

## Review Article

# Dry Capture of Low-level CO<sub>2</sub> from Public Indoor Spaces using Chemically Modified Carbonaceous Adsorbents - A Review

Sujeong Heo, Wooram Kim, Tae Jung Lee, Adedeji A. Adelodun<sup>1)</sup>, Young Min Jo\*

Department of Applied Environmental Science, Kyung Hee University, Yugin-si, Gyeonggi-do, Republic of Korea  
<sup>1)</sup>Department of Marine Science & Technology, School of Earth & Mineral Sciences, The Federal University of Technology, P.M.B. 704, Akure, Nigeria

**\*Corresponding author.**

Tel: +82-31-201-2485

E-mail: [ymjo@khu.ac.kr](mailto:ymjo@khu.ac.kr)

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**ABSTRACT** Although CO<sub>2</sub> is prominent as the most important greenhouse gas, responsible for 64% of anthropogenic global warming, it is also a viable indicator for indoor air quality (IAQ). Due to the incessant increase in the human population and residence time indoors, the need to control indoor CO<sub>2</sub> levels has become exigent. To this effect, dry-phase removal technology via adsorption with zeolites, activated carbons (AC) and activated carbon fibers (ACFs) had sufficed. Chemically modified AC and ACF surfaces through alkali impregnation have been used to improve their selectivity toward CO<sub>2</sub> at room temperature. Here we appraise the various methods in the literature and carry out performance evaluation based on the physical and chemical modification induced by the chemical agents and experimental conditions. This study reviews the improved adsorption of low concentration (0.3%) via surface reformation of commercial carbon-based adsorbents, and the highest adsorption capacity was 2.2 mmol/g CO<sub>2</sub> at the indoor level, which was achieved by AC pellets doped with ammine functionalities.

**KEY WORDS** Indoor Air Quality, CO<sub>2</sub> adsorption, Activated carbon, Activated carbon fiber, Surface Chemistry

## 1. INTRODUCTION

Aside from being the main anthropogenic contributor to global warming, CO<sub>2</sub>, an index indicating indoor air quality (IAQ), is harmful at relatively high concentrations, especially in confined spaces such as offices, schools, subway stations, cars, airplanes, and submarines. Indoor CO<sub>2</sub> concentration (IAQ<sub>CO<sub>2</sub></sub>) above 1000 ppm, causes unpleasantness, fatigue and headache. Therefore, effective management of IAQ<sub>CO<sub>2</sub></sub> is necessary for human health (Satish *et al.*, 2012).

According to Park *et al.* (2020), IAQ<sub>CO<sub>2</sub></sub> in classrooms usually falls in the range of 1,000 to 3,000 ppm. Elsewhere, office meeting rooms have exhibit between 446 and 1,450 ppm CO<sub>2</sub> with normal ventilation, which could rise to 4,920 ppm without ventilation (Vehviläinen *et al.*, 2016). When uncrowded, the IAQ<sub>CO<sub>2</sub></sub> of subway cabins ranges from 550 to 2,680 ppm (Lee *et al.*, 2014). However, it often exceeds 5,000 ppm during rush-hours. Due to the commonly elevated IAQ<sub>CO<sub>2</sub></sub> in public indoor spaces, the need for its monitoring and control. As set by the Environmental

Protection Agency (EPA),  $IAQ_{CO_2} > 1,000$  ppm requires efficient control technologies to ensure the safety of humans during their regular long-duration operations in confined spaces (Vehviläinen *et al.*, 2016).

Among the various techniques for  $CO_2$  capture, dry-phase adsorption is the only process feasibly applicable to indoor scenarios. Considering the low-concentrations of  $IAQ_{CO_2}$ , natural (inevitable) and artificial (necessary) sources, incessant generation, and evasiveness, an adsorbent with a high affinity for  $CO_2$  molecules, hydrophobicity, sanitary safety, ready availability, mechanical stability and easy regeneration is required for sustainable use. Such an adsorbent should be efficient enough to operate under ambient conditions.

Thus, this study comprehensively explores plausible techniques to capture  $CO_2$  at low levels of 3,000 ppm, as studied in some references (Jeong *et al.*, 2019; Kim *et al.*, 2017; Adelodun and Jo, 2013), focusing more on dry adsorption. In this review, ultra-high-efficiency materials such as metal organic frames (MOF), carbon nanotubes (CNT) and hierarchically porous zeolites including ZSM and SBA were not addressed due to their high cost and lack of feasibility for indoor fields.

## 2. ADSORPTION OF $IAQ_{CO_2}$

In the past two decades, researchers have intensified efforts to develop adsorption technology to control of  $IAQ_{CO_2}$  at room temperature. Popular among the approaches is the use of amine-based or amine-functionalized adsorbents, derived by tethering amine groups onto supports by grafting or chemical impregnation and immobilization of liquid amines (Moloney *et al.*, 2011).

When compared to other technologies, adsorption offers some distinct advantages, particularly for indoor air control through product recovery, non-necessity of pollutant control during product recycling, automation, selectivity at trace and ultra-trace levels, excellent control and response to process change, cost-effectiveness, environmentally friendliness, high adsorbent reusability cycles, high energy efficiency etc.

The major factors that influence adsorption include the chemical properties of the chemical properties of the adsorbate and adsorbent, the adsorbent's surface area and porosity, experimental conditions such as temperature, humidity, flow rate, gas matrix, pressure, etc.

Solid adsorbents (in packed or fluidized beds) are more energy efficient than wet scrubbers, in which  $CO_2$  molecules dissolve into the fluid. Whereas in adsorption, the process involves either weak physisorption by van der Waals attractions or chemisorption by covalent bonding interactions between the  $CO_2$  molecules (adsorbate) and an adsorbent surface. However, no sharp distinction exists between physical and chemical adsorptions. Rather a somewhat gradual transition scaled by the strength of the interaction at the interface occurs. At the equilibrium point at which adsorption rate equals that of desorption, the  $CO_2$ -laden solid can be refreshed by subjecting it to one of pressure, vacuum, or temperature swing adsorptions, i.e. PSA, VSA and TSA respectively. Recently, combinations of two or more of these techniques have been adopted to achieve faster and more efficient regeneration.

Zeolites, composed of Al and Si oxides, are an easily-attainable, commercially-available adsorbent with a high adsorption affinity for various gases. Zeolites are microporous crystalline materials with adsorptive and ion exchange properties, often used to capture gaseous pollutants due to their characteristic high surface area and three-dimensional pore structures (Ye *et al.*, 2012; Liu *et al.*, 2011). The type of surface cations, polarity, surface area, and pore volume determine zeolites' adsorption capacities (Plaza *et al.*, 2007).

Amongst the noble functional zeolitic crystals, ZSM structures are excellent ones purposefully designed for  $CO_2$  capture. These zeolites tend to maximize electrostatic interactions between the surface and  $CO_2$  molecules. Besides, metal organic framework (MOF) is another porous solid material of immense porosity up to the region of  $10,000 \text{ m}^2/\text{g}$  (Ghanbari *et al.*, 2020). These three-dimensional adsorbents consist of secondary building units and connectors, whose length can be adjusted depending on the target molecule's kinetic diameter. However, the cost of gaseous adsorption using these 'future' materials is still high, and it also requires handling expertise (Jiao *et al.*, 2016; Wang *et al.*, 2014).

On the other hand, carbon-based adsorbents such as activated carbon (AC) and activated carbon fiber (ACF) are incredibly porous materials with appreciably high surface area to unit weight ratio. These carbon-based adsorbents are currently the most popularly used due to their ruggedness, robustness, relatively low-cost, and reusability (Yu and Chuang, 2017). Capturing indoor air pollutants using carbonaceous adsorbents at normal

temperatures is considered sustainable. Thus, recent advances on the surface modification of carbon-based adsorbents to enhance their selective and capacitive IAQ<sub>CO<sub>2</sub></sub> adsorption are appraised in this study.

Adsorption selectivity is the measure of the preferential attraction of a material towards one or a few components in a mixture or matrix in which efficient contact is feasible between the target and the material (Adelodun *et al.*, 2015).

Selectivity in a physical adsorption system may depend on differences in either equilibrium or kinetics, but the great majority of adsorption separation depends on equilibrium-based selectivity (Clarkson and Bustin, 2000). The selective adsorption of CO<sub>2</sub> has become a tremendous research interest in recent years. Most of these studies on the selective separation of CO<sub>2</sub> from N<sub>2</sub> atmospheres include the use of metal-organic frameworks (Wu *et al.*, 2014; Chen *et al.*, 2013; Hou *et al.*, 2013), synthetic zeolites (Sethia *et al.*, 2014; Dangi *et al.*, 2012), polymers (Nagarkar *et al.*, 2011) and carbon-based adsorbents (Chandra *et al.*, 2012; Thote *et al.*, 2010; Plaza *et al.*, 2009a).

When properly modeled, the experimental investigation for the selective capture of IAQ<sub>CO<sub>2</sub></sub> proved that the matrix of the inlet gases should comprise other common air constituents, be they natural such as Ar and moisture or anthropogenic such as VOCs and particulate matters. Thus, with CO<sub>2</sub> being the major weak Lewis acid in indoor air, its selective adsorption is feasible by increasing the surface basicity of the adsorbent (Adelodun *et al.*, 2015).

### 3. ACTIVATED CARBON (AC)

Due to the established demerit of zeolites with CO<sub>2</sub> adsorption at high humidity, activated carbons are sufficient as suitable alternatives. ACs do not require any moisture removal, and they present a high adsorption capacity at ambient pressure. Moreover, they can easily regenerate for multiple reuses with no or insignificant drop in original efficiency (Plaza *et al.*, 2009b). The surface chemistry of AC can strongly affect the adsorption of CO<sub>2</sub> (Pevida *et al.*, 2008). Due to the acidic nature of CO<sub>2</sub> with weak Lewis acid, it is expected that the introduction of Lewis bases onto the AC surfaces may increase the adsorbent's affinity toward CO<sub>2</sub> molecules. Besides, surface impregnation, basifying the AC surfaces also

includes increasing the  $\pi$ -electron density of the basal planes, which has also been reported to exhibit basic properties (Lahaye, 1998). To achieve surface basification of AC, heat treatment and amine-doping such as amination and ammoxidation are the respective physical and chemical approaches popularly used (Dali *et al.*, 2012). In addition to this, the CO<sub>2</sub> capture capacity could be enhanced by engineering the porous structure of AC in a hierarchical design adapted for the adsorbate. Sevilla and Fuertes (2011) and Sevilla *et al.* (2011) reported that ACs predominated with 1–2 nm width pores often exhibit excellent CO<sub>2</sub> adsorption selectivity, unlike those with narrower micropores or with a mesoporous pore network.

Elsewhere, some efforts had been exemplified to use AC to support the highly successful CO<sub>2</sub> absorbents by implying monoethanolamine (MEA) and glycine (Gly). This approach was conceived to improve the basic adsorbents' portability, a property imperative for indoor use (Lee *et al.*, 2013). Later, chemical modification by alkali-metal ion exchange also was carried out on Gly-based absorbents (Lim *et al.*, 2016). However, such alkali impregnation significantly reduce the adsorbent's specific surface area and microporosity. Such depreciation in the textural properties resulted in a decrease in pure CO<sub>2</sub> sorption implying low CO<sub>2</sub> storage capability. Contrarily, the 5 M supported MEA exhibited significantly enhanced low-level CO<sub>2</sub> capture capacity from 0.016 to 1.064 mmol/g consistently.

The surface of the chemically modified AC adsorbents as observed by X-ray photoelectron spectroscopy (XPS) was tethered with amine, pyrrole and quaternary-N. In reducing potency, pyridine, amine, and pyrrolic-N have been identified as N-functionalities that contribute significantly to CO<sub>2</sub> selective adsorption (Adelodun *et al.*, 2015; Lim *et al.*, 2015).

#### 3.1 Amination of AC

A highly bilateral interforce between adsorbent and CO<sub>2</sub> molecules could be formed by providing strong covalent bonds with metal oxides as those found in MOFs. However, the financial cost to fabricate, handle, and regenerate MOFs are excessively high (DeSantis *et al.*, 2017). For this reason, efforts have been made to up the selective efficiency of AC, being a cheaper and readily available alternative. In accordance, amine groups are on the AC surface using amination, a process in which thermally fragmented ammonia radicals are impregnated

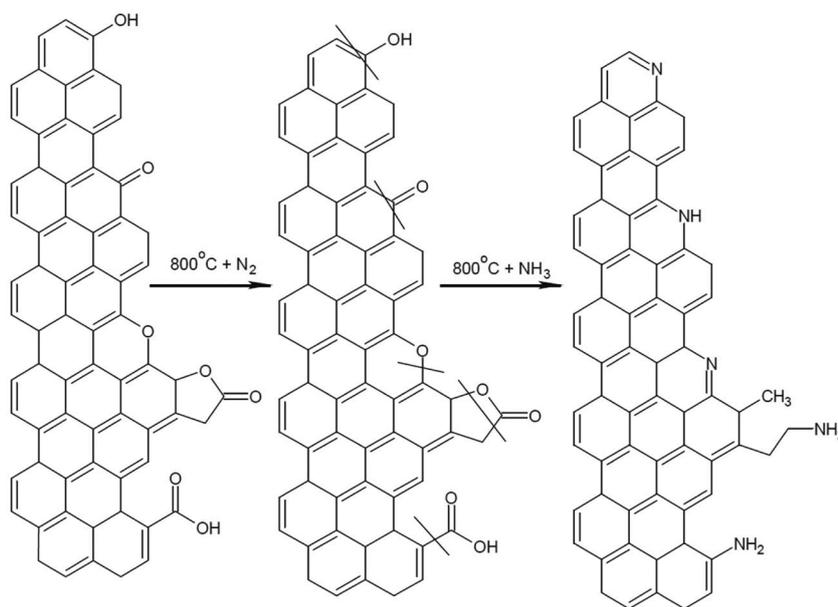


Fig. 1. Schematic diagram showing the amination process (Kim *et al.*, 2017).

Table 1. Summary of surface amination of AC adsorbents for CO<sub>2</sub> capture.

Material	Treatment	Target	Reference
Almond shell	800°C; 2 h	CO <sub>2</sub>	Plaza <i>et al.</i> , 2009
Commercial	200–1000°C; 2 h	CO <sub>2</sub>	Przepiorski <i>et al.</i> , 2004
Wood & peat	200–800°C; 2 h	CO <sub>2</sub>	Pevida <i>et al.</i> , 2008
Olive stone	400–900°C; 2 h	CO <sub>2</sub>	Plaza <i>et al.</i> , 2009
Biomass, sludge, coke	400°C; 2 h	CO <sub>2</sub>	Gray <i>et al.</i> , 2004

on AC surfaces. The process is theoretically depicted in Fig. 1.

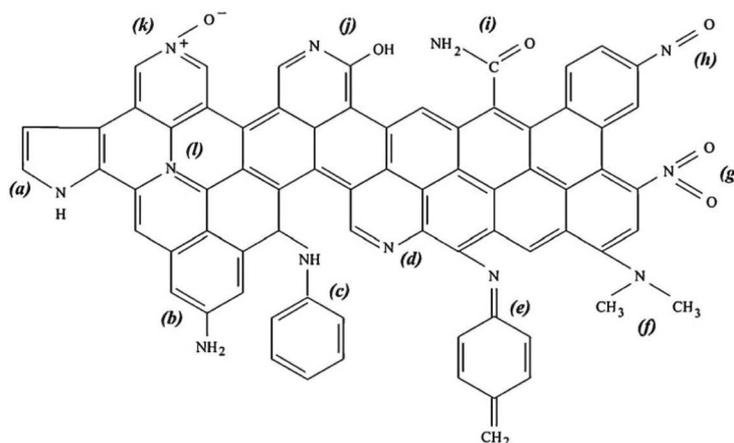
At temperature between 200 and 1, 200°C, the amine radicals viz. ·NH<sub>2</sub>, :NH, atomic :N and ·H, would attack the carbon surface to form surface nitrogen functionalities (SNFs) (Shafeeyan *et al.*, 2010; Przepiorski *et al.*, 2004; Soto-Garrido *et al.*, 2003). Amination simply basicifies the AC surface, turning it into a Lewis base-laden environment onto which CO<sub>2</sub>, a Lewis acid, could be easily scavenged. The pioneering studies on amination in the past decade are summarized in Table 1. Here, we found the stock material for the AC to vary widely.

### 3.2 Ammoxidation of AC

Instead of pure ammonia gas used in amination, ammoxidation involves using ammonia-air gas mixture for nitrogen enrichment of carbon surface. Jansen and Bekkum (1994) observed differences in the surface chemistry of

aminated and ammoxidized carbons carried out at 600–900°C. Therein, they reported differences in the amide-lactam-imide ratio between the techniques. The respective nitrogen contents and surface fractions obtained were 4.4 wt% and 0.14 wt% respectively for amination, while ammoxidation achieved 3.2 wt% and 0.11 wt%.

According to Pietrzak *et al.* (2007), ammoxidation with ammonia: air of 1 : 3 at 300°C, depends mainly on the period of the process ammoxidation is performed. They reported that the ammoxidation of brown coal decreases its surface area. Whereas, when used as a thermal activation process, it favors the formation of surface oxygen groups (SOFs) as well as enhancing the carbon's basicity. However, using the similar ammoxidation condition (NH<sub>3</sub> : air = 1 : 3) but at 350°C, a further study carried out by Pietrzak (2009) on volatile bituminous coal resulted in enhancement of both the textural (pore structure and surface area) and surface basic property as varieties



**Fig. 2.** Detailed expression of nitrogen-containing complexes possibly found on activated carbons (Shafeeyan *et al.*, 2010).

and large amount of SNFs were incorporated.

The thermogravimetric (TG-DTG) curves assessed by Wachowski and Hofman (2006) showed that coal samples ammoxidized at higher temperature exhibit slightly lower thermal stability. This occurrence could be due to the formation of less refractory SOFs usually associated with air-treatment at low temperatures.

A group of researchers investigated and identified a peculiar application of ammoxidized carbons. They reported their series of experimentals over five years, that ammoxidation could improve the capacitance of ACs. Their research focused on the capacitance behavior of ammoxidized coal and nanotube carbon materials purportedly designed for the electrical industry (Jurewicz *et al.*, 2006, 2004 and 2002).

Shafeeyan *et al.* (2010) has provided the precise description SNFs plausibly found on activated carbons, especially after amination as depicted in Fig. 2. The research team saw the need to distinguish pyridine and pyridines from each other, mainly when the study of CO<sub>2</sub> adsorption is concerned due to the varied affinity for the gas molecules.

Chemical activation of carbon-based materials with a strong alkali such as concentrated KOH solution has been investigated and reported in recent works (Labus *et al.*, 2014; Mitome *et al.*, 2013). Identical methods that involve soaking and mixing the solid adsorbent with KOH solution, then sintering at a very high temperature under an inert atmosphere were used in all the reports. Usually, due to the increased instability of the carbon surface and the possible presence of highly reactive K<sub>2</sub>O, the treated carbons are then transferred into a concentrated acid

solution (usually HCl) for neutralization. Some possible inside reactions are presented in equations (1) to (5). Results showed that improved microporosity achieved does enhance CO<sub>2</sub> capacity capture of prepared adsorbents.



Before 2014, the reported CO<sub>2</sub> capture selectivity at indoor levels by modified adsorbents did not show impressive achievement despite of several efforts for the adsorption of IAQ CO<sub>2</sub> by AC. Even pre-oxidation treatment was later incorporated to populate the AC surface before the displacement reaction induced by the amine radicals. However, not until Adelodun *et al.* (2014b) adopted the alkalization method was the target selectivity met. Later, the KOH treatment concept was adopted for basifying AC surface through alkylation, in which the thermally doped KOH was left on the AC by amine-stabilization (Adelodun *et al.*, 2014b). It was the first time that sintered KOH could retain on a carbonaceous surface under air atmosphere without any burn-off due to oxidation. In their report, Adelodun *et al.* (2014b) were able to match the maximum adsorption capacity of modified AC ( $q_{\text{max}}(100\%-\text{CO}_2)$ ) with its indoor (0.3%-CO<sub>2</sub>) selective adsorption capacity of 2.23 and 2.20 mmol/g, respectively.

## 4. ACTIVATED CARBON FIBER (ACF)

Generally activated carbon fibers (ACFs) used for CO<sub>2</sub> adsorption are prepared from polymeric materials such as polyacrylonitrile (PAN). The systematic processes involved oxidation, activation and amination, focusing on the impregnating SNFs (Adelodun and Jo, 2013). Similar to the basification of AC, the specific surface area and pore volume of ACF were reduced after chemical amination. This is attributed to the presence of more SNFs on the external surface and inner pore walls. Amination is the substitution of a hydrogen bound to a carbon with an amino group.

In our previous studies, the ratio of micropores to the total volume ranged between 0.85 and 0.91, with an average pore width of 1.6 nm. Additionally, the induced SNFs such as imine, pyridine, and pyrrole present favorable affinity with CO<sub>2</sub>. The aminated ACF exhibited enhanced low-level (3,000 ppm) CO<sub>2</sub> adsorption from 0.40 mmol/g at room temperature. The selective separation of CO<sub>2</sub> against dry air (O<sub>2</sub> & N<sub>2</sub>) also increased from 1.00 to 4.66 by amination.

### 4.1 Alkalization of ACF

While amination is a dry-phase process where thermally decomposed ammonia gas impregnate basic N-groups on the carbon surface, in this study, alkalization is the wet-phase alternative of concentrated solutions of alkali to incorporate alkali or alkali earth metals on the adsorbent. To optimize the advantage of fibrous adsorbents, activated carbon nanofibers (ACNFs) were fabricated via electrospinning, using polyacrylonitrile (PAN) as a precursor (Kim *et al.*, 2017). The spun fibers were 390 to 580 nm in thickness, with a surface area in the range of 27.3 to 300 m<sup>2</sup>/g. The surface structure was improved by programmed thermal activation at 800°C in a CO<sub>2</sub> atmosphere, resulting in enhancement of specific surface area up to 542 m<sup>2</sup>/g. It was also found that the nitrogen-groups including pyrrole and pyridine were created during the activation, which in turn facilitated the selective adsorption of CO<sub>2</sub>. The final adsorption capacity was 2.74 mmol/g for pure CO<sub>2</sub> flow and 0.74 mmol/g for 3,000 ppm.

Further, the fabrication of doped-ACFs from melamine-blended polyacrylonitrile (MACNFs) improved their IAQ-CO<sub>2</sub> removal (Jeong *et al.*, 2019). Here, melamine was used as an alkali dopant for basic SNFs. Upon final CO<sub>2</sub> activation, the specific surface area and micro-

porosity were enhanced. As per chemical properties, the surface basicity was improved through the significant tethering of pyridine, allowing for preferential adsorption of CO<sub>2</sub> over N<sub>2</sub>. The optimum melamine doping condition achieved the high CO<sub>2</sub> adsorption capacity of 3.15 mmol/g.

Another attempt to tether SNFs on activated carbon nanofibers (ANF) was made using tetraethylenepentamine (TEPA) solution (Wang *et al.*, 2020) Further impregnation of TEPA was achieved with preliminary oxidation of the nanofibers with 70% HNO<sub>3</sub>. The effects of HNO<sub>3</sub> and TEPA treatments on the modified ANFs were found that although TEPA impregnation reduced the specific surface area and pore volume of the ANFs from 673.7 and 15.61 to 278.8 m<sup>2</sup>/g and 0.28 cm<sup>3</sup>/g, respectively, the HNO<sub>3</sub> pre-oxidation increased the number of carboxylic groups on the ANF. Upon TEPA loading, pyridinic nitrogen was formed, which was further enhanced by pre-oxidation. The surface treatment cumulatively increased the amine content from 5.81% to 13.31%. The adsorbent's final adsorption capacity toward 3,000 ppm and 100% CO<sub>2</sub> levels improved from 0.20 and 1.89 to 0.33 and 2.96 mmol/g, respectively.

### 4.2 Comparison between AC and ACF

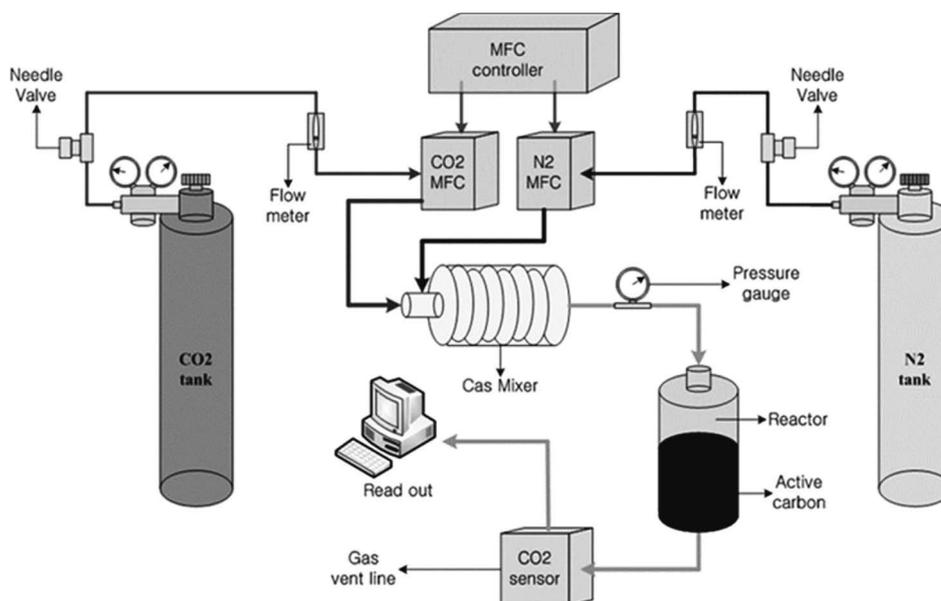
Based on literature review on the properties of AC and ACF with respect to CO<sub>2</sub> adsorption, the respective advantages and disadvantages of the adsorbents are summarized in Table 2.

## 5. EVALUATION OF ADSORPTION CAPACITY

### 5.1 Initial Adsorption Performance

Porous activated carbons are very effective for gas adsorption as driven by the sorbents' pore structure and surface chemistry. Since the adsorbents are used for target harmful gases in the ambient condition, selectivity is an imperative property that ACs and ACFs should possess. For this reason, their capabilities for a lean gas treatment prioritized over their adsorption capacity for pure CO<sub>2</sub>, as required for CO<sub>2</sub> storage and sequestration (Adelodun and Jo, 2013). The present review investigated mainly selective capture of CO<sub>2</sub> in terms of chemical or physical modification of carbonaceous adsorbents.

The adsorption capacity toward 3,000 ppm CO<sub>2</sub> (in binary mixture with N<sub>2</sub>) was fed to the bed filled with



**Fig. 3.** A schematic representation of lab-scale set-up for low-level CO<sub>2</sub> adsorption.

**Table 2.** Comparison of the advantages and disadvantages of AC and ACF.

S/N	Property	Activated carbon	Activated carbon fiber
1	Adsorption capacity	Slightly higher	Slightly lower
2	Adsorption selectivity	Significantly superior	Inferior
3	Specific surface area	Lower	Higher
4	Porosity	Hierarchical	Uniform & super-microporous
5	Durability	Pellets are durable	Depends on precursor
6	Regeneration	Requires high temperature or pressure	Easier to regenerate
7	Moisture resistance	Higher, but trapped moisture is refractory to desorb	Lower, but easier to desorb trapped moisture
8	CO <sub>2</sub> concentration	Better at higher partial pressures	Better at low partial pressures
9	Fabrication	Numerous starting materials; process is more tedious	Fewer starting materials; process is less tedious
10	Surface modification	Less homogenous	More homogenous

adsorbents. Fig. 3 shows a typical lab-scale adsorption set-up for the evaluation (Adelodun *et al.*, 2015; Lee and Jo, 2009). The SENSEair detector was equipped with a non-dispersive infra-red (NDIR) sensor for a continuous sampling. The amount of adsorbed CO<sub>2</sub> (mmol/g) was evaluated using Equation (1):

$$q = \frac{Q C_0}{M} \left[ \int_0^t \left( 1 - \frac{C_t}{C_0} \right) dt \right] \quad (1)$$

where  $q$  = amount of CO<sub>2</sub> adsorbed (mmol/g),  $Q$  = inflow rate of CO<sub>2</sub> (200 mL/min),  $C_0$  = CO<sub>2</sub> inflow concentration (3000 ppm (i.e., 0.3%)),  $C_t$  = CO<sub>2</sub> concentration (ppm) at saturation time,  $t$  = CO<sub>2</sub> sensor intermittent read time (s), and  $M$  = mass of test sample (g).

As received ACs have been reported to exhibit 3.27 mmol/g adsorption capacity toward pure CO<sub>2</sub> flow (Lim *et al.*, 2013). However, its propensity for low level CO<sub>2</sub> such as 0.3%, as frequently found in public indoor spaces, was extremely poor (0.016 mmol/g). To maintain a fresh and safe IAQ with less than 2000 ppm, various modifications on the carbonaceous adsorbents that have been experimented are summarized in Table 3. Focusing on field plausibility, commercial-grade alkaline reagents were used as dopants to improve the surface affinity of ACs and ACFs toward CO<sub>2</sub>.

Although, surface reformation often reduces the intrinsic specific surface area of carbonaceous adsorbents, causing a low adsorption tendency for pure CO<sub>2</sub>, such a modi-

**Table 3.** CO<sub>2</sub> adsorption capacities achieved using ACs and ACFs.

Basis adsorbent	Modification	CO <sub>2</sub> conc. (%)	Ads. capacity (mmol/g)	Reference
AC	Ca-impregnation	0.3%	0.31	Lee <i>et al.</i> , 2012
	Impregnation of Li with glycine	pure	3.08	Lim <i>et al.</i> , 2012
	NH <sub>3</sub> with KOH	0.3%	0.41	Hong <i>et al.</i> , 2013
	UV-oxidation & NH <sub>3</sub>	0.3%	0.38	Adelodun <i>et al.</i> , 2014a
	NH <sub>3</sub> with KOH	0.3%	2.20	Adelodun and Jo, 2013
	Hi-tem. activation	pure	0.1–0.3	Huang <i>et al.</i> , 2015
ACF	TEPA impregnation	pure	1.89	Wang <i>et al.</i> , 2020
		0.3%	0.33	
	Melamine doping	pure	3.15	Jeong <i>et al.</i> , 2019
		0.3%	0.47	
	Acetonitrile & ferrocene	pure	1.92	Yu and Chuang, 2017
	Urea doping	pure	2.98	Kim <i>et al.</i> , 2017
	KOH treatment	0.3%	0.05	Hwang <i>et al.</i> , 2016
	Ammonia treatment @ 700°C	pure	0.4	Hwang <i>et al.</i> , 2016
	Modified ACF	pure	6.58	Choma <i>et al.</i> , 2016
	Graphite fiber-KOH	pure	1.61	Yuan <i>et al.</i> , 2016
	Modified ACF	pure	3.1	Díez <i>et al.</i> , 2015
	Modified ACF	pure	3.4	Díez <i>et al.</i> , 2015
	KOH treatment	Pure	5.68	Lee and Park, 2013
	ACF	pure	5.68	Lee <i>et al.</i> , 2013
Composite	pure	2.26	An <i>et al.</i> , 2009	
CO <sub>2</sub> activation	0.3%	0.74		

fication adds bilateral interforce, resulting in high selectivity for low CO<sub>2</sub> concentration.

Thus, the adsorption capacities of ACs and ACFs for pure CO<sub>2</sub> are generally larger than 1 mmol/g, while those of IAQ-CO<sub>2</sub> could be as high as 2.2 mmol/g.

Most surface treatments focus on textural modifications, increasing the proportion of supermicropores or ultramicropores with an appreciable degree of consistency. These micro scale pores are preferable for CO<sub>2</sub> molecules whose kinetic diameter is approximately 3.3 nm.

### 5.2 Regeneration and Reusability

One of the demerits of zeolites as a viable CO<sub>2</sub> adsorbent is their affinity for moisture and high energy requirement for regeneration. The latter is due to the predominance of covalent bonding in attaching with CO<sub>2</sub> during chemisorption (Lee *et al.*, 2012). However, since carbon-based adsorbents interact with CO<sub>2</sub> based on weaker physical or physical-chemical interactions, the energy requirement for their regeneration is lower, and hence more feasible (Jeon *et al.*, 2016).

Usually, few adsorption studies attempt to perform regeneration studies after estimating the maximum

adsorption capacity. And, because pure CO<sub>2</sub> adsorption studies are more popular than those of low-level adsorptions, fewer the regeneration and reusability studies on the latter exist (Krishnamurthy *et al.*, 2019). Notably, Adelodun *et al.* (2014b) achieved the highest adsorption amount for low-level (3,000 ppm) CO<sub>2</sub> through five-cycle regenerations on the optimized sample. Generally, the regeneration of AC and ACF sorbents achieves adsorption capacity more than 90% of the initial cycle, provided the physical properties of the sorbent was not compromised during thermal or pressure swing adsorption refreshment.

## 6. MODELING OF ADSORPTION CHARACTERISTICS

During adsorption, the free, non-adsorbed gas and the adsorbed gas molecules are in a dynamic equilibrium, influenced by temperature, partial pressure or the adsorbate's concentration, and the gas composition. The amount of adsorbate can be expressed using either surface concentration (mol/m<sup>2</sup>), or as surface coverage ( $\theta$ , in

%) which is defined as the ratio of the number of adsorption sites occupied to the number of adsorption sites available (Buekens *et al.*, 2008).

The equilibrium partition between the free and the adsorbed state depends on temperature and pressure. It also can be described in terms of an empirical function  $q = f(P, T)$ .

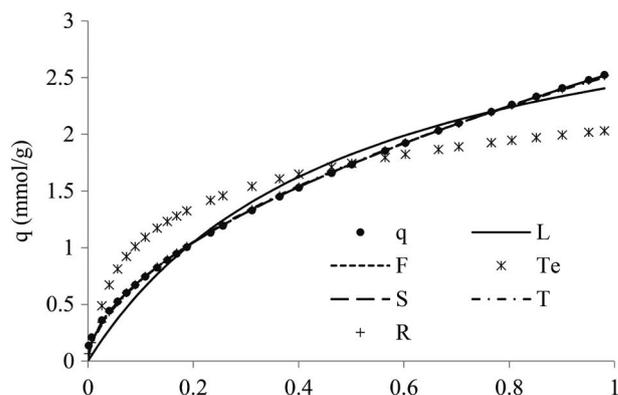
$q = f(P)$	Isotherm	at constant T
$q = f(T)$	Isobar	at constant P
$q = f(P, T)$	Isostere	fixed surface coverage

The adsorption isotherm is the most widely used form for representing equilibrium data, because it is the most amenable with experimental data. Moreover, this is the form in which theoretical treatments are usually developed. Gas separation by adsorption is achieved by one of the three mechanisms viz steric, equilibrium or kinetic effect (Jasra *et al.*, 1991). When the adsorption equilibrium state is determined at a constant temperature, an adsorption isotherm is obtained with changes in pressure. Adsorption isotherms can be used to estimate the surface area and pore volume in various porosity regimes, assessments of the surface chemistry of the adsorbent and fundamental information on the efficiency of various ACs.

The efficiency of adsorbents is estimated on the basis of its capacity, adsorption rate, mechanical strength, and possibility of regeneration and reusability. The adsorbent capacity is the most crucial parameter and the first often determined (Brdar *et al.*, 2012).

Equilibrium data are fundamental information to design an adsorption process. At adsorption equilibrium, the chemical potential of the solute in the gaseous phase is equal to that in the solid phase. In this perspective, equilibrium relationships, describe how pollutants interact with adsorbents. Thus, they are critical for optimizing the adsorption pathways, expressing the surface properties and capacities of adsorbents, and the effective design of the adsorption system (El-Khaiary, 2008).

In general, an adsorption isotherm is an invaluable curve describing the phenomenon governing the retention (or release) or mobility of adsorbate from a fluid-phase to a solid-phase at a constant temperature (Limousin *et al.*, 2007). In our previous study (Adelodun *et al.*, 2015), six models of the numerous isotherms available (Foo and Hamed, 2010) were employed to analyze the adsorption of gaseous CO<sub>2</sub>: 2-parameter models (Langmuir (L), Freundlich (F) and Temkin (Te) models) and



**Fig. 4.** A typical isotherm model by non-linear fitting for pure CO<sub>2</sub> flow with a surface aminated activated carbon.

3-parameter ones (Sips (S), Toth (T) and Redlich-Peterson (R) models), as could be seen in Fig. 4.

Concerning the theoretical  $q_{\max}$  values, only the Redlich-Peterson model was close to the experimental values. Meanwhile, Sips and Toth models, which have been reported in some other studies to exhibit a high degree of fitness with gas adsorption showed conformity to a reasonable extent (Adelodun *et al.*, 2016). Freundlich is the most reliable two-parameter model to describe the adsorption of CO<sub>2</sub> on either pristine or modified AC, while Sips and Redlich-Peterson models are better expressions, probably due to the extra parameter which improves their flexibility.

Therefore, the adsorption isotherm study confirms the presence of various SNGs (surface nitrogen groups) heterogeneously distributed on the modified AC. The process favors Freundlich and Langmuir isotherm at low or high CO<sub>2</sub> concentrations, respectively (Lim *et al.*, 2016).

## 7. CONCLUSIONS AND RECOMMENDATION

Despite the impressive adsorption capacities exhibited by activated carbons (ACs) and activated carbon fibers (ACFs), their selectivity toward indoor-level CO<sub>2</sub> is poor. For this reason, chemical modification with various organic and inorganic dopants have been used to basify their surfaces, especially in the past two decades. Since the present review focuses on the field available process, high selectivity and capacity materials that are costly and imperfective molecular organic frames (MOFs) or novel mesoporous zeolitic structure were not considered.

We found that amination and ammoxidation offer varying degrees of success with improving both the surface chemistry and textural properties. In addition, some alkali materials such as glycine, urea, melamine and tetraethyleneamine etc. have been explored to modify the adsorbent surface including pore channels. In particular, the selective adsorption for low level CO<sub>2</sub> capture could be improved by alkalization with concentrated KOH.

Another carbonaceous adsorbent, ACF, is also useful in forming various functional groups suitable for CO<sub>2</sub> molecules, because it can be designed to meet the specific purposes from the initial spinning step. The isotherm studies for this diluted CO<sub>2</sub> capture seemed to result in Freundlich and Langmuir model implying simple gas adsorption.

The feasibility and performance evaluation for the real-life application of established CO<sub>2</sub> adsorbents should be aimed from the view of practical environment. Based on the few empirical studies currently available on the CO<sub>2</sub> adsorption from indoor spaces, the use of readily available materials such as waste, biomass, etc. for low-cost adsorbent preparation should be propagated to minimize regeneration cost. Process design studies including reactors further should continue with a focus on improving Indoor Air Quality (IAQ).

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