1 Geological setting of Jharia coalfield (Moonidih area)

Volumetric sorption setup was used to estimate the sorption capacity of coal. The setup consists of reference cell, sample cell, flow line, digital pressure transducer, data acquisition system, etc. (Figs. 2, 3). The volumetric setup was kept immersed in water bath maintained at uniform temperature of 27 °C (± 1 °C). The volume of excess sorption was determined at constant temperatures as a function of pressure at equilibrium.

Determination of sorption capacity

About 100 g of coal sample was taken in the calibrated sample cell. The setup was evacuated to remove unwanted gas present in coal using vacuum pump (make: Scientific Instrument Services, Inc., USA). Helium gas was injected at 0.68–3.5 MPa pressure to determine the volume of sample as well as reference cell using volume expansion method as follows (Zhang et al. 2013):

$$V_s = \frac{\psi}{\psi - \xi} \times V_{sb}, \tag{1}$$

$$\psi = \frac{\frac{P_{s_2}}{Z_{s_2}} - \frac{P_{s_1}}{Z_{s_1}}}{\frac{P_{r_1}}{Z_{r_1}} - \frac{P_{r_2}}{Z_{r_2}}}, \tag{2}$$

$$\xi = \frac{\frac{P_{s_2}}{Z_{s_2}} - \frac{P_{s_1}}{Z_{s_1}}}{\frac{P_{r_1}}{Z_{r_1}} - \frac{P_{r_2}}{Z_{r_2}}}, \tag{3}$$
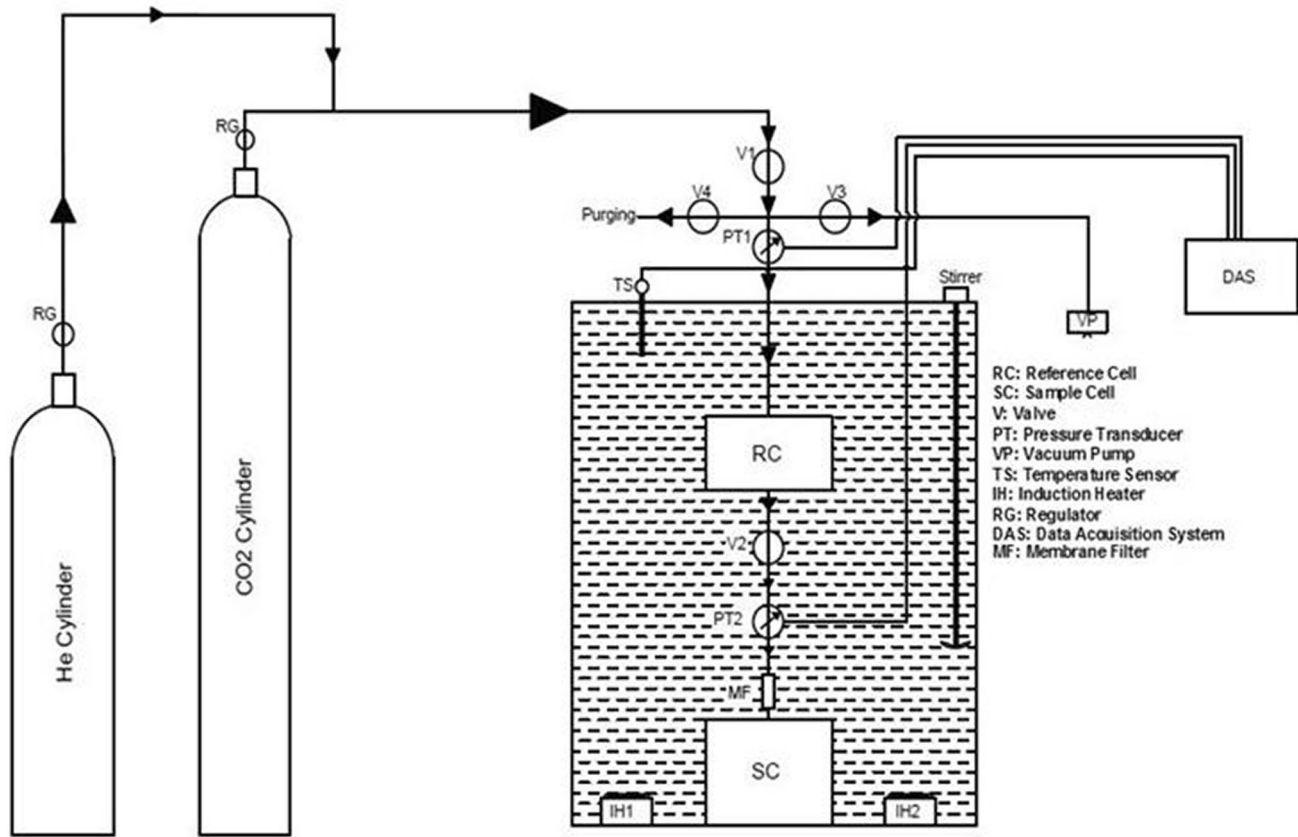
Fig. 2 Schematic diagram of volumetric sorption setup

where V_s is the volume of sample cell, ψ is the constant with iron ball in the sample cell, ξ is the constant without iron ball in sample cell, V_{sb} is the total volume of the iron ball, P_{s2} and P_{s1} are the pressure of helium gas in sample cell before and at the equilibrium condition, P_{r2} and P_{r1} are the pressure of helium gas in reference cell before and at the equilibrium condition, $Z_{s2}, Z_{s1}, Z_{r2}, Z_{r1}$ are the compressibility factor at the respective pressure. Similarly, the volume of reference cell was determined as (Zhang et al. 2013):

$$V_r = \frac{\psi \times \xi}{\psi - \xi} \times V_{sb}. \tag{4}$$

Compressibility factor was calculated using American gas association report no. 8, 1992 (GAS Calc.5.0). Void volume (V_v) was calculated as (Zhang et al. 2013):

$$V_v = \frac{\frac{P_{r_1}}{Z_{r_1}} - \frac{P_{r_2}}{Z_{r_2}}}{\frac{P_{s_2}}{Z_{s_2}} - \frac{P_{s_1}}{Z_{s_1}}} \times V_r. \tag{5}$$

Mole transferred was determined as

$$n_{total} = \frac{P_r V_r}{Z_r RT}, \tag{6}$$

where n_{total} is the mole transferred to the reference cell, R is the gas constant and T is the temperature at which the experiment was conducted (27 °C).

CO_2 gas (99.99% pure) was first filled in the reference cell at regular interval of 0.68 ± 0.068 MPa.

Total mole transferred to the reference cell was determined as (Zhang et al. 2013):

$$n_r = \frac{\frac{P_{r_1}}{Z_{r_1}} - \frac{P_{r_2}}{Z_{r_2}}}{R \times T} \times V_r, \tag{7}$$

$$n_s = \frac{\frac{P_{s_2}}{Z_{s_2}} - \frac{P_{s_1}}{Z_{s_1}}}{R \times T} \times V_s. \tag{8}$$

The sorption of gas was estimated through mole balance. Gas adsorbed was determined from the difference between mole injected and mole present in void:
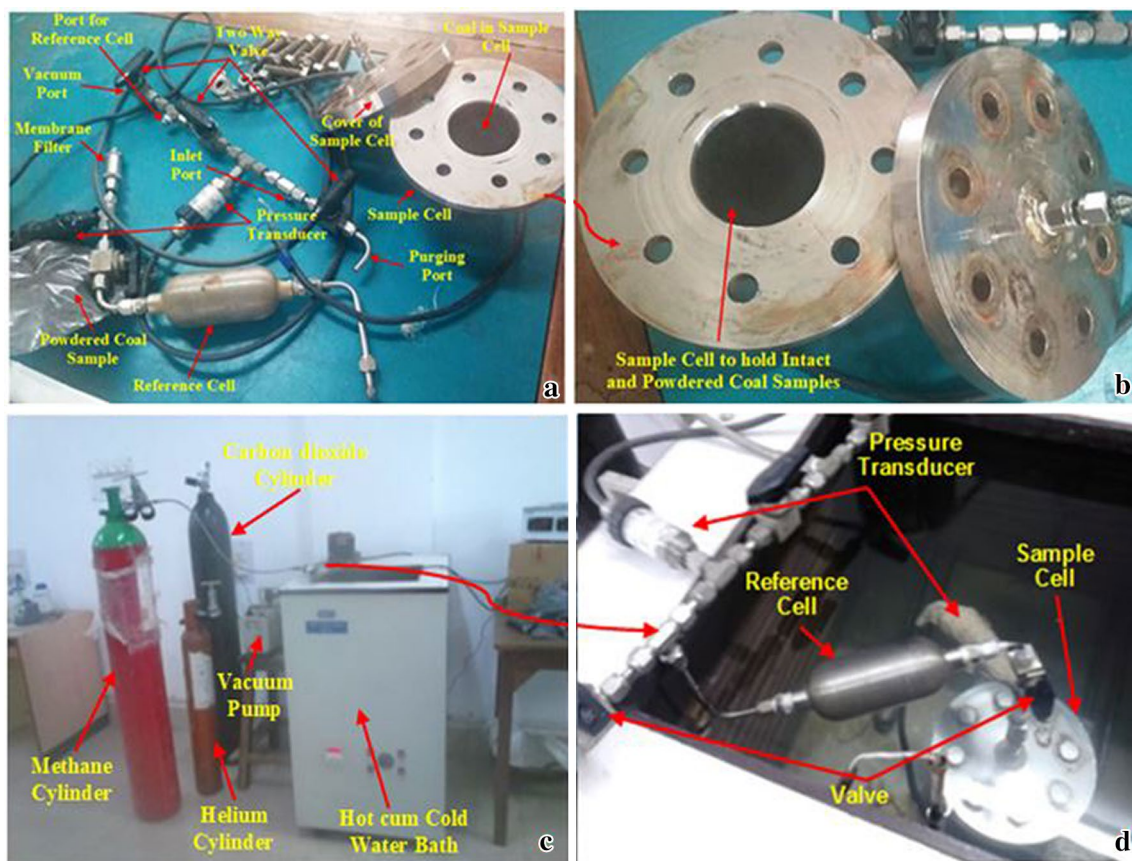
$$n_{adsorbed} = n_{injected} - n_{void}. \tag{9}$$

Fig. 3 Experimental setup for sorption test. **a** Detailed view of the different parts in setup. **b** Sample cell to hold intact as well as powdered coal samples. **c** Setup with gas cylinder as well as vacuum pump. **d** Setup in place

Results and discussion

The results of proximate and ultimate analyses are shown in Table 1. VM (daf basis) varies between 21.02 and 34.48, while FC (daf basis) found in between 65.52 and 78.98 indicates medium-volatile bituminous coal. Variation in moisture and ash content from 0.93 to 1.4 and 9.01 to 20.64

reflects medium-volatile bituminous coal. Value of vitrinite reflectance was determined using the established Rice formula (Rice 1993) as.

$$R_0(\%) = -2.712 \times \log\left(VM_{daf}\right) + 5.092, \tag{20}$$

where R_0 is the vitrinite reflectance (%) and $VM_{(daf)}$ is the volatile matter (dry ash-free basis) (%).

Table 1 Results of proximate analysis of coal samples

Parameters	Sample ID				
	JH-MD-S$_1$	JH-MD-S$_2$	JH-MD-S$_3$	JH-MD-S$_4$	JH-MD-S$_5$
Depth (m)	580	520	500	450	400
M (wt%)	0.93	1.19	1.27	1.28	1.40
A (wt%)	9.01	13.46	15.38	18.49	20.64
VM (wt%)	18.93	22.53	24.81	25.28	26.88
FC (wt%)	71.13	62.82	58.54	54.95	51.08
VM (d) (wt%)	19.11	22.80	25.13	25.61	27.26
FC (d) (wt%)	71.80	63.58	59.29	55.66	51.81
VM (daf) (wt%)	21.02	26.40	29.77	31.51	34.48
FC (daf) (wt%)	78.98	73.60	70.23	68.49	65.52
R0 (%)	1.51	1.24	1.10	1.03	0.92

Table 2 Sorption capacity of coal

Samples	Depth (m)	Excess sorption amount (cc/g)	
		CO_2	CH_4
JH-MD-S$_1$	580	20.52	13.29
JH-MD-S$_2$	520	16.54	12.29
JH-MD-S$_3$	500	14.26	11.23
JH-MD-S$_4$	450	10.02	9.99
JH-MD-S$_5$	400	11.11	9.49
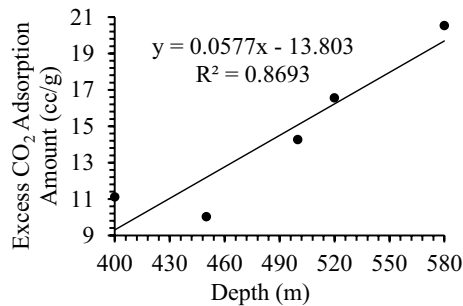
Fig. 4 Variation of excess CO_2 sorption with depth

$y = 0.0577x - 13.803$
$R^2 = 0.8693$
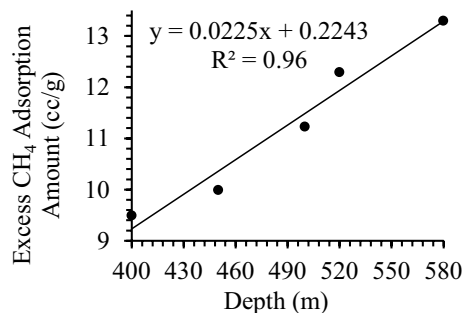
Fig. 5 Variation of excess CH_4 sorption with depth

$y = 0.0225x + 0.2243$
$R^2 = 0.96$

The variation of R_0 between 0.92 and 1.51 indicates medium-volatile bituminous coal as per the rank parameter discussed elsewhere (Diessel 1992) (Table 1). The values of vitrinite reflectance observed in this study lie in between the threshold value of 0.7–2.0% which confirms commercial CBM prospects as described elsewhere (Chandra 1950).

Sorption capacity of coal bed

Sorption capacity was measured at the highest pressure of 7.5 MPa for methane and 5.5 MPa for CO_2 at 300 K. CO_2 and CH_4 sorption capacity varied from 11.11 to 20.52 and 9.49 to 13.29 cc/g, respectively (Table 2). Sharp increase in sorption capacity was observed with increase in depth of coal seam (Figs. 4, 5). Increase in sorption capacity with depth is due to the increase in carbon content as well as porosity of coal and reduction in moisture content. The sorption ratio of CO_2/CH_4 was from 1.6:1 to 1.2:1. Adsorption of CO_2 was observed to be higher than that of CH_4 with an increase in pressure. The difference between adsorption amounts is due to the higher density of CO_2 and its interaction with coal than that with CH_4. It is also due to the variation in molecular diameter of CO_2 (0.33 nm) than CH_4 (0.38 nm). This increase in maturity and higher rank of coal allow more sorption. CO_2 sorption capacity of coal of study area was also compared with different coalfields worldwide (Table 3).

Determination of sorption isotherm based on experimental data

The sorption capacity of CO_2 and CH_4 in coal was studied and sorption isotherm was plotted (Figs. 6, 7). Experimental values were compared with other isotherm models such as D–R, Temkin, Freundlich and Langmuir, respectively, using linear regression method. Values were analysed statistically to determine the regression coefficient of all isotherm

Table 3 Sorption capacity of coal at different coalfields worldwide

Location	Depth (m)	Temp. (°C)	Pressure (bar)	Excess sorption amount (cc/g)	References
Monte Sinni coal mine (Carbosulcis, Cagliari, Italy)	500	45	190	45.302	(Mares et al. 2009)
Huntly Coalfield, New Zealand	520	32	40	23.718	(Gruszkiewicz et al. 2009)
Black Warrior Basin, West Central Alabama	731	35	69	17.968	(Saghaf 2010)
Australian coals	750	27	51	24.998	(31)
Great Britain, Slovenia and Poland	–	45	69	39.754	(Garnier et al. 2011)
Kyungdong coal (South Korea)	756	65	101	27.401	(33)
		45	76	33.016	
Waterberg Coalfield, South Africa	250	30	45	44.471	(Maphala 2012)
		30	42	20.910	
Jharia Coalfield, India	580	27	55	20.52	Study area

Fig. 6 a Comparison of CO_2 excess sorption amount of different coals at 27 °C. **b** Comparison of different CO_2 sorption isotherm models at 580 m depth and 27 °C
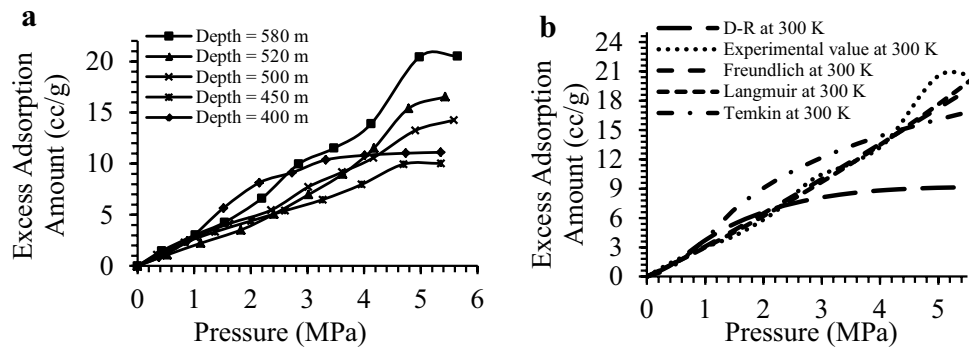

Fig. 7 a Comparison of CH_4 excess sorption amount of different coals at 27 °C. **b** Comparison of different CH_4 sorption isotherm models at 580 m depth and 27 °C
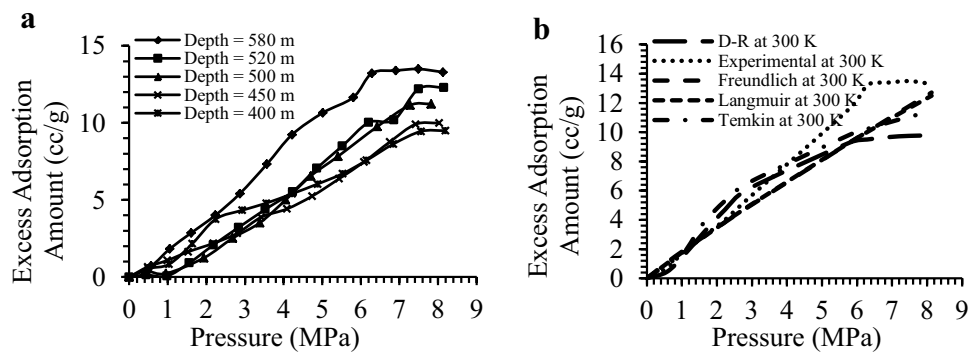

Table 4 Constant of CO_2 sorption isotherm model at 27 °C

Depth (m)	Freundlich isotherm		Langmuir isotherm	
	$1/n$	K_f	q_m	K
580	0.9693	0.0151	76.9230	0.000257
520	1.2061	0.0049	21.0970	0.000565
500	0.9693	0.0208	30.3951	0.000669
450	0.818	0.0438	28.9017	0.000647
400	0.5153	0.0208	14.7928	0.004244

Table 5 Constant of CH_4 sorption isotherm model at 27 °C

Depth (m)	Freundlich isotherm		Langmuir isotherm	
	$1/n$	K_f	q_m	K
580	0.9260	0.0181	83.3333	0.000149
520	1.3753	0.0008	16.8067	0.000390
500	1.8539	0.00003	40.9836	0.000340
450	1.1074	0.0040	22.3713	0.000373
400	0.8555	0.0228	30.2114	0.000382

models. Freundlich and Langmuir models were observed to be best fitted to the experimental values. This is due to the monolayer as well as multilayer sorption of gas in coal obtained from different depths of occurrence.

The constant for each model is shown in Tables 4 and 5. Constant, q_m obtained from Langmuir isotherm model indicates monolayer sorption process. Values of $1/n$ and K_f were determined from Freundlich isotherm model. The values of $1/n$ were found below 1 and above 1 indicating normal as well as cooperative sorption process. Thus, the monolayer as well as multilayer deposition of gas was observed in gas sorption process in coal. Temkin isotherm and D–R isotherm models were less fitted to the experimental values; therefore, the constants associated with the Temkin and D–R isotherms were not discussed in the results. The fitting of the Langmuir as well as Freundlich isotherm models to the experimental values is due to well-developed pores, pore size distribution and porosity in coal samples.

Langmuir volume and pressure

Langmuir volume and pressure are widely acceptable parameters in reservoir engineering. In a physical sense, the Langmuir volume constant (V_L) represents the maximum amount of gas that can be sorbed onto the given sample at infinite pressure. When pressure equals the Langmuir pressure constant (P_L), gas content is equal to half of V_L. Thus, the Langmuir pressure constant represents the half-saturation pressure, i.e. the pressure at which the coal holds one-half the maximum gas volume. Langmuir pressure and volume were determined for all coal samples as shown in

Table 6. Langmuir volumes of CO_2 and CH_4 sorption were 11.11–20.53 and 9.5–13.29 cc/g at pressures 1.52–2.9 and 3.59–5.32 MPa, respectively, at 27 °C. The Langmuir pressure and volume are important to determine the saturation level as well as critical desorption pressure in initial reservoir conditions.

Effect of moisture content on sorption capacity of coal

The moisture content of coal at varying depths was correlated with CO_2 as well as CH_4 sorption capacity and an increasing trend of gas sorption capacity with a decrease in moisture content was observed. Linear correlation with R^2 of 0.81 and 0.84 was observed between sorption capacity and moisture content reflecting a strong influence of moisture content on gas sorption capacity of coal (Fig. 8a, b).

Increase in gas sorption capacity with loss of moisture content is due to increases in porosity and gas sorption site in coal. The moisture in coal occupies more sorption sites and restricts the sorption of gas. Presence of moisture in coal is also responsible for matrix shrinkage and decrease in gas sorption capacity. The reduction in moisture content in coal facilitates more sorption site by producing more micro-pores and porosity in coal matrix.

Effect of ash content on sorption capacity of coal

Results obtained in experimentation reflect the reduced sorption capacity of both CO_2 as well as CH_4 with enrichment of ash content in coal. Linear correlation with R^2 of 0.93 and 0.97 was obtained between excess sorption amount and ash content reflecting the strong correlations between them (Fig. 9a, b). Decrease in gas sorption capacity of coal with

Table 6 Langmuir volume and pressure of coal samples at 27 °C

Depth (m)	CO₂ sorption			CH₄ sorption		
	P_L (MPa)	V_L (cc/g)	R^2	P_L (MPa)	V_L (cc/g)	R^2
580	2.90	20.53	0.98	5.32	13.29	0.99
520	3.45	16.55	0.99	4.59	12.3	0.99
500	3.31	14.26	0.98	4.34	11.23	0.90
450	2.38	10.02	0.99	4.22	8.97	0.99
400	1.52	11.11	0.99	3.59	9.5	0.99

Fig. 8 Effect of moisture content on sorption capacity of coal. **a** CO_2 sorption and **b** CH_4 sorption
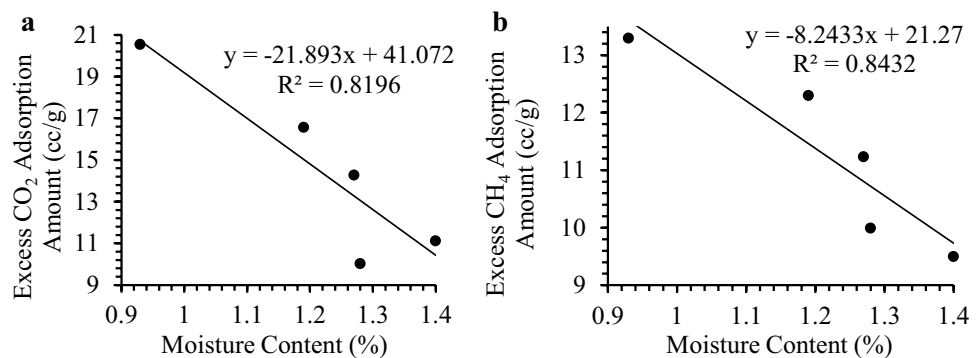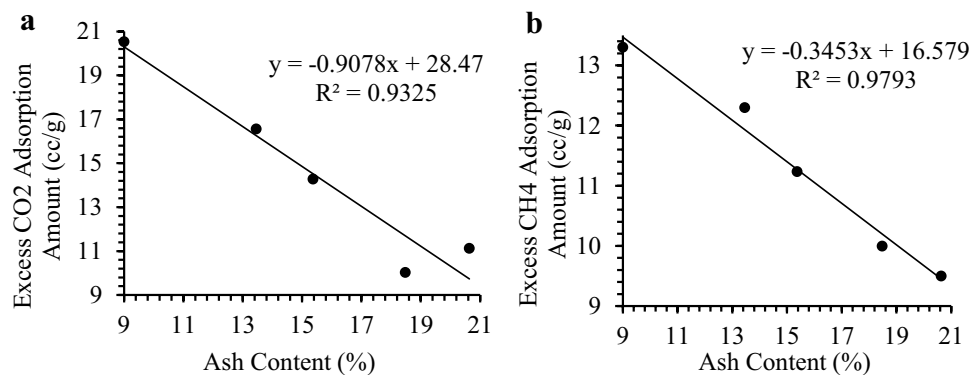

Fig. 9 Effect of ash content on sorption capacity of coal. **a** CO_2 sorption and **b** CH_4 sorption
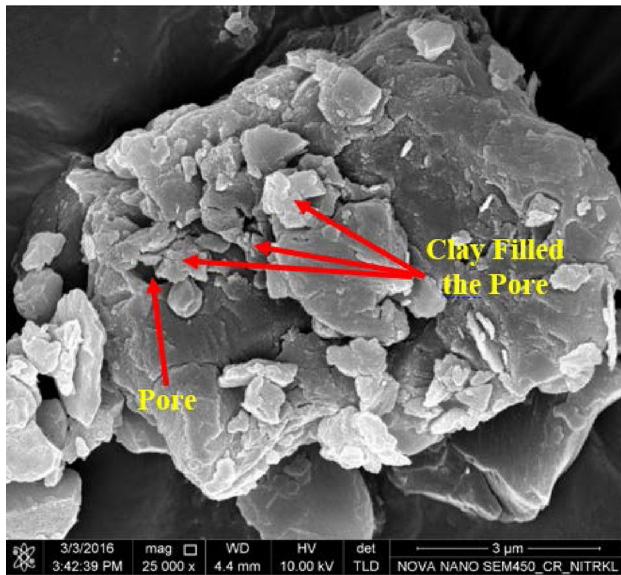
Fig. 10 FESEM image showing infilling of pores in coal sample

ash content is attributed to the infilling and blockage of pores, cleats and fracture system with inherent and extraneous mineral matter such as clays and carbonates in coal. The infilling of the pores is also indicated in the FESEM image shown in Fig. 10.
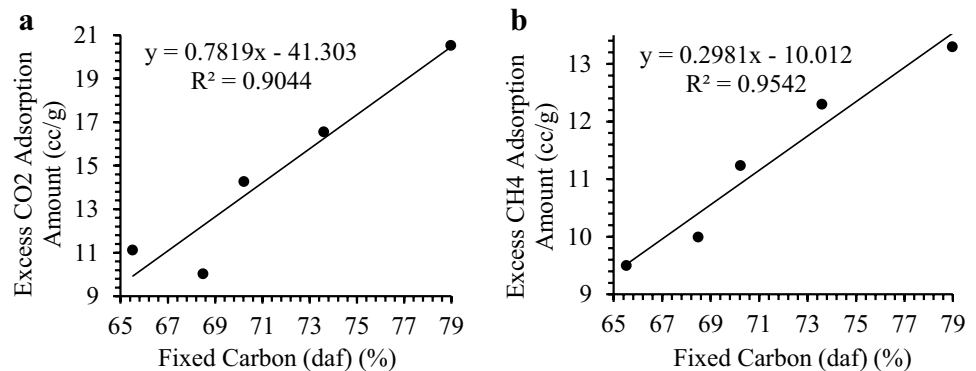
Effect of fixed carbon (daf basis) on sorption capacity of coal

Experimentally obtained sorption isotherm of both CO_2, as well as CH_4, was correlated with fixed carbon (daf basis) of coal samples (Fig. 11a, b). Linear and positive correlation with R^2 0.90 and 0.95 was obtained with CO_2 as well as CH_4 excess sorption capacity, respectively, reflecting strong influence of fixed carbon on sorption capacity of coal. Increased sorption capacity was observed with fixed carbon (daf basis). Increase in fixed carbon develops more micro- as well as meso-pores and facilitates gas sorption site in coal matrix (Ali et al. 2018).

Effect of vitrinite reflectance on sorption capacity of coal

The correlation was established with excess CO_2 as well as CH_4 sorption amount of coal and percentage of calculated vitrinite reflectance of coal (Fig 12a, b). Linear and positive correlation with R^2 0.90 and 0.93 was obtained with CO_2 as well as CH_4 excess sorption capacity, respectively, reflecting strong influence of vitrinite reflectance on sorption capacity of coal. Increased sorption capacity was observed with vitrinite reflectance. It is attributed to increase in porosity, pore size distribution, and specific pore surface area with increase

Fig. 11 Effect of fixed carbon (daf basis) on sorption capacity of coal. **a** CO_2 sorption and **b** CH_4 sorption

Fig. 12 Effect of vitrinite reflectance on sorption capacity of coal. **a** CO_2 sorption and **b** CH_4 sorption

in vitrinite reflectance of coal ([19], Suman and Harpalani 2019; Ali et al. 2018). The vitrinite reflectance value of the coal in study area indicates medium-ranking coal (medium-volatile bituminous coal) which is favourable for meso- and macro-pores. These pores are favourable to sorption, diffusion, and migration of coal bed methane as well as injection of CO_2 in coal matrix.

Thus, from the above investigation, it was observed that the depth of occurrence, molecular size of gases, affinity of gas towards coal, density, porosity, coal rank, etc. are the major controlling factors for the adsorption capacity of the coal.

Conclusion

The following conclusion is drawn from the observation of coal under investigation at Jharia coalfield (Moonidih area).

- Coal of study area was found in between medium and low volatile bituminous rank.
- Maximum sorption capacity of coal was observed to be 20.53 cc/g for CO_2 and 13.29 cc/g for CH_4 at 580 m depth.
- Carbon dioxide sorption capacity of coal was observed to be higher than that of the methane due to the higher density of CO_2 and its interaction with coal than that with CH_4. It is also due to variation in molecular diameter of CO_2 (0.33 nm) than CH_4 (0.38 nm).
- Monolayer as well as multilayer sorption of gas was exhibited by the coal samples at variable depths of occurence.
- Increase in gas sorption capacity was observed with fixed carbon, vitrinite reflectance and decrease in ash content and moisture content, respectively.

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