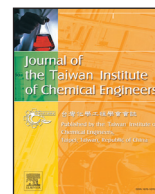




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Selective adsorption of greenhouse gases on the residual carbon in lignite coal liquefaction

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ABSTRACT

Adsorption of CO₂, CH₄, and N₂ on activated carbon now is considered as a promising approach for greenhouse gas capture and/or separation. The objective of this study was to determine whether the residual carbon in lignite coal liquefaction could be used as an inexpensive and effective replacement for activated carbon. The chemical structure and functional groups transformation of residual carbon were analyzed using Fourier transform infrared (FTIR) and nuclear magnetic resonance (NMR). The adsorption properties of CO₂, CH₄, and N₂ on the residual carbon were investigated at temperatures of 0 °C, 10 °C and 20 °C up to a pressure of 100 kPa. The adsorption kinetics and thermodynamics were also discussed from the uptake curves with the isothermal model. The separation of CO₂/CH₄, CH₄/N₂, and CO₂/N₂ binary mixtures was determined using ideal adsorbed solution theory (IAST) model. At 0 °C and 100 kPa, the simulated selectivities of CO₂/CH₄, CH₄/N₂, and CO₂/N₂ in 50/50 vol.% were 35, 11, and 38, respectively. Significant increased values than have previously been reported in the literature. The residual carbon from solvothermally treated lignite coal could be developed for greenhouse gas capture or biogas purification to achieve fuel grade quality.

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1. Introduction

Separation of CO₂ and N₂ from CH₄ is the most important issue to effectively utilize energy and to sequester the greenhouse gases [1]. Most of CO₂ is released from flue gases in which the mixture consists of ~70% N₂ and 15% CO₂. CH₄, having a much higher global warming potential, mainly originates from the anaerobic digestion of organic waste material in landfill sites. The N₂ level is as high as ~20% in landfill gas. Therefore, CO₂ and CH₄ capture and separation from N₂ are important to limit the release of these chemicals to the atmosphere and to pure or more concentrated CH₄ to achieve fuel grade quality [2].

Efforts through membrane, chemical and physical adsorption [3–5] have successfully for CH₄ storage and CO₂ capture from N₂. Of the diverse technologies being developed for gas separation, adsorption is an applicable and cost-efficient technology [3,5–7]. Most recently, various materials, such as activated carbons, zeolites, porous clays, and alumina, have been investigated as ad-

sorbent. Any attempt to utilize the waste with enriched functional groups is beneficial to environmental issue.

Carbon materials mostly derived from the waste have been widely studied on adsorption kinetic studies of CH₄ and CO₂. For example, Carrott et al. [8] evaluated the kinetic properties of pure-component CO₂, O₂, N₂ and CH₄ on carbon materials prepared from polyester textile fibers. Rodríguez-Blanco et al. [9] prepared carbon materials from cotton fabrics by different chemical activating methods. They correlated the kinetic data with the activating conditions. Introduction of amine groups into porous materials [6,10,11] have improved affinity to CO₂. From a view of industrial utilization, however, preparation of carbon materials is a complex process involving several steps to obtain high pore volumes and sufficient functional groups.

Lignite is the lowest rank of coal, and high CO₂ emission from direct combustion results in its environmental incompatibility [12]. Coal liquefaction is considered a technically possible, efficient and environmentally friendly means of low rank coal utilization. Avid et al. [13] and Liu et al. [14] indicated coals can be easily and rapidly solubilized in a suitable solvent at elevated temperatures to obtain valued gas and oil. After solvothermal treatment, oxygen-containing functional group of residual carbon was substituted by

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Table 1
Proximate and ultimate analysis of lignite coal.

	Moisture, M^a (%)	Ash, A^d (%)	Volatile matter, V^{daf} (%)	Sulfur S_t^d (%)	Caloric value, Q^{daf} (kcal/kg)	Carbon, C^{daf} (%)	Hydrogen, H^{daf} (%)	Nitrogen N^{daf} (%)	Oxygen O^{daf} (%)
Lignite	9.4	13.3	47.0	0.56	5712	64.2	5.74	0.26	29.8

O^{daf} - by difference, daf = dry, ash-free.

hydrogen or carbon-substituent [14] that dominant selective adsorption for gas separation.

In this study, the potential application of the residual carbon from lignite coal after liquefaction process in greenhouse gas adsorption and separation was investigated. The adsorption properties of CO_2 , CH_4 , and N_2 were studied on the residual carbon. Isotherm data were used to evaluate the adsorptive separation of CO_2/CH_4 , CH_4/N_2 , and CO_2/N_2 mixtures through IAST. The adsorption kinetics and thermodynamics of CO_2 , CH_4 , and N_2 were also investigated.

2. Experimental

2.1. Residual carbon from coal liquefaction

The two-step coal treatment process involves the pretreatment of coal and liquid upgrading of the products. The detailed is described in Supporting Information.

2.2. Characterization of the samples

The FTIR spectra were recorded on Interspec 200-X FTIR spectrometers. A KBr disk with 1% finely ground samples was also used in PIKE diffusion IR accessories. The scan range is $4000\text{--}400\text{ cm}^{-1}$, and each sample was executed with 32 scans. 1H NMR and ^{13}C NMR spectra of the samples dissolved in deuteriochloroform with chloroform were characterized using a BRUKER AVIII 600 MHz spectrometer.

2.3. Pure gas adsorption measurements

In this study, the gas-adsorbed quantity was measured with Micromeritics ASAP 2020 by increasing the pressure in 15 steps to 100 kPa, allowing an equilibrium time for each step of 600 s from the moment the rate of pressure change less than 0.5%. Exactly the same procedure was used for each determination at $0^\circ C$, $10^\circ C$ and $20^\circ C$, respectively. The gas purity specifications of CO_2 , CH_4 , and N_2 were both 99.99%. Some of the tested samples were duplicated and the results were taken as average. Analytical errors were 2.1%, 1.5%, and 1.6% for CO_2 , CH_4 , and N_2 , respectively. The detection limitation of analytical measurements was 0.01 mmol/g.

2.4. Adsorption selectivity

The selectivities of the binary mixtures (CO_2/N_2 , CH_4/N_2 , and CO_2/CH_4) in the upgraded solid product are predicted by using Ideal Adsorbed Solution Theory (IAST), described elsewhere [15]. The selectivity factor is described as Eq. (1).

$$S = \frac{x_i/x_j}{y_i/y_j} \quad (1)$$

where x_g , y_g are the equilibrium mole fractions of component g ($g = i, j$) in the adsorbed and gas phase, respectively.

2.5. Adsorption thermodynamics

During the adsorption process, the temperature change could be predicted using the isosteric heat of adsorption (Q_{st} , kJ/mol). Q_{st}

Table 2
Main inorganic elements in lignite coal and ash (% db).

element	Lignite coal	Ash
Si	2.4	10.2
Al	1.3	5.3
Fe	0.79	2.1
Ca	3.7	12.4
Mg	0.17	0.86
S	–	2.9
Na	–	–
K	0.14	0.52

– Not detectable.

can be calculated from the adsorption isotherms at different temperature [16] using Clausius–Clapeyron equation given by Eq. (2).

$$Q_{st} = -RT^2 \left(\frac{\partial \ln P}{\partial T} \right)_q \quad (2)$$

where T , P , R and q refer to the temperature (K), the pressure (kPa), the gas constant, and the adsorbed amount (mmol/g), respectively. Q_{st} is obtained from a line by plotting $\ln P$ versus $1/T$ for the same adsorbed amount for different isotherms.

2.6. Adsorption kinetics

In this study, the isothermal model was used while assuming that diffusion is slower than heat transfer. At a pressure of 2 kPa, the adsorption kinetic curves of CO_2 , CH_4 , and N_2 on the upgraded solid were measured at three different temperatures ($0^\circ C$, $10^\circ C$ and $20^\circ C$). Prior to gas adsorption, the samples were degassed under below 1 Torr at $120^\circ C$ for at least 1 h. The diffusion time constants (t) is obtained using Eq. (3) while the fractional uptake is larger than 0.7.

$$1 - \frac{m_t}{m_\infty} = \frac{6}{\pi^2} \exp\left(-\frac{D_c}{r_c^2} \pi^2 t\right) \quad (3)$$

where m_t/m_∞ is the fractional uptake, and D_c/r_c^2 is the diffusion time constant.

3. Results and discussion

3.1. Physico-chemical characteristics

The residual carbon used in this study was obtained from lignite coal liquefaction. First the properties of lignite coal were investigated. Table 1 shows the results from the proximate and ultimate analyses, indicating the coal is rich in vitrinite and with low ash and sulfur contents. The volatile matter and carbon contents were $V^{daf} = 47.0\%$ and $C^{daf} = 64.2\%$, respectively. The chemical composition of ash is shown in Table 2. The contents of the main inorganic elements in ash show the relatively lower silicates and higher calcium and magnesium oxides than those in bituminous ash. The petrographic photographs, the maceral group compositions, and chemical characterization through FTIR spectroscopy as well as ^{13}C and 1H NMR analysis are presented in Supporting Information. The results show that this coal is low-rank B2 mark lignite that is suitable for thermal processing.

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