



Full Length Article

Interactions of nitric oxide with various rank coals: Implications for oxy-coal combustion flue gas sequestration in deep coal seams with enhanced coalbed methane recovery



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HIGHLIGHTS

- Sips model can describe NO adsorption equilibrium on coals.
- Elovich equation can fit NO adsorption kinetics on coals.
- Interactions with NO change the nitrogen speciation compositions of coals.
- Chemisorption is the main interaction mechanism between NO and coals.

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ABSTRACT

Oxy-coal combustion is a promising option for capturing CO₂ emitted from coal-fired power plant. Meanwhile, CO₂ sequestration in deep coal seams with enhanced coal-bed methane (CH₄) recovery (CO₂-ECBM) can store the captured CO₂ in geologic period. Thus, if the oxy-coal combustion flue gas directly sequestered in deep coal seams is successfully implemented, the emissions of greenhouse gas (CO₂) and the main gaseous pollutants (NO_x, SO₂) contained in flue gas will be simultaneously mitigated and CH₄ as a by-product can also be recovered. It is acknowledged that the fluid sequestration in the deep coal seams is mainly attributed to the strong adsorption performance of coal matrix, thus the interactions of NO with various rank coals including adsorption behaviors and the possible interaction mechanism were primarily addressed in this work. Adsorption equilibrium study shows that the Sips isotherm model can well describe NO adsorption behavior on various rank coals, which is probably related to the high heterogeneity of coals. The Elovich equation can fit NO adsorption kinetics on coals successfully, indicating that the chemisorption probably plays a dominant role between NO and coals. Both Fourier Transform Infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS) characterization results further confirm that the interactions with NO can change the nitrogen speciation compositions of all the test coals, and chemisorption is the main interaction mechanism between NO and coals. The chemisorption mechanism between NO and coals will contribute to the steady storage of NO in the target coal seams.

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1. Introduction

Carbon dioxide capture and sequestration (CCS) is of great potential to reduce the main anthropogenic greenhouse gas carbon dioxide (CO₂) emissions into the atmosphere. CCS mainly incorporates

CO₂ capture from large stationary CO₂ sources, transport via pipelines and sequestration into a target location [1]. With considerations of CO₂ capture, pre-combustion, post combustion, chemical looping combustion and oxy-fuel combustion can generate concentrated CO₂ from energy-related sources. Among the options for CO₂ capture, oxy-coal combustion using high purity oxygen (≥95%) can generate concentrated CO₂ source [2]. As illustrated in Fig. S1 (a), Supplementary data, the conventional oxy-coal combustion process requires denitration (DeNO_x) and flue gas desulfuration (FGD). With

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regard to CO₂ sequestration, CO₂ sequestration in deep coal seams with enhanced coalbed methane recovery (CO₂-ECBM) has attracted considerable attention [3,4]. Quantum chemistry calculation shows that the adsorption potential well of CO₂ is deeper than that of coalbed methane, thus the adsorbed CO₂ molecules will replace methane when CO₂ adsorption occurs (Fig. S1 (b), Supplementary data) [5]. This conclusion has been verified in many experimental investigations [6–8]. Therefore, the methane recovery from the target coal seams will compensate for the cost of CO₂ sequestration.

From the aforementioned presentations, it is assumed that if the oxy-coal combustion flue gas without DeNO_x and FGD treatment were successfully injected into the deep coal seams, the emissions of greenhouse gas (CO₂) and the main gaseous pollutants (NO_x, SO₂) contained in flue gas would be simultaneously mitigated due to the substantial adsorption capacity of coal under the reservoir conditions [4]. This proposal may be particularly beneficial to eliminate the capital cost of oxy-coal combustion process. We nominate this novel technology as oxy-coal combustion flue gas sequestration in deep coal seams with enhanced coalbed methane recovery (Fig. S1 (c), Supplementary data).

Previous investigations have shown that the fluid sequestration in the deep coal seams is mainly due to the strong adsorption capability of coal matrix [3,4]. Hitherto, there are numerous publications focused on NO adsorption on various carbon-based materials other than coals. Based on the results of ab initial simulations, Vasylenko et al. proposed that the presence of vacancies on Single-Walled Carbon Nanotubes (SWCNT) could prompt surface reconstruction of SWCNT and thus enhanced its chemisorption performance of NO [9]. Chen et al. found that NO molecules adsorbed on Ordered Mesoporous Carbon (OMC) were mainly via the formation of C—O—N=O, which was formed by the bond of C (O) groups on OMC with NO [10]. Li et al. demonstrated that NO could be kept in a more confined state in micropores of platinum (Pt) supported on Activated Carbon Fiber (Pt-ACF) due to the strong interaction existed between electrodeposited Pt particles and NO [11]. The above literature review indicates that although the interactions of NO with many carbon-based materials have been well addressed, the interactions of NO with coals as a widely spread carbon-based material have not been well elaborated up to now. In addition, it is acknowledged that coal is of higher complexity and heterogeneity in physical structure and chemical structure [12,13]. Thus, the exposure process of coal to NO molecules probably incorporate more complex interactions. As a preliminary research on oxy-coal combustion flue gas sequestration in deep coal seams, the interactions of NO_x with coals were addressed in this work. The related conclusions will not only elaborate the mechanism of oxy-coal combustion flue gas components sequestration in deep coal seams, but also provide preliminary knowledge for oxy-coal combustion flue gas sequestration in deep coal seams with enhanced coalbed methane recovery process.

2. Material and methods

2.1. Samples collection and preparation

Three rank coal samples collected from northwest and north China were selected in this study: HB coal, SM coal and YQ coal. Each coal sample was preserved in a sealed plastics container with inert helium (He) preventing undesired alterations of physico-chemical property due to the atmospheric oxidation effect [14].

The proximate analysis including ash, volatile matter and fixed carbon, the equilibrium moisture content, and the maximum vitrinite reflectance coefficient denoted by $R_{o\ max}$ were determined in our previous work [5]. As shown in Table 1, HB coal and SM coal are of high volatile matter content, which are categorized into

Table 1
Properties of coal samples studied in this work.

Property	Sample		
	HB	SM	YQ
Ash (dried basis), wt%	13.26	10.20	19.42
Volatile matter (dried basis), wt%	35.40	31.47	8.92
Fixed carbon (dried basis), wt%	51.35	58.34	71.68
Equilibrium moisture, wt%	18.18	9.73	5.31
$R_{o\ max}$ %	0.77	0.88	2.62

bituminous coals. Contrarily, YQ coal has the lowest volatile matter content, which is classified as anthracite together with $R_{o\ max}$ of 2.62%. Prior to the exposure of NO_x to coals, each sample was uniformly crushed and sieved to obtain particles with particle size range of 125–150 μm.

2.2. Adsorption and desorption test

It is known that NO_x emitted by coal-fired air or oxy-fuel combustion comprises NO and NO₂, and NO accounts for around more than 90% of the total NO_x emissions [15]. Therefore, NO with purity above 99.99% was selected as the representative to study the interactions of NO_x with coals. Many studies have shown that the adsorption effect is the dominant mechanism for gas sequestration in deep coal seams [4,5,16]. Thus, this investigation about the interactions of NO with coals primarily focused on the adsorption behavior of NO on coals. It is necessary to mention that the process of oxy-coal combustion flue gas sequestration in the deep coal seams aims to store fluid in coal seams in geologic time. Thus, the sequestration process is totally different from the adsorption process operated in a fixed bed. Thus, the volumetric method is widely adopted for high-pressure fluid adsorption on coals [17,18]. In this work, the images of schematic diagram of volumetric method for NO adsorption equilibrium and kinetics behaviors study was shown in Fig. S2 (a), Supplementary data. Both high-precision pressure transducers with a precision of 0.05% of the scale span 20 MPa (Super TJE, Honeywell International Inc., U.S. A.) and natural convection air oven with temperature fluctuation range lower than 0.1 °C (UN450, Memmert GmbH & Co. KG, Germany) were employed. The detailed information of the apparatus and the operation procedures for volumetric method could be found in Refs. [5,8,19].

NO can be easily oxidized when exposed to oxygen (2NO + O₂ = 2NO₂), thus all the cells including sample cell and reference cell shown in Fig. S2 (a), Supplementary data, were flushed with inert helium (He) and completely evacuated to effectively drive out the trace amounts of air (oxygen) previously presented in the cells. In addition, Gas Chromatography (GC) analysis toward the gas sample collected from the cells after He purge also confirms the non-existence of air (oxygen) in cells as shown in Fig. S3, Supplementary data.

The true experimental observed value, called Gibbsian surface excess adsorption amount (GSE) [20], can be calculated by Eq. (1):

$$\Delta GSE = \frac{1}{RmT} \left(\frac{P_2 V_{RC}}{Z_2} + \frac{P_1 V_V}{Z_1} - \frac{P_3 V_{RC}}{Z_3} - \frac{P_4 V_V}{Z_4} \right) \quad (1)$$

where R is the universal gas constant, 8.314 J·mol⁻¹·K⁻¹; m is the mass of the coal sample, g; T is the isothermal temperature, K; P_1 and P_2 are the initial pressures of sample cell and reference cell, respectively, Pa; P_3 and P_4 are the final pressures of reference cell and sample cell corresponding to the adsorption equilibrium, respectively, Pa; V_{RC} and V_V are the volumes of reference cell and the void volume of sample cell contained coal sample determined by He, respectively, ml; Z_1 , Z_2 , Z_3 and Z_4 are compressibility factors of NO corresponding to P_1 , P_2 , P_3 and P_4 at T , respectively. By careful

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