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# Biomass Valorization to Produce Porous Carbons: Applications in $CO_2$ Capture and Biogas Upgrading to Biomethane—A Mini-Review

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Porous carbon materials, derived from biomass wastes and/or as by-products, are considered versatile, economical and environmentally sustainable. Recently, their high adsorption capacity has led to an increased interest in several environmental applications related to separation/purification both in liquid- and gas-phases. Specifically, their use in carbon dioxide ($CO_2$) capture/sequestration has been a hot topic in the framework of gas adsorption applications. Cost effective biomass porous carbons with enhanced textural properties and high $CO_2$ uptakes present themselves as attractive alternative adsorbents with potential to be used in $CO_2$ capture/separation, apart from zeolites, commercial activated carbons and metal-organic frameworks (MOFs). The renewable and sustainable character of the precursor of these bioadsorbents must be highlighted in the context of a circular-economy and emergent renewable energy market to reach the EU climate and energy goals. This mini-review summarizes the current understandings and discussions about the development of porous carbons derived from bio-wastes, focusing their application to capture $CO_2$ and upgrade biogas to biomethane by adsorption-based processes. Biogas is composed by 55–65 v/v% of methane ($CH_4$) mainly in 35–45 v/v% of $CO_2$. The biogas upgraded to bio-$CH_4$ (97%v/v) through an adsorption process yields after proper conditioning to high quality biomethane and replaces natural gas of fossil source. The circular-economy impact of bio-$CH_4$ production is further enhanced by the use of biomass-derived porous carbons employed in the production process.

Keywords: biomass, porous carbons, adsorption, biogas upgrading, pressure swing adsorption, biomethane, $CH_4$, $CO_2$

## INTRODUCTION

Biogas produced from several biomasses is a renewable fuel source of methane that after proper conditioning can be injected into the gas pipeline networks or used as energy for transport (Scarlat et al., 2018; Ferella et al., 2019). However, $CO_2$ removal from biogas is a critical step for biogas upgrading and limits its application. Upgrading technologies comprise absorption, adsorption, membrane permeation, and cryogenic strategies (Zhou et al., 2017; Kapoor et al., 2019). Among them, adsorption has attracted attention due to its environmentally friendly nature, low energy demand and capital costs (Surra et al., 2019a; Lombardi and Francini, 2020). The key element in

adsorption-based processes is the need for an effective adsorbent with high adsorption capacities for the species to capture, low productions cost and low environmental impacts. Specific surface area and pore volume are the critical properties for these materials. Recent developments were achieved concerning adsorbents for $CO_2$ removal (Zhou et al., 2017), complemented with molecular simulations (Peng and Cao, 2013) to predict/produce the best materials (Esteves et al., 2008; Surra et al., 2019b; Bernardo et al., 2020; Liu et al., 2020b). Moreover, fresh overviews devoted to the state of the art of adsorbents for $CO_2$ removal have been focused on biomass derived porous carbons (Singh et al., 2019; Xu and Strømme, 2019; Sher et al., 2020). However, much of the research lacks the use of biomass-derived adsorbents employed themselves as key-materials to capture $CO_2$ and upgrade biogas to bio-$CH_4$ by adsorption-based processes (PSA), in a concept of a circular-economy (Cheng et al., 2020; Sherwood, 2020; D'Adamo et al., 2021; Kumar and Verma, 2021). Hence, this mini-review offers a succinct summary of this topic presenting the most recent literature (2020–2021) of $CO_2$ uptake with biomass-derived porous carbons and the last 5 years literature of biocarbons application in PSA technology for biogas upgrading. The existing gaps and potential future paths are also discussed.

## Biomass-Derived Porous Carbons

Converting the biomass into porous carbons involves carbonization (pyrolysis) and activation that can be of physical and/or chemical types. Physical-activation is a two-step process: the raw material is carbonized in the absence of $O_2$ at temperatures between 400–850°C, followed by activation of the resulting char with oxidant/gasifying gases like steam, air, $N_2$, $O_2$, $NH_3$, $CO_2$ or a mixture of these gases, at temperatures around 600–1000°C (Marsh and Rodríguez-Reinoso, 2006; González-García, 2018). The activation with $CO_2$ produces porous carbons with narrow micropore size distribution, providing optimum pore size for $CO_2/CH_4$ separation, while steam activation generates carbons with wider pore size distribution and smaller micropore volume (Marsh and Rodríguez-Reinoso, 2006). Nevertheless, in the physical-activation there is a poorer control of the porosity. The reactions during physical-activation can form surface oxygen functional groups, while activation with $NH_3$ adds N-containing groups on carbon surface; however, this is usually coupled with other gas to provide more porosity (Tan et al., 2017). The high temperatures usually used in physical-activation represent an energetic disadvantage.

Chemical-activation can be a one-step or two-step method since the impregnation with the activating chemical (dehydrating agents and/or oxidants) can be made directly in the biomass or in the resulting char from the first step of carbonization. After impregnation, the mixture of precursor and activating agent is heated under inert atmosphere at temperatures between 400–800°C (Marsh and Rodríguez-Reinoso, 2006; González-García, 2018). As physical-activation, chemical-activation provides porosity development and functional groups at carbon surface. Acids, alkalis, and salts like $H_3PO_4$, $H_2SO_4$, $ZnCl_2$, $K_2CO_3$, NaOH, and KOH are usually used in chemical-activations of biomass precursors (Yahya et al., 2015; González-García, 2018).

Chemical-activation usually provides biomass-derived porous carbons with high surface areas and a good control of the porosity, but washing the produced carbon to remove the residual activating agent present in the carbon matrix turns the process into a time- and energy-consuming one, and environmentally less friendly.

## Tailoring Carbon Surface Chemistry

When producing biomass-derived porous carbons, it is possible to tune their properties by an appropriate choice of the precursor and activation conditions. Nonetheless, the resulting material can be further tailored, specifically the surface chemistry properties to increase the $CO_2$ uptake capacity. Modification treatments can be envisaged to add and/or increase relevant functional groups on the surface of the carbon to enhance $CO_2$ retention. Increasing the basicity of the carbon is the most efficient way to improve the adsorption efficiency toward $CO_2$ uptake, namely through the removal of acidic functional groups from carbon surface and/or introduction of nitrogen groups that provide basic sites able to attract the acidic $CO_2$ (Adelodun et al., 2015; Rashidi and Yusup, 2016). Depending on the biomass precursor and if an acid is used as activating agent, several oxygen functional groups with strong or mild acidity might be present on the carbon surface. To remove these oxygenated groups, heat treatments under inert ($N_2$, Ar, He) or $H_2$ atmosphere are usually performed, but this requires very high temperatures (800–1000°C) (Shafeeyan et al., 2010).

Heat treatment with ammonia can be employed at temperatures between 200 and 1000°C (Rashidi and Yusup, 2016). Since carbon surface is typically non-reactive to $NH_3$, pre-oxidation of the surface is required prior to amination. After the treatment, the carbon surface is usually enriched with nitrogen functionalities like $-NH_2-$, $-CN$, pyridinic, pyrrolic, and quaternary $N_2$ (Shen and Fan, 2013). Nitrogen-rich biomass precursors, such as chitosan (Fujiki and Yogo, 2016), crab and prawn shell (Chen et al., 2015; Gao et al., 2016), and protein enriched biowaste (Huang et al., 2015; Shi et al., 2019), can directly provide porous carbons enriched with nitrogen functionalities.

Metal impregnation has been also employed to increase the carbon $CO_2$ uptake capacity and selectivity, although this strategy is not usual as nitrogen-doping. Metal oxides of alkaline-earth and transition elements can provide catalytic active sites able to interact with $CO_2$ although the presence of ultra microporosity remains an important feature.

**Table 1** presents the most recent works (2020–2021) dealing with biomass derived-porous carbons for $CO_2$ uptake. The biomass precursor, activation conditions, textural properties and $CO_2$ uptake capacity are shown. It should be highlighted that most of the studies use powder carbons, and static $CO_2$ adsorption measurements are performed volumetrically using gas sorption apparatus or through thermogravimetric analysers. Pure $CO_2$ or synthetic gas mixtures under controlled conditions are typically used. Dynamic $CO_2$ adsorption breakthrough tests and real gas samples are scarcely studied for the biomass-based adsorbents, although these conditions are closer to industrial

**TABLE 1 |** Recent literature (2020—2021) of $CO_2$ uptake with biomass-derived porous carbons.

| Biomass precursor | Activation conditions | $S_{BET}$ ($m^2/g$) | $V_{micro}$ ($cm^3/g$) | $CO_2$ uptake (mmol/g) | References |
|---|---|---|---|---|---|
| *Alligator* weed | KOH, 800°C, 2 h | 1779 | 0.60 | 6.4 (273 K, 1 bar) | (Singh et al., 2021) |
| Tobacco stem | KOH, 800°C, 2 h | 1922 | 0.79 | 7.9 (273 K, 1 bar) | (Ma et al., 2020c) |
| Peanut shell | KOH, 750°C, 1 h | 900 | 0.33 | 3.9 (298 K, 1 bar) | (Sher et al., 2020) |
| Palm sheath | $NaNH_2$, 550°C 1 h | 2181 | 0.65 | 5.8 (273 K, 1 bar) | (Liu et al., 2020a) |
| Citrus Aurantium leaves + Spirulina Platensis | $ZnCl_2$ + $CO_2$, 800°C, 2 h | 937 | 0.30 | 8.4 (273 K, 1 bar) | (Balou et al., 2020) |
| Acai fruit stone | KOH, 850°C, 1 h | 2612 | 0.84 | 6.0 (273 K, 1 bar) | (de Souza et al., 2020) |
|  | $CO_2$, 850°C, 8 h | 1150 | 0.39 | 3.0 (298 K, 1 bar) |  |
| Tobacco stem + urea | KOH, 600°C, 1 h | 2690 | 0.81 | 3.5 (298 K, 1 bar) | (Ma et al., 2020b) |
| Starch | $CO_2$, 900°C, 3 h | 1096 | 0.39 | 2.5 (298 K, 1 bar) | (Wu et al., 2020) |
| Garlic peel | KOH, 700°C, 1 h | 1248 | 0.52 | 5.1 (273 K, 1 bar) | (Huang et al., 2020) |
|  |  |  |  | 4.1 (298 K, 1 bar) |  |
| Oil residue | $NaNH_2$, 500°C 1 h | 2113 | 0.94 | 5.6 (273 K, 1 bar) | (Yang et al., 2020b) |
|  |  |  |  | 3.5 (298 K, 1 bar) |  |
| Pine sawdust | KOH, 700°C, 2 h | 1729 | 0.67 | 4.2 (298 K, 1 bar) | (Quan et al., 2020) |
| Banana sheets | KOH, 800°C, 1 h | 1988 | 0.67 | 5.3 (273 K, 1 bar) | (Li et al., 2020) |
|  |  |  |  | 4.2 (298 K, 1 bar) |  |
| Grape marc | KOH, 800°C, 2 h | 1727 | 0.59 | 6.7 (273 K, 1 bar) | (Ismail et al., 2020) |
|  |  |  |  | 3.9 (298 K, 1 bar) |  |
| *Lotus* stalk | KOH, 600°C | 1188 | 0.43 | 5.1 (273 K, 1 bar) | (Yang et al., 2020a) |
|  |  |  |  | 3.7 (298 K, 1 bar) |  |
| Olive mill waste | KOH, 700°C, 0.5 h | 1036 | 0.35 | 5.1 (273 K, 1 bar) | (González and Manyà, 2020) |
|  | $CO_2$, 850°C, 3 h | 1135 | 0.37 | 2.3 (273 K, 1 bar) |  |
| Sugarcane bagasse | Air, 850°C, 2 h | 99 | 0.03 | 1.6 (298 K, 1 bar) | (Guo et al., 2020) |
|  | $CO_2$, 850°C, 2 h | 622 | 0.30 | 2.6 (298 K, 1 bar) |  |
|  | $H_3PO_4$, 750°C, 1.5 h | 873 | 0.18 | 2.7 (298 K, 1 bar) |  |
|  | NaOH, 850°C, 1.5 h | 1149 | 0.08 | 4.3 (298 K, 1 bar) |  |
| Palm kernel shell + urea | $N_2$, 700°C, 0.5 h | 1700 | 0.56 | 5.3 (298 K, 1 bar) | (Ma et al., 2020a) |
| Walnut shell + urea | KOH, 850°C, 1 h | 2354 | 0.97 | 5.1 (273 K, 1 bar) | (Yang et al., 2020c) |
|  |  |  |  | 3.0 (298 K, 1 bar) |  |

applications. Also, dynamic adsorption/desorption/regeneration cycles allow a more efficient utilization of the adsorbent capacity; however, most of the published studies do not consider this important feature. Considering the lack of knowledge about the performance of biowastes derived porous carbons in continuous flow systems and their regeneration, it is of utmost importance that studies move forward to reflect realistic applications.

## Biogas Upgrading to Biomethane and $CO_2$ Capture

The selection of a biogas upgrading technology depends mainly on plant location, investment availability, biogas composition, bio-$CH_4$ aim quality, and plant productivity. Adsorption-based processes like Pressure-Swing Adsorption (PSA) is one of the most established know-how used in gas separation/purification/capture applications (Riboldi and Bolland, 2017). Its compacted set of fixed-bed adsorption columns, operating with pressure modulation in a cyclic mode, use the adsorbent(s) to selectively adsorb and desorb the undesired gases like $CO_2$ in biogas (Esteves, 2005; Esteves and Mota, 2007; Augelletti et al., 2017; Canevesi et al., 2018). The selective adsorption occurs due to different equilibrium capacities of the species (equilibrium adsorbent) or distinct gas uptake rates (kinetic adsorbent) in the adsorbent's surface. Activated carbons have demonstrated to perform effectively in relevant operating conditions, surpassing zeolites when $CO_2$ partial pressure overpasses a certain threshold

(ca. 1.7 bar) (Riboldi and Bolland, 2017). This means that what really matters is not the total $CO_2$ adsorption capacity of the solid adsorbent at a given pressure, but the pressure difference necessary to be applied between adsorption and desorption to obtain a satisfactory gas separation in PSA (adsorption isotherms). Carbons have higher adsorption capacity than zeolites at high pressure, but zeolites are regenerated by vacuum desorption with very small pressure variation. Carbon materials have the advantages of high thermal/chemical/mechanical/moisture stabilities, electrical and heat conductivities and reasonable cost. Their affinities for $CO_2$ can be improved with functionalization (Lee and Park, 2015). Achieve higher $CO_2$ selectivities requires the development of better adsorbents that, additionally, need to be easy regenerable for proper application in a PSA system that, otherwise, would require reduction in partial pressures or increase in the operating temperature. Recently, MOFs appeared as potential highly selective materials for gas separation/capture (Ferreira et al., 2019; Ribeiro et al., 2019), although they still need further research related with their cost, particle shaping, and stability (structural, mechanical, moisture, aging, etc.).

Upon this scenario, the use of porous biocarbons opens a double opportunity, in the sense that biomass turns to be a useful resource that can contribute, thereafter, to capture $CO_2$ and separate it from biogas, aiming to upgrade it to bio-$CH_4$ (Álvarez-Gutiérrez et al., 2014). This pathway closes the loop regarding carbon neutrality and contributes significantly to the

**TABLE 2 |** Last 5 years literature of biocarbons application in PSA technology for biogas upgrading.

| Biomass precursor | $S_{BET}$ (m$^2$/g) | $V_{micro}$ (cm$^3$/g) | Includes | $CO_2$ uptake (mol/kg) | References |
|---|---|---|---|---|---|
| Cherry stones | 1045.0 (CS-$CO_2$) | 0.40 | Dynamic column breakthrough measurements with 50/50 vol% $CO_2/CH_4$ feed mixture | 3.60 (CS-$CO_2$, 303 K, 5 bar) | (Álvarez-Gutiérrez et al., 2016) |
| | 998.0 (CS-$H_2O$) | 0.38 | | 3.53 (CS-$CO_2$, 303 K, 5 bar) | |
| Pine wood pellets | 561.0 | 0.22 | Breakthrough experiments with 60/40 vol% $CO_2/CH_4$ feed mixture | 2.14 (303 K, 1.5 bar) | (Vivo-Vilches et al., 2017) |
| Silver fir sawdust | 881.0 (HCA_200_0) | 0.24[a] | Breakthrough runs with 50/50 vol% $CO_2/CH_4$ and $CO_2/N_2$ feed mixture | 6.57 (in $N_2$, 5 bar) | (Gallucci et al., 2020) |
| | 284.0 (HCA_200_120) | 0.11[a] | | 3.64 (in $N_2$, 5 bar) | |
| Moso bamboo | 370.5 (BC 300) | n.a | Breakthrough and PSA experiments with 60/40 vol% $CO_2/CH_4$ feed mixture | 2.26[b] | (Seo et al., 2016) |
| | 419.9 (BC 500) | | | 2.70[b] | |
| | 494.0 (BC 900) | | | 2.76[b] | |
| | 288.1 (BC 1000) | | | 1.61[b] | |

[a]BJH Desorption Pore Volume ($V_{BJH}$, cm$^3$/g).
[b]At saturation.

field of renewable energy. In the last 5 years, when crossing PSA, biogas upgrading and activated carbon topics in Web of Science (WOS) Core Collection, one gets only 16 publications. Adding $CO_2$ capture topic to this search, only nine articles are found. From those papers, only four of them are about biomass-derived porous carbons applied to PSA. **Table 2** depicts those publications. The lack of research on bioadsorbents followed by their effective transfer to adsorption-based processes applied to biogas upgrading is still noticed in a sector so relevant to answer to the renewable energy and environmental concerns of the 21st century.

Despite the good potential demonstrated by porous carbons developed from cherry stones, pine wood pellets, silver fir sawdust and modo bamboo, the few studies published in the last 5 years emphasizes the need to gather more data on biomass-based activated carbons for biogas upgrading under operational conditions similar to real cases. Besides exhibiting high selectivity and optimal uptake capacity for $CO_2$, one of the features to be considered is to obtain porous carbons with the desired particle size to be directly packed in fixed bed columns. This avoids high pressure drops in columns that turn the process unfeasible. Some of these biomass derived adsorbents are obtained as powders and therefore their shaping into pellets, granules or spheres is another challenge for the near future.

## CONCLUSIONS AND POTENTIAL FUTURE DEVELOPMENTS

A succinct mini-review of current understandings about the development of porous carbons derived from bio-wastes, focusing their application to capture $CO_2$ and upgrade biogas to biomethane by adsorption-based processes (PSA) is carried out in this paper. Porous carbons derived from several biomass precursors as $CO_2$ adsorbents had a huge

development only in the last year of 2020, confirming that in fact this is a hot topic. However, the works were mainly directed to use pure $CO_2$ or synthetic mixtures in static experiments, neglecting the importance of studying the dynamic behavior of the new developed adsorbents as well as their performance in regeneration cycles. Further research in this field is highly recommended. Additional assessments on the effective use of biocarbons in adsorption processes need to be undertaken and explored, especially regarding their use in the inherent dynamic PSA process operation. Moreover, shaping of the porous carbons powders is a challenge to overcome, and the impact of that shaping procedure on the efficiency of the adsorbent must also be assessed. Finally, but not less important, an interesting concept that should be explored in this subject is an integrated techno-environmental evaluation of biomethane production from biogas (obtained by biocarbons in PSA), combined with $CO_2$ capture and storage (CCS), to check the potential for net negative greenhouse-gas emissions.

## AUTHOR CONTRIBUTIONS

MB (Conceptualization; Writing—Original Draft); NL and IF (Discussion; Writing—Review and Editing); IE (Conceptualization; Writing—Original draft; Funding acquisition).

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