Biowaste-Based Porous Adsorbent for Carbon Dioxide Adsorption

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Abstract. Activated carbon is known as one of the effective adsorbents in carbon dioxide adsorption, which is mainly attributed to its highly porous structure. Utilizing biowaste as the precursor for the activated carbon synthesis is considered as an attractive approach due to the biowaste being an inexpensive, renewable, abundant, and environmental-friendly material. This review article covers the various biowaste precursors used for activated carbon synthesis along with the comparison of its activating methods (which are physical and chemical activation). The relation between the activation methods and carbon dioxide adsorption performance is elucidated for better understanding. The functionalization of the biowaste-based activated carbon with basic nitrogen functionalizing agent as well as the challenges in the application of these adsorbents in carbon dioxide adsorption are also discussed.

1. Introduction
Carbon dioxide (CO₂) is renowned as one of the major greenhouse gases that can directly contribute to the notorious climate change phenomena. The occurrence of climate change can result to unwanted environmental effects such as rising sea level, flood, and ocean storms. To mitigate these issues, the emission of carbon dioxide needs to be reduced through an established strategy called “Carbon Dioxide Capture and Storage (CCS)”. This strategy is mostly integrated in power plant as 86% of the total CO₂ emission originated from fossil fuel burning in power plant [1]. The implementation of CCS in power plant involves capturing CO₂ from the industrial flue gas before it gets released to the atmosphere. The typical flue gas compositions from post-combustion in power plant is 15-16 % CO₂, 5-7 % H₂O, 3-4 %O₂, and 73-77 %N₂ [2]. Amine-based liquid absorption is apparently the only commercial method used in industry to capture CO₂. Although it is widely used in industry nowadays, there are drawbacks in utilizing this method which includes being corrosive to the equipment as well as being expensive due to the substantial energy required for the regeneration process[3].

Adsorption is currently one of the most researched methods in replacing the amine-based liquid absorption due to its simplicity, low-cost, highly effective, and non-corrosive to the equipment. One of the most promising adsorbents for CO₂ adsorption is activated carbon due to it being inexpensive, water-resistant, and merely requires low regeneration energy. Moreover, it is also known to possess...
excellent adsorption capability because of its highly porous structure[4]. Even though common commercial activated carbon is readily available to be utilized as an adsorbent, it is still considered as a very expensive material. Therefore, an alternative method which involves synthesizing activated carbon from a cheap precursor is recommended. Various low-cost materials have been extensively researched in the past for the precursor role in synthesis of activated carbon. Biowaste is considered as one of the most researched low-cost materials as a precursor due to its abundancy in which its utilization would certainly help to reduce the waste disposal in landfills. Therefore, the main aim of this article is to review the biowaste-based activated carbons as adsorbents for CO₂ adsorption application.

2. Bio Waste-based Activated Carbon

Biowaste is considered as a discarded, unused and low-valued organic material. It is mostly comprised of agricultural, industrial and food wastes. Besides biowaste being an inexpensive, abundant and environmental-friendly material, its high carbon and low ash composition are other major reasons it is considered as a promising material for a precursor role in the synthesis of activated carbon. High carbon and low ash composition are necessary to attain a high yield activated carbon for future industrial application. The synthesizing route of biowaste-activated carbon usually involves two steps – which are carbonization and activation. The first step (carbonization) is a process that requires a heating medium (inert gas) to heat the precursor at a high temperature until the biochar is produced. The requirement of the first step is to ensure the elimination of impurities, moisture and volatiles from the precursor [5]. The second step (activation) is the process that is responsible in creating the pores of the activated carbon by using an activation agent. There are also a few researches that carried out a mere single-step method which combines carbonization and activation in one-step. The activation method is categorized into two methods (physical and chemical), in which the following sections will discuss more on the activation methods along with its comparison on the CO₂ adsorption performance.

2.1. Physical Activation

Activated carbon synthesis using physical activation is an activation method that does not involve a chemical agent being utilized in the process. The physical activation methods that were used in previous studies are air, steam, and CO₂ activation. This activation method generates pores by diffusing an activating gas (air, steam or carbon dioxide) into the biochar, where an oxidation reaction of the gas with the biochar will develop the intended pores in the adsorbent.

For steam activation, Erto et al.,[6] investigated the activated carbon synthesized from apricot stone, peach stone and olive stone for CO₂ adsorption at 25°C and 0.15 bar. From this research, it was found that the CO₂ adsorption capacity of apricot stone (0.75 mmol/g) is the highest, followed by peach stone (0.65 mmol/g) and olive stone (0.50 mmol/g) [6]. Although the surface area of apricot stone (830 m²/g) is lower than olive stone (910 m²/g), apricot stone still showcased its superior CO₂ adsorption capacity towards olive stone due to the higher fraction of micropore volumes existed on the adsorbent, which is calculated at around 72% of the total pore volume [6]. This indicates CO₂ adsorption of an adsorbent is dependable on the type of pores that are dominant in the adsorbent, in which for this case is micropore. Micropore (pore diameter <2nm) seems to be the highest-functioning pore in adsorbing CO₂ due to the overlapping inter-particle interactions of the pore walls of the micropores with the CO₂ molecules, which leads to the retention of the CO₂ molecules inside the pores [7].

CO₂ activation is another physical activation method that has proven to generate abundant micropores for CO₂ adsorption. For instance, activated carbon synthesized from sugarcane bagasse through CO₂ activation attained a considerably high CO₂ adsorption capacity of 2.55 mmol/g at ambient condition due to a majority of the developed pores are micropores (79% of the total pore volume) [8]. In fact, its CO₂ adsorption capacity value of 1.12 mmol/g under post-combustion flue gas condition (60°C and 0.1 bar) was found to be even higher than the aforementioned biowaste flue gas from steam activation. A comparison made between CO₂ activation and air activation of
sugarcane bagasse-derived activated carbon has also validated that CO₂ activation gas produces a more effective activated carbon as opposed to utilizing air as the activation gas [8]. The reasoning for this is obviously due to the higher volume of pores generated in the CO₂-activated carbon compared to the air-activated carbon.

Another high CO₂ adsorption capacity value was acquired from activated carbon synthesized from rice husk (3.1 mmol/g at ambient condition) – in which its high adsorption capacity is attributed to both micropores and mesopores created through CO₂ activation[9]. The development of mesopores (pore diameter between 2 and 50 nm) on the adsorbent played a role in increasing the CO₂ diffusion speed into the pores by reducing its pathway resistance. On the other hand, a slightly lower CO₂ adsorption capacity was obtained from walnut shell-derived activated carbon (1.58 mmol/g) at ambient condition compared to other biowaste-based activated carbon synthesized from CO₂ activation [10]. This is probably due to the lower activation temperature used in the activating process that causes an incomplete activation of the adsorbent.

Table 1 is the summary on the previous studies of the biowaste-based activated carbon synthesized via physical activation for CO₂ adsorption. Among all of the physical activating methods, it is inferred that CO₂ activation is the most effective physical activation method due to the pores (micropores and mesopores) that are more developed on the adsorbent compared to other activation methods, where its CO₂ adsorption capacity in the range of 1.5 to 3 mmol/g is attained at ambient condition.

| Precursor          | Activation method | Activation temperature (°C) | Surface area (m²/g) | Total pore volume (cm³/g) | Micropore volume (cm³/g) | Adsorption condition | CO₂ adsorption capacity (mmol/g) | Reference |
|--------------------|-------------------|-----------------------------|---------------------|---------------------------|--------------------------|----------------------|----------------------------------|-----------|
| Apricot stone      | Steam             | 800                         | 830                 | 0.500                     | 0.360                    | 25°C, 0.15 bar         | 0.75                             | [6]       |
| Peach stone        | Steam             | 800                         | 820                 | 0.410                     | 0.278                    | 25°C, 0.15 bar         | 0.65                             | [6]       |
| Olive stone        | Steam             | 800                         | 910                 | 0.630                     | 0.335                    | 25°C, 0.15 bar         | 0.50                             | [6]       |
| Sugarcane bagasse  | Air               | 850                         | 99                  | 0.070                     | 0.030                    | 25°C, 1 bar            | 1.61                             | [8]       |
|                    | CO₂               | 850                         | 622                 | 0.380                     | 0.300                    | 60°C, 0.1 bar          | 0.74                             | [8]       |
|                    |                   |                             |                     |                           |                          | 25°C, 1 bar            | 2.55                             | [8]       |
| Date seeds         | CO₂              | 900                         | 798.38              | -                         | 0.280                    | 25°C, 1 bar            | 2.94                             | [11]      |
| Rice husk          | CO₂              | 900                         | 1097                | -                         | 0.340                    | 40°C, 0.1 bar          | 1.62                             | [9]       |
| Walnut shell       | CO₂              | 500                         | 810.85              | 0.340                     | -                        | 20°C, 1 bar            | 1.58                             | [10]      |
| Palm kernel shell  | CO₂              | 850                         | 423.9               | 0.220                     | -                        | 25°C, 1 bar            | 2.13                             | [12]      |

2.2. Chemical Activation

Chemical activation is an activation method that uses a chemical activation agent to react with the carbon element for promoting pore formation in the adsorbent. The chemical activation method involves the impregnation of a carbon source with the chemical activation agent, followed by heating the mixture at a high activation temperature in an inert condition. Some of the chemical activation agents that have been used in the past for activated carbon synthesis are phosphoric acid (H₃PO₄), sodium hydroxide (NaOH), potassium hydroxide (KOH), and potassium carbonate(K₂CO₃). For H₃PO₄ activation, the acid is used to catalyze the bond cleavage reaction and form crosslinks in the adsorbent, where formation of phosphate and polyphosphate groups promotes pores generation inside the adsorbent [13]. Whereas, for alkali metal hydroxide (NaOH and KOH) activation, it is understood that its porosity development is acquired through gasification reaction of the elemental carbon, the redox reaction of the alkali metal hydroxide with the elemental carbon as well as the intercalation of the alkali metal into the carbon matrix obtained from the reduction of its alkali metal hydroxides [14]. K₂CO₃ activation is similar with KOH activation in the aspect where the development of pores in K₂CO₃ activation is also attributed to the gasification reaction, redox reaction and intercalation of
potassium metal in the carbon matrix [15]. The previous studies of biowaste-based activated carbon synthesized via chemical activation for CO$_2$ adsorption can be referred from Table 2.

| Precursor                  | Activation method | Activation parameter | Surface area (m$^2$/g) | Total pore volume (cm$^3$/g) | Micropore volume (cm$^3$/g) | Adsorption condition | Adsorption capacity (mmol/g) | Reference |
|----------------------------|-------------------|----------------------|-------------------------|-----------------------------|-----------------------------|-----------------------|-----------------------------|-----------|
| Sugarcane bagasse          | H$_2$PO$_4$       | -                    | 873                     | 1.06                        | 0.18                        | 25°C, 1 bar           | 6.22                         | [8]       |
|                            | NaOH              | -                    | 1149                    | 1.73                        | 0.08                        | 25°C, 1 bar, 60°C, 0.1 bar | 4.28                         | [8]       |
| Argan fruit shell          | NaOH              | 850                  | 1827                    | 0.96                        | 0.73                        | 25°C, 1 bar           | 3.73                         | [5]       |
|                            | KOH               | 850                  | 1890                    | 0.87                        | 0.80                        | 25°C, 1 bar           | 5.63                         | [5]       |
| Yellow mombin fruit stone  | KOH               | 500                  | 1384                    | 0.63                        | 0.50                        | 25°C, 1 bar, 50°C, 1 bar | 7.3                          | [16]      |
| Pine cone shell            | KOH               | 650                  | 3135                    | -                           | -                           | 25°C, 1 bar           | 4.73                         | [17]      |
| Pine sawdust               | KOH               | 700                  | 1729                    | 0.70                        | 0.67                        | 25°C, 1 bar           | 4.21                         | [18]      |
| Spent coffee ground        | KOH               | 700                  | 1624                    | 0.66                        | 0.59                        | 25°C, 1 bar, 50°C, 1 bar | 4.42                         | [19]      |
| Sunflower seed shell       | KOH               | 700                  | 1790                    | 0.77                        | -                           | 25°C, 1 bar, 25°C, 0.15 bar | 4.61                         | [20]      |
| Peanut shell               | KOH               | 700                  | 956                     | 0.43                        | -                           | 25°C, 1 bar, 25°C, 0.15 bar | 4.03                         | [20]      |
| Garlic peel                | KOH               | 700                  | 947                     | 0.51                        | 0.50                        | 25°C, 1 bar           | 4.22                         | [21]      |
| Carrot peels               | KOH               | 700                  | 1379                    | 0.58                        | 0.51                        | 25°C, 1 bar           | 4.18                         | [22]      |
| Pomegranate peels          | KOH               | 700                  | 585                     | 0.28                        | 0.20                        | 25°C, 1 bar           | 4.11                         | [22]      |
| Cashew nutshell            | K$_2$CO$_3$       | 850                  | 1225                    | 0.66                        | 0.36                        | 0°C, 1 bar            | 6.22                         | [15]      |

A recent research investigated the potentiality of a sugarcane bagasse-based activated carbon for CO$_2$ adsorption that was synthesized from various activation methods including H$_2$PO$_4$ activation and NaOH activation [8]. It is exhibited from Table 1 that NaOH activation of sugarcane bagasse reached a higher CO$_2$ adsorption capacity than H$_2$PO$_4$ activation. This is due to a larger pore volume generated through NaOH activation where substantial pores are available for CO$_2$ diffusion. Regarding alkali metal hydroxide activation, Boujibar et al.,[5] carried out a comparison between KOH activation and NaOH activation method using argan fruit shell as the precursor, where it was found that KOH activation is a more effective method in producing narrow micropores (<0.7nm) for CO$_2$ adsorption. The reason for this is probably due to the ability of potassium metal to intercalate better into the material compared to a sodium metal, thus developing more pores inside the activated carbon [14].

KOH activation is apparently the most frequently studied activation method for activated carbon synthesis due to its ability in forming large quantity of micropores inside the activated carbon. This is evident from the relatively high CO$_2$ uptakes at ambient condition recorded for KOH-activated carbons from biowaste precursors such as peanut shell (4.03mmol/g) [20], pine sawdust (4.21 mmol/g) [18], spent coffee ground (4.42 mmol/g) [19], pine cone shell (4.61 mmol/g) [17], argan fruit shell (5.63 mmol/g) [5] and yellow mombin fruit stone (7.3mmol/g) [16]. An impressive CO$_2$ adsorption capacity is observed from the yellow mombin fruit stone due to a an immense narrow micropores (0.85nm-1.0nm) developed inside the activated carbon, where it represented 89% of the total pore volume [16]. In replacement for KOH activation, K$_2$CO$_3$ activation of cashew nutshell was investigated for an environmental-friendly activating agent alternative where it attained a moderately high adsorption capacity of 6.22mmol/g at 0°C and 1 bar[15]. Although the pore development
mechanism of KOH and K$_2$CO$_3$ activation are similar, it is reported that KOH has a rather stronger etching effect for pore creation than K$_2$CO$_3$ activation [23].

For achieving an optimal CO$_2$ adsorption performance, it is necessary for the activation parameters to be optimized. Activation parameters including the activation temperature and activating agent impregnation ratio (agent:carbon source) play a vital role in attaining the highest adsorption capacity. For instance, the optimal CO$_2$ adsorption capacity of a pine sawdust-based activated carbon (4.21mmol/g) at ambient condition was reached at an activation temperature of 750°C, whereby a further increase in the activation temperature to 800°C leads to a reduction in the CO$_2$ adsorption capacity (3.46 mmol/g)[18]. The reason for the decrease in adsorption capacity beyond the optimal activation temperature is because of the pore collapse at an extreme high activation temperature. A similar trend is also observed in the KOH impregnation ratio of a peanut shell, in which the increase in impregnation ratio from 1:1 to 1.5:1 causes a pore collapse in the activated carbon, and consequently reducing its CO$_2$ uptake of the adsorbent [20].

From the numerous previous researches, it is ostensible that chemical activation method is a preferential method over physical activation method due to the higher CO$_2$ adsorption capacity registered for biowaste-activated carbon synthesized via chemical activation. Amongst the chemical activation method, KOH activation is considered as the most common and effective activation method to synthesize biowaste-based activated carbon at ambient condition attributing to its ability to generate abundant pores, specifically narrow micropores (<1nm) for CO$_2$ adsorption.

### 3. Nitrogen functionalization of biowaste-based activated carbon

Besides acquiring abundant pores in the adsorbent, it is also pertinent for the surface of the adsorbent to be highly basic in order to produce an effective adsorbent for CO$_2$ adsorption. Increasing the surface basicity of the adsorbent will enhance its CO$_2$ adsorption performance through the chemical reaction between the basic functional group of the adsorbent with the acidic CO$_2$ molecules. This approach is especially beneficial in adsorbing CO$_2$ from a simulated post combustion flue gas condition, in which the flue gas mixture is generally at a high temperature (40-127°C) and low CO$_2$ partial pressure condition (0.08-0.16 bar) [2][24]. One of the most researched methods in increasing the surface basicity of the adsorbents is functionalizing the adsorbent’s surface with basic nitrogen functional groups obtained from a nitrogen-rich source. These functionalized nitrogen groups on the adsorbent will adsorb CO$_2$ by acting as the adsorption site to selectively bind with the acidic CO$_2$ molecule. The past studies of various nitrogen functionalizing agent used on the biowaste-based activated carbon for CO$_2$ adsorption application can be referred from Table 3.

| Precursor          | Activation agent | Nitrogen functionalizing agent | Surface Area (m$^2$/g) | Micropore volume (cm$^3$/g) | Nitrogen content (wt%) | Adsorption condition | Adsorption capacity (mmol/g) | Reference |
|--------------------|------------------|--------------------------------|-------------------------|----------------------------|------------------------|-----------------------|---------------------------|-----------|
| Coconut shell      | KOH              | Urea                           | 1535                    | 0.56                       | 0.91                   | 25°C, 1 bar           | 4.80                      | [25]      |
| Walnut shell       | KOH              | Urea                           | 2354                    | 0.97                       | 0.86                   | 25°C, 1 bar           | 3.08                      | [23]      |
| Broom sorghum stalk | KOH and H$_2$PO$_4$ | Urea and TETA                 | 614                     | 0.274                      | -                      | 25°C, 1 bar           | 2.33                      | [27]      |
| Bagasse            | ZnCl$_2$ and KOH | TEPA and TETA                  | 1235                    | -                          | -                      | 60°C, 0.15 bar        | 6.25                      | [28]      |
| Waste tea          | KOH              | EDA                            | 11.80                   | 10.15                      | 25°C, 1 bar            | 2.48                  | [29]                      |
| Palm Shell         | Steam            | MEA and DEA                    | 392.4                   | -                          | 4.54                   | 70°C, 1 bar           | 2.36                      | [30]      |
| Pine cone          | H$_2$PO$_4$      | Polyaniline                    | 1279                    | 0.524                      | 4.46                   | 25°C, 1 bar           | 3.96                      | [31]      |
| Rice husk          | H$_2$PO$_4$      | Melamine                       | 1163                    | -                          | 4.90                   | 75°C, 0.1 bar         | 3.56                      | [32]      |
According to a myriad of previous studies represented in Table 3, urea is obviously one of the often-used nitrogen-functionalizing agent on biowaste-based activated carbon for CO\(_2\) adsorption. Both coconut shell [25] [26] and walnut shell [23] were functionalized with nitrogen functional group in a similar method where biowaste and urea are mixed together under a heated gas before the activation process. The CO\(_2\) uptake achieved for both walnut shell and coconut shell at ambient condition are comparable with the previous biowaste-based activated carbon synthesized via chemical activation without urea modification. The high nitrogen content expected for the activated carbon was not acquired for the activated carbon due to its decomposition during the activation process [25]. This indicates that the adsorption mechanism of the coconut shell and walnut shell-based activated carbon are mainly governed by physical adsorption attributed to its highly microporous structure with minimal contribution from the nitrogen functionalizing agent. However, it was surmised by Chen et al., [26] that the nitrogen presented in the carbon structure had helped in creating more micropores instead due to its decomposition. On the other hand, the broom sorghum stalk grafted with urea after activation process is able to adsorb CO\(_2\) as a result of the reaction between the functionalized nitrogen on the adsorbent with CO\(_2\) molecule. Its porosity has a lesser effect on the CO\(_2\) adsorption because of the resultant blocked pores caused by urea grafting [27].

Modifying the surface with amine is another common method to introduce the nitrogen functional group onto the adsorbent. Some of the amine functionalizing agent that have been used in the past for biowaste-based activated carbons are monoethanolamine (MEA), diethanolamine (DEA), ethylenediamine (EDA), triethylenetetramine (TETA) and tetraethylenepentamine (TEPA). The broom sorghum stalk-based activated carbon grafted with triethylenetetramine (TETA) showcased a higher CO\(_2\) adsorption capacity value of 3.22 mmol/g at ambient condition than the urea-grafted adsorbent due to its higher surface area and micropore volume [27]. The DEA impregnation of palm shell-based activated carbon enhanced the CO\(_2\) uptake of the unmodified-activated carbon by 3 times due to the higher acquired nitrogen content after impregnation [30]. At a simulated post-combustion flue gas condition (60\(^\circ\)C and 0.15 bar), the bagasse-derived activated carbon impregnated with TETA and TEPA reached an excellent CO\(_2\) adsorption capacity value of 3.62 mmol/g and 3.49 mmol/g respectively, which are among the highest for biowaste-based activated carbon [28]. These values are noticeably higher that the unmodified sugarcane bagasse-derived activated carbon (1.31 mmol/g) at the same simulated post-combustion flue gas condition [8]. Additionally, rice husk-derived activated carbon functionalized with melamine has also attained a relatively high CO\(_2\) adsorption capacity (3.56 mmol/g) at a post combustion flue gas condition (70\(^\circ\)C and 0.1 bar) attributing to its basicity and porosity [32].

From these previous studies on nitrogen functionalization of biowaste-based activated carbon, it can be inferred that nitrogen functionalization method does enhance the carbon dioxide uptake of biowaste-based activated carbon especially at a simulated post-combustion flue gas condition, where approximately 3–4 mmol/g of CO\(_2\) is adsorbed. However, this value is still considered relatively low in comparison to the amine-functionalized mesoporous silica adsorbents that adsorb 7.79 mmol/g CO\(_2\) at simulated post-combustion flue gas condition [33] [34].

In the aspect of the nitrogen functionalizing method, there is apparently a dire issue in acquiring an optimal balance between porosity and basicity of the adsorbent, in which the porosity is usually cancelled out when the nitrogen groups are functionalized after the activation process due to the blocked pores from nitrogen-functionalizing agent. At the same time, the surface basicity can also be cancelled out if the nitrogen group is functionalized before the activation process due to the decomposition of nitrogen during activation. Hence, an improvement in this aspect is necessary to achieve an optimal CO\(_2\) capacity of an adsorbent.

4. Conclusion
In conclusion, biowaste-based activated carbon in general has shown to be an effective adsorbent in capturing CO\(_2\), with chemical activation being a better activation method to synthesize the adsorbent attributing to its higher micropore generation in the carbon matrix. However, its application in post-
combustion flue condition (high temperature and low CO₂ partial pressure) is still limited due to its relatively low CO₂ uptake at this condition. Therefore, an improvement in its surface basicity without compromising its porosity is recommended in future researches to further enhance its CO₂ adsorption performance at a post-combustion flue gas condition.

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