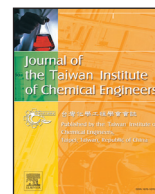




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Surface modifications of carbonaceous materials for carbon dioxide adsorption: A review

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ABSTRACT

Carbon capture and storage is a crucial technology for reducing atmospheric CO₂ emissions from power plants and other industrial facilities. While many technologies have been developed, the high cost and energy requirements of current CO₂ capture methods still are the primary barriers. Adsorption is one of the cost-effective options for CO₂ capture because of its low energy requirement, cost advantage, and ease of applicability over a relatively wide range of temperature and pressure. However, the success of adsorption methods depends on the development of the adsorbents with high CO₂ selectivity and capacity and easy regeneration. Although some adsorbents exhibited good CO₂ adsorption capacity, their adsorption for water vapor is an important issue for this purpose because water usually exists in flue gas streams. Activated carbons are proposed as suitable candidates for CO₂ capture: they do not require moisture removal, present a high CO₂ capacity at ambient pressures, and are easy to regenerate. However, the low selectivity to CO₂ for activated carbons is the restriction. This article reviews recent advances in surface microstructure or chemistry of carbonaceous materials as CO₂ adsorbents with emphasis on the reactions of surface functionalization, CO₂ adsorption performance and mechanism on the carbons with different features.

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1. Introduction

The growing environmental concerns for global warming and climate change are reflected in the successive activities related to improve CO₂ capture and storage. Warming of the climate system was unequivocal, and since the 1950s, many of the observed changes were unprecedented over decades to millennia [1]. The Intergovernmental Panel on Climate Change (IPCC) in 2014 reported that scientists were more than 95% certain that global warming was mostly being caused by anthropogenic activities, mainly increasing concentrations of greenhouse gases such as methane and CO₂.

The IPCC [2] recommended carbon capture and storage (CCS) as a three-stage strategy for reducing CO₂ emissions, which involves: (i) separation, (ii) transportation, and (iii) storage of CO₂. The two latter steps have been already developed; however, CO₂ capture is

still an unsolved issue. This step accounted for about two thirds of the total cost, which prevented large-scale application of CCS [3]. Therefore, intensive studies have been carried out worldwide to improve the existing technologies and/or develop new ones for CO₂ capture.

Carbon dioxide emissions from fossil fuel combustion and industrial processes contribute about 78% to the total greenhouse gases (GHG) emission increase between 1970 and 2010, with a contribution of similar percentage over the 2000–2010 period. Fossil-fuel-related CO₂ emissions reached 32 ± 2.7 Gt CO₂/yr, in 2010, and grew further by about 3% between 2010 and 2011, and by 1 to 2% between 2011 and 2012. Carbon dioxide remains the major anthropogenic GHG, accounting for 76% of total anthropogenic GHG emissions in 2010. Of the total, 16% comes from CH₄, 6.2% from N₂O, and 2.0% from fluorinated gases (F-gases) [1]. The global warming potential of CO₂ is less than that of other GHG, however, its high annual emission into the atmosphere makes it the most imperative anthropogenic GHG.

Flue gas is generally 40–60 °C with a partial pressure of CO₂ at 0.13–0.16 bar. The main component CO₂ must be separated from N₂ and small amounts of other gases. A typical untreated flue

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Table 1

Typical untreated flue gas composition from a power plant burning low sulfur eastern bituminous coal [4].

Species	Concentration (by volume)	
	Unit	Value
N ₂	%	73–77
CO ₂	%	15–16
H ₂ O	%	5–7
O ₂	%	3–4
SO ₂	ppm	800
NO _x	ppm	500
HCl	ppm	100
CO	ppm	20
SO ₃	ppm	10
Hydrocarbons	ppm	10
Total Hg	ppb	1

gas composition from a power plant burning low sulfur eastern bituminous coal was shown in Table 1 [4]. Since most anthropogenic CO₂ was a byproduct of the combustion of fossil fuels from coal-fired plants and large industrial sources, CO₂ capture technologies were commonly classified as pre-combustion capture, post-combustion capture and oxy-combustion systems [5]. Among the different CCS methods available, post-combustion capture appeared to be the most feasible approach as it can be retrofitted to existing power plants without upgrading or modifying the existing systems [6]. Accordingly, much effort has been expended to develop various physical and chemical methods for post-combustion CO₂ capture.

Carbon capture and storage (CCS) is the only technology able to deliver significant CO₂ emission reduction from the use of fossil fuels. Capturing CO₂ was by far the most energy-expensive part of CCS technologies. CCS can reduce emissions not only from power generation, but also from industrial sectors such as iron and steel, refining, petrochemical, and cement manufacturing. The first CCS project applied to coal-fired power generation commenced operation in 2014 in Saskatchewan, Canada. To 2015, 15 large-scale CCS projects are already operating globally, with a further 7 expected to come online by 2018 [7].

To date, many separation technologies can be used: physical absorption, chemical absorption, adsorption, cryogenics separation and membranes. Most of commercial CO₂ capture plants used amine-based processes and wet scrubbing systems, but it had remarkable disadvantages such as oxidative degradation of absorbents, high corrosion, high energy intensity and not cost-effectiveness associated with the solvent regeneration (25–40% for a coal-fired power station). Moreover, amine and water may enter the gas stream, thus releasing amines (as gas and liquid) emissions to air, while possibly also forming other compounds in the atmosphere after emission. Cryogenic distillation is generally not considered as a practical method to separate CO₂ from flue gases because of high energy consumption [8].

For repeated use, physical adsorption is a preferred CO₂ capture mechanism since it is based on much weaker intermolecular forces and the adsorbent can easily be regenerated by applying a small amount of energy. There have been many different solid adsorbents used to remove CO₂ in post-combustion separation processes. These materials have successfully been adapted to pressure swing adsorption (PSA), which recently has been shown to be a competitive technology even with low-cost technologies such as amine scrubbing [9]. Adsorption is considered as one of the cost-effective options for CO₂ separation because of the low energy requirement, cost advantage, and ease of applicability over a relatively wide range of temperatures and pressures. However, the success of this method depends on the development of the adsorbents with a high CO₂ selectivity and adsorption capacity, easy regeneration and high stability.

The adsorbents of CO₂ should satisfy the following requirements: (i) high CO₂ uptake, (ii) high adsorption rate, (iii) high selectivity against other molecules (*i.e.*, N₂) competing for adsorption sites, (iv) easy regeneration, (v) low-cost and high availability, (vi) stable adsorption capacity of CO₂ after repeated adsorption/desorption cycles, and (vii) adequate mechanical strength [9–11]. Some of the adsorbents for CO₂ separation and capture such as activated carbons [10], activated carbon fibers (ACFs) [12], carbon nanotubes (CNTs) [13], graphene sheets [14], graphene oxide [15], carbon molecular sieves [16], zeolites [17], metal-organic frameworks (MOFs) [3], microporous coordination polymers (MCPs) [18], zeolitic imidazolate frameworks (ZIFs) [19], and metal oxides [20] have been successfully and widely employed in literature.

Industrial applications of these materials, despite the appreciable adsorption capacity, are limited. Among them, MOF and nanoporous carbons have gained much attention. Yazaydin et al. [21] showed that MOFs exhibit high CO₂ adsorption capacities up to 8.5 mmol/g at 1 bar and 25 °C. Despite the excellent adsorption capacities of MOFs, they are much more expensive than most carbonaceous adsorbents, especially commercially available activated carbons [22]. Moreover, MOFs are water vapor sensitive, they can adsorb a large amount of water but not all water can be desorbed due to chemical adsorption [23,24]. Flue gas contains a considerable amount of water vapor; thus, MOFs are unlikely to be used in fossil fuel-fired power plants as CO₂ adsorbents unless they have significant improvement for hydrophobicity and long-term stability towards moisture. In addition, it still suffers from structural/chemical instabilities of metal ion or hydrogen bonding. Due to these reasons, MOFs are less feasible from the viewpoint of industrial applications [22].

Zeolites have also shown promising results for separating CO₂ from gas mixtures. However, zeolites are very hydrophilic, which is a problem as the flue gas contain significant amount of water. Therefore, the presence of water inhibits CO₂ adsorption capacity of zeolites [25]. MCPs and ZIFs synthesized with N-containing organic compounds as the crosslinker [26] often suffered from structural instability and inefficiency for CO₂ selectivity in the presence of water and thus were limited in their widespread use [27].

In contrast, porous carbon-based materials display a comparatively advantages over a wide range of operating conditions, standing out in terms of low cost, high thermal stability, availability, large surface area, easy-to-design pore structure, high resistance to water due to their hydrophobicity, good chemical resistance to both alkaline and acidic media, low energy requirements, and high efficiency for regeneration [28]. Activated carbons are being proposed as suitable candidates for CO₂ capture: they do not require any moisture removal, present a high CO₂ adsorption capacity at ambient pressure and, moreover, they are easy to regenerate. However, the low selectivity to CO₂ for activated carbon is the restriction. It has been recognized that the surface chemistry of activated carbons can strongly affect the adsorption capacity [11].

Carbon-based adsorbents have adequate adsorption capacity for CO₂ at low concentrations and ambient temperatures. However, the adsorption capacity reduces with increasing temperature, which can be enhanced greatly using chemical modification. For instance, when the basic groups were introduced on the surface of porous carbonaceous adsorbents, the basic sites having intermediate affinity would exhibit stronger interaction with CO₂ than that of pure porous carbon adsorbents, but the affinity was too weak to induce pure chemical adsorption for CO₂. In addition, the effect of the surface area of carbon-based adsorbents on adsorption capacity of CO₂ becomes less important while increasing temperature. This indicates adsorption mechanism of CO₂ at high temperature is not completely physical adsorption [9].

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