Efficient nitrogen doped porous carbonaceous CO$_2$ adsorbents based on lotus leaf

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**A B S T R A C T**

In this work, the waste biomass lotus leaf was converted into N-doped porous carbonaceous CO$_2$ adsorbents. The synthesis process includes carbonization of lotus leaf, melamine post-treatment and KOH activation. For the resultant sorbents, high nitrogen content can be contained due to the melamine modification and advanced porous structure were formed by KOH etching. These samples were carefully characterized by different techniques and their CO$_2$ adsorption properties were investigated in detail. These sorbents hold good CO$_2$ adsorption abilities, up to 3.87 and 5.89 mmol/g at 25 and 0°C under 1 bar, respectively. By thorough investigation, the combined interplay of N content and narrow microporous volume was found to be responsible for the CO$_2$ uptake for this series of sorbents. Together with the high CO$_2$ adsorption abilities, these carbons also display excellent reversibility, high CO$_2$/N$_2$ selectivity, applicable heat of adsorption, fast CO$_2$ adsorption kinetics and good dynamic CO$_2$ adsorption capacity. This study reveals a universal method of obtaining N-doped porous carbonaceous sorbents from leaves. The low cost of raw materials accompanied by easy synthesis procedure disclose the enormous potential of leaves-based carbons in CO$_2$ capture as well as many other applications.

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**Introduction**

Since the industrial revolution, the global atmospheric CO$_2$ concentration has gradually increased due to the massive burning of fossil fuels, which is up to 415 ppmV by April 2020. If atmospheric CO$_2$ concentration is still increasing at this rate, irreparable harm to the climate and the environment would be produced. In order to cope with this situation, effective measures must be taken to reduce the CO$_2$ emissions. Among all kinds of strategies, carbon capture and storage (CCS) has been proposed as the most operative one to reduce CO$_2$ release and mitigate climate change (Guo et al., 2016). In recent years, some CCS-related technologies such as amine scrubbing (Rochelle, 2009), membrane separation (Yu et al., 2017) and ionic liquid absorption (Chen et al., 2016; Liu et al., 2018) have made great progress. Compared with the above technologies, adsorption thru solid sorbents is a potential alternative, which has the superiorities of low capital input, easy operation, high energy efficiency and free of corrosivity (Huang et al., 2016; Liu et al., 2019a; Peng et al., 2019b). The attainment of solid sorbents with superior CO$_2$ capture performance is of utmost importance.
capture capacities is the crucial for the effective implementation of this strategy. Thus, large number of researches have been focused on the fabricating porous materials with high CO₂ adsorption capacity such as metal-organic frameworks (MOFs) (Chen et al., 2019; Millward and Yaghi, 2005), zeolite (Bae et al., 2013; Dabbawala et al., 2020), porous carbon (An et al., 2019; Kamran and Park, 2020; Kou and Sun, 2016; Liu et al., 2020b) and porous polymer (Mame et al., 2018; Sun et al., 2015). Of these adsorbents, porous carbons attracted the special interest due to the merits of low cost, easy to make, ease to regeneration, tunable surface area and porosity, easy modification of surface functionalization and hydrophobicity etc. (Du et al., 2018, 2019; Yang et al., 2020). Owing to the highly developed porous structures, porous carbons exhibited good CO₂ adsorption performances at 25°C and ambient pressure. Such as, a petroleum coke derived porous carbon exhibited CO₂ uptake of 3.68 mmol/g (Jang et al., 2018).

While, porous carbonaceous adsorbents prepared from coconut shell possess the highest CO₂ adsorption ability of 4.23 mmol/g (Yang et al., 2017). Moreover, adsorption ability up to 5.0 mmol/g was achieved for the biomass-based porous carbon synthesized by Deng et al. (2014). Based on the previous literature, the significance of narrow micropores (< 1 nm) on the CO₂ uptake of the carbonaceous adsorbents under 1 bar and 25°C was constantly emphasized (Presser et al., 2011; Sevilla and Fuertes, 2011). Therefore, various strategies of improving narrow microporosity of carbonaceous adsorbents have been explored (Liu et al., 2020b; Yang et al., 2020). Besides enhancing the narrow microporosity of the sorbents, nitrogen incorporation into the carbon framework has also been proved as an effective method to increase the CO₂ adsorption abilities for porous carbons under ambient conditions (Hao et al., 2010; Xing et al., 2012; Yang et al., 2015). On this theme, excellent CO₂ uptake higher than 5.0 mmol/g were reported for several N-doped porous carbons derived from polymers (Sethia and Sanyari, 2015) or biomass materials (Adeniran and Mokaya, 2015). The synthesis of novel nitrogen-doped carbonaceous CO₂ adsorbent has become a hotspot in the field of CO₂ capture recently.

One advantage of carbonaceous adsorbents is their easy preparation from almost any raw materials containing carbon element. From the previous reports, various precursors have been utilized as starting materials to make porous carbons, such as coal (Arami-Niya et al., 2016), petroleum coke (Bai et al., 2015), polymer (Peng et al., 2019a) and biomass materials (Liu et al., 2020a; Rao et al., 2019) etc. Among all kinds of precursors, biomass materials attract the most attention from the standpoint of both economic and eco-environmental. On one hand, the inexpensive and abundant biomass precursors can decrease the massive production cost of carbonaceous adsorbents, which makes their practical application feasible. On the other hand, the massive pile-up of biomass materials might cause severe environmental problems. The proper disposal of these waste biomass materials can effectively avoid ecosystem imbalance. Thus, the conversion of biomass materials into excellent CO₂ adsorbents is very attractive in terms of both low-cost preparation of adsorbents and environmental protection.

When it comes to doping nitrogen into the carbon framework, post treatment method is the most explored and effective strategy. The direct reaction between carbon precursor and nitridation agent can efficiently incorporate large amount of N element into the carbon skeletons. Among various nitrogen-containing agents, melamine is cheap, abundant and contains high N content of 67%, which is an effective nitridation agent.

In this work, novel nitrogen-doped porous carbons were made employing lotus leaf as the starting materials, melamine as the nitrizing agent and KOH as the activator, respectively. Lotus leaf is a widely available biomass material, which is taken as a representative to explore the potential of leaves for the synthesis of N-doped porous carbons. To our best knowledge, there are very limited amount of literature on the utilization of various leaves for the production of carbonaceous CO₂ adsorbents (Liu et al., 2019b; Wang et al., 2012b; Zhu et al., 2015) and hence we believe that this investigation can provide new information in the field of both preparation of porous carbon and CO₂ capture.

In this work, the adsorbents’ synthesis employed a three-step process including carbonization of lotus leaf, post-nitriding by melamine and KOH activation. By changing two preparation parameters i.e. KOH quality and activation temperature, the porous textural and chemical properties of the adsorbents were adjusted. The most important factor that decides the CO₂ uptake was determined by detailed investigation.

1. Materials and methods

Lotus leaves were dried and broken into powders. Through sieving, particles with size of 74–150 μm were used as precursor to carry out carbonization. The carbonization process is performed under the protection of inert N₂ at 500°C for 1 hr. The resultant carbonized lotus leaf was named by LC. Next, N incorporation is executed by direct reaction of LC and melamine with a mass ratio of 1:1 at 500°C for 2 hr under N₂ stream. The resulting N-enhanced sample was branded by LCM. Subsequently, KOH activation process was carried out using LCM as the raw materials. During activation, activation temperature is set at 500, 550, 600, or 650°C and mass ratio of KOH to LCM of 1, 2 or 3 were chosen, respectively. The final adsorbents were assigned as LCM-X-Y, in which X and Y represents activation temperature and mass ratio of KOH to LCM, respectively.

The yield of these N-doped porous carbons ranges from 69% to 25%.

The detailed carbonization, post-nitridation and KOH activation processes as well as material characterization, sorbent performance test were recorded in Appendix A. Supplementary data.

2. Results and discussion

2.1. Porous properties

Fig. 1 shows the N₂ adsorption-desorption isotherms for these LCM-X-Y porous carbons. Significant N₂ adsorption arises at the low relative pressure (P/P₀ < 0.01) for all the sorbents,
Fig. 1 – N₂ sorption isotherms of the samples prepared at different conditions. Filled and empty symbols represent adsorption and desorption branches, respectively. LCM-X-Y: N-doped porous carbons synthesized from N-enhanced carbonized lotus leaf (LCM) at the activation temperature X and mass ratio KOH/LCM of Y. (a) Y = 1, (b) Y = 2, and (c) Y = 3. STP: standard temperature and pressure.

Fig. 2 – Pore size distribution of the samples prepared at different mass ratios of KOH/LCM (a) 1, (b) 2, and (c) 3. Dv(d): differential pore volume.
Table 1 - Porous textural, elemental compositions, and CO₂ uptakes of adsorbents derived from lotus leaf under different conditions.

<table>
<thead>
<tr>
<th>Sample</th>
<th>S_{BET} (m²/g)</th>
<th>V₀ (cm³/g)</th>
<th>V₁ (cm³/g)</th>
<th>Vₙ (cm³/g)</th>
<th>N (wt.%)</th>
<th>C (wt.%)</th>
<th>H (wt.%)</th>
<th>CO₂ uptake (mmol/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>25 °C</td>
</tr>
<tr>
<td>LC</td>
<td>3.82</td>
<td>0.02</td>
<td>–</td>
<td>0.13</td>
<td>1.55</td>
<td>64.37</td>
<td>2.57</td>
<td>1.00</td>
</tr>
<tr>
<td>LCM</td>
<td>4.32</td>
<td>0.02</td>
<td>–</td>
<td>–</td>
<td>19.68</td>
<td>63.38</td>
<td>2.14</td>
<td>0.46</td>
</tr>
<tr>
<td>LCM-500-1</td>
<td>687</td>
<td>0.34</td>
<td>0.24</td>
<td>0.33</td>
<td>7.69</td>
<td>67.35</td>
<td>2.69</td>
<td>3.21</td>
</tr>
<tr>
<td>LCM-500-2</td>
<td>1392</td>
<td>0.62</td>
<td>0.53</td>
<td>0.58</td>
<td>5.53</td>
<td>66.87</td>
<td>3.02</td>
<td>3.66</td>
</tr>
<tr>
<td>LCM-500-3</td>
<td>1491</td>
<td>0.67</td>
<td>0.56</td>
<td>0.63</td>
<td>3.97</td>
<td>67.02</td>
<td>2.91</td>
<td>3.49</td>
</tr>
<tr>
<td>LCM-550-1</td>
<td>1122</td>
<td>0.52</td>
<td>0.41</td>
<td>0.48</td>
<td>6.83</td>
<td>66.50</td>
<td>3.64</td>
<td>3.50</td>
</tr>
<tr>
<td>LCM-550-2</td>
<td>1487</td>
<td>0.69</td>
<td>0.54</td>
<td>0.61</td>
<td>4.09</td>
<td>68.15</td>
<td>4.11</td>
<td>3.87</td>
</tr>
<tr>
<td>LCM-550-3</td>
<td>2074</td>
<td>1.01</td>
<td>0.85</td>
<td>0.79</td>
<td>3.01</td>
<td>70.63</td>
<td>3.21</td>
<td>3.67</td>
</tr>
<tr>
<td>LCM-600-1</td>
<td>1272</td>
<td>0.64</td>
<td>0.51</td>
<td>0.57</td>
<td>4.97</td>
<td>66.15</td>
<td>2.47</td>
<td>3.50</td>
</tr>
<tr>
<td>LCM-600-2</td>
<td>2182</td>
<td>1.16</td>
<td>0.94</td>
<td>0.78</td>
<td>3.67</td>
<td>67.84</td>
<td>2.03</td>
<td>3.20</td>
</tr>
<tr>
<td>LCM-600-3</td>
<td>2340</td>
<td>1.35</td>
<td>1.16</td>
<td>0.81</td>
<td>2.54</td>
<td>71.18</td>
<td>1.73</td>
<td>2.94</td>
</tr>
<tr>
<td>LCM-650-1</td>
<td>1500</td>
<td>0.79</td>
<td>0.58</td>
<td>0.61</td>
<td>3.21</td>
<td>72.34</td>
<td>1.54</td>
<td>3.08</td>
</tr>
<tr>
<td>LCM-650-2</td>
<td>1860</td>
<td>1.11</td>
<td>0.75</td>
<td>0.65</td>
<td>2.29</td>
<td>74.63</td>
<td>1.13</td>
<td>2.76</td>
</tr>
<tr>
<td>LCM-650-3</td>
<td>2124</td>
<td>1.48</td>
<td>1.24</td>
<td>0.69</td>
<td>1.26</td>
<td>79.46</td>
<td>0.78</td>
<td>2.53</td>
</tr>
</tbody>
</table>

S_{BET}: surface area was calculated using the BET method at P/P₀ = 0.01–0.1; V₀: total pore volume at P/P₀ = 0.99; V₁: evaluated by the t-plot method; Vₙ: pore volume of narrow micropores (<1 nm) obtained from the CO₂ adsorption data at 0°C.

Fig. 3 – N 1s X-ray photoelectron spectroscopy (XPS) spectra of the samples (a) LCM, (b) LCM-550–2, (c) LCM-550–3 and (d) LCM-650–1. N-5: pyrrolic/pyridonic-N; N-6: pyridinic-N.

which suggests the presence of abundant micropores. With the further increasing of P/P₀, the adsorbents obtained under relatively low activation temperature (500 and 550°C) display near-linear adsorption characteristic. This performance matches to the monolayer and multilayer adsorption onto the available adsorption sites. Essentially, the adsorbents synthesized under 600 and 650°C but at KOH/CM ratio of 1 also presented the same adsorption feature. While, for the adsorbents LCM-X-Y (X = 600 or 650°C; Y = 2 or 3), a very wide knee was found at relative pressure ranging from 0.01 to 0.4, suggesting
the existence of certain amount of larger micropores. Commonly, these isotherms can be sorted as type I isotherm according to the International Union of Pure and Applied Chemistry (IUPAC) classification. However, it should be noted that small hysteresis exists for several samples, suggesting the presence of mesopores in the porous structures. The pore size distribution (PSD) curves of these adsorbents were demonstrated in Fig. 2, from which larger pore size and wider PSD can be detected with the increasing of both activation temperature and KOH dosage. All the textural characteristics obtained from the N₂ sorption measurement for these sorbents were summarized in Table 1. From the data of Table 1, both LC and LCM possess limited values in every textural characteristic i.e. BET surface area (S_BET), total pore volume (V₀), and micropore pore volume (Vₙ), indicating their non-porous nature. While, the activated samples have much developed porous structure, with the maximum S_BET, V₀, and V₁ of 2340 m²/g, 1.48 and 1.24 cm³/g, respectively. Although these adsorbents own high S_BET and V₀, it is well admitted that narrow micropore (< 1 nm) is the crucial to decide the CO₂ uptake under ambient conditions. Consequently, the volume of narrow micropores (Vₙ) of these sorbents need to be known, which is calculated by applying the Dubinin–Radushkevich (D–R) equation to the CO₂ adsorption data at 0°C. Table 1 includes the Vₙ values for these sorbents, which is between 0.33–0.81 cm³/g. The maximum Vₙ belongs to LCM-600-3, while higher temperature of 650°C leads to a decreased Vₙ, suggesting the negative effect of high temperature on the construction of narrow micropores.

Fig. 4 – Scanning electron microscopy (SEM) images of (a) LC, (b) LCM (c) and (d) LCM-550-2, (e) transmission electron microscopy (TEM) image and (f) X-ray diffraction (XRD) pattern of LCM-550-2.
2.2. Surface chemical, morphological and phase structural properties

From the results of elemental analysis, 64.37 wt.% C, 2.57 wt.% H and 1.55 wt.% N were found on LC. The N content increased to 19.68 wt.% for LCM after melamine treatment. This can prove that melamine is an excellent nitriding agent, and the post-treatment method can effectively dope nitrogen into the carbon material. The N content of the activated samples decreased compared with LCM, which decreased with the increasing KOH dosage and activation temperature. This agrees well with the previous studies (Bai et al., 2015; Yang et al., 2015). The N content for the samples synthesized under moderate activation can still keep 3–4 wt.% according to Table 1. While, under the harshest activation condition in this work, the resulting sample i.e. LCM-650-3 even possesses the less N content than the biomass precursor. On one hand, more nitrogen species will decompose at higher temperature. On the other hand, more N-containing functional groups will be consumed by reaction with more available KOH.

The nature of N species on the surface of these adsorbents were recognized by the X-ray photoelectron spectroscopy (XPS) measurement. As shown in Fig. 3a, XPS N 1s spectra of LCM, two peaks at binding energy of ca. 398.4 and 400.0 eV were found, which can be associated to pyridinic-N (N-6), and pyrrolic-/pyridonic-N (N-5), respectively. The same two peaks can be observed on the N 1s XPS spectra of the selected activated sorbents i.e. LCM-550-2, LCM-550-3 and LCM-650-1 (Fig. 3b-d). This finding indicates that there are no other N species such as oxidized nitrogen and quaternary-N in these activated sorbents. Moreover, it can be found that the quantity of N-6 is higher than that of N-5 for LCM, while for all N-doped porous carbons, the opposite results are observed. This seems to suggest that N-6 species has a higher reactivity with KOH than N-5, so more N-6 was consumed during the KOH activation. Higher N-5 content in these N-doped porous carbons may be more conducive to CO₂ adsorption, since it has been reported in some previous literatures that N-5 is more effective in CO₂ adsorption than N-6 (Ma et al., 2014; Sánchez-Sánchez et al., 2014; Sevilla et al., 2011).

Scanning electron microscopy (SEM) was employed to analyze the surface morphology of LC, LCM and a characteristic N-doped porous carbon LCM-550-2. As illustrated in Fig. 4a, LC shows a blocky morphology with smooth surface. There are no cavities or holes on its surface. After melamine treatment, LCM exhibits almost unchanged morphological features, only with the presence of some large pores on the surface (Fig. 4b). After further KOH activation, the surface of activated carbon LCM-550-2 shows obvious corrugations and unevenness (Fig. 4c), which is caused by the KOH etching. If the SEM image is further enlarged (Fig. 4d), a large number of small holes can be observed on the surface of LCM-550-2, indicating its highly advanced porous structure. TEM analysis was used to further characterize the morphological characteristics of LCM-550-2. As presented in Fig. 4e, copious randomly distributed vermicular microspheres were observed for LCM-550-2. This agrees well with the findings of N₂ sorption test, confirming its microporous structural feature.

Furthermore, X-ray diffraction (XRD) analysis was used to investigate the phase structure of LCM-550-2. As exhibited

![Fig. 5 - CO₂ adsorption isotherms at 25°C (empty symbols) and 0°C (filled symbols) for lotus leaf-based nitrogen-doped carbons prepared under different conditions. (a) Y = 1, (b) Y = 2, and (c) Y = 3.](image-url)
in Fig. 3f, one broad peak centered at $2\theta = 22^\circ$ and another much weaker peak at $2\theta = 43^\circ$ were observed. These two peaks are respectively associated with (002) and (100) diffractions of amorphous carbon (Liu et al., 2020a). This finding reveals the low crystallinity of the sample. The amorphous feature for these N-doped carbons is conducive for the generation of high levels of porosity, which is beneficial for the CO$_2$ uptake.

2.3. CO$_2$ adsorption analysis

The CO$_2$ adsorption isotherms of these lotus leaf-derived sorbents were tested at 25 and 0°C respectively. Fig. 5 displays the corresponding isotherms and Table 1 summarizes the CO$_2$ adsorption values. For all the sorbents at two temperatures, CO$_2$ uptake increase with increasing of pressure, of which half of the total CO$_2$ uptake occurs at less than 0.2 bar. This can be
ascribed to the CO₂ filling of the narrow micropores’ space at the low-pressure zone. With the further increasing of pressure, the increase in CO₂ adsorption capacity slows down for all the samples. Nevertheless, no sign of saturation in CO₂ uptake can be observed at 1 bar for these sorbents, suggesting that higher CO₂ adsorption capacity can be achieved at elevated pressure. As discovered in Table 1, CO₂ uptake range from 2.53–3.87 and 4.04–5.89 mmol/g were found for these sorbents at 25 and 0°C, respectively. While at 25°C and 0.15 bar (the CO₂ partial pressure in flue gas), the CO₂ uptake for these carbons ranges from 0.61 to 1.38 mmol/g. These values are still far behind some N-doped porous carbons derived from other biomass materials (Adeniran and Mokaya, 2015; Coromina et al., 2016; Deng et al., 2014; Sethia and Sayari, 2015). But they are similar to and even higher than numerous former described carbonaceous sorbents (Chen et al., 2018a, 2018b; Geng et al., 2017; Hao et al., 2011; Liu et al., 2012, 2011; Sevilla and Fuertes, 2012; Wang et al., 2012a, 2013) or other similar porous adsorbents such as COFs (Furukawa and Yaghi, 2009), ZIFs (Banerjee et al., 2009) and MOFs (Millward and Yaghi, 2005; Sumida et al., 2012). In Appendix A. Supplementary data, a Table lists and compares the CO₂ uptake for various sorbents at 25°C and 1 bar.

To obtain the main factors that decide the CO₂ uptake at 25°C and 1 bar, the CO₂ adsorption ability versus each porous textural characteristic as well as N content of these adsorbents was presented in Fig. 6. No direct correlation between CO₂ uptake and any single factor can be found, indicating that CO₂ adsorption capacity must not be determined solely by a single factor. From the previous literature, it is well recognized that CO₂ uptake can be improved by either enhancing the amount of narrow micropores (Deng et al., 2014; Wickramaratne and Jaroniec, 2013) or incorporating more N into the carbon framework (Hao et al., 2010; Xing et al., 2012). Since both factors are vital in determining the CO₂ uptake, it is possible that CO₂ uptake for these sorbents are determined by the combined interplay of narrow micropore volume and N content. The data provided in Table 1 can support this statement. Among all the adsorbents, LCM-500-1 has the highest N content but only possesses the medium CO₂ adsorption capacity. While, the highest Vₚ belongs to LCM-600-3, which shows even less CO₂ uptake than LCM-500-1. The low CO₂ adsorption ability for LCM-500-1 can be ascribed to its smallest Vₚ of all the adsorbents, while only 1.73 wt.% N content of LCM-600-3 can be responsible for its insufficient adsorption capacity. On the contrary, LCM-550-2 and LCM-500-2 have the first and third highest CO₂ uptake of all the samples, both of which only possess medium value of Vₚ and N content. The reason of higher CO₂ uptake of LCM-550-2 than LCM-500-2 could be attributed to the higher Vₚ in LCM-550-2, even though LCM-500-2 possesses the higher N content. In fact, when considering any pair of adsorbents in this work, their relative CO₂ adsorption capacity can be explained by comparing the value of nitrogen content and Vₚ. Thus, it is believed that the combined effects of both factors decide the CO₂ adsorption capacity for these adsorbents.

For the potential CO₂ capture application, the adsorbent recyclability must be considered. In order to test the recyclability, the best performance sample LCM-550-2 was degassed under 200°C before each CO₂ adsorption experiment for five continuous cycles at 25°C. As demonstrated in Fig. 7, only less than 4% loss with respect to the origin adsorption capacity can be found after the 5th cycle, confirming its stable recyclability.

Besides high CO₂ adsorption capacity and excellent recyclability, a good CO₂ sorbent should have high CO₂/N₂ selectivity as well. To obtain the CO₂/N₂ selectivity, the N₂ adsorption isotherm of LCM-550-2 was obtained under the identical conditions of CO₂ adsorption, which is illustrated in Fig. 8a. Ideal adsorbed solution theory (IAST) (Myers and Prausnitz, 1965) was applied to evaluate the CO₂/N₂ selectivity of LCM-550-2 on a CO₂–N₂ mixture (10:90 V/V) at 25°C and 1 bar. The CO₂/N₂ selectivity of LCM-550-2 reaches 20, which is higher than many carbonaceous sorbents reported formerly (Bai et al., 2015; Hao et al., 2011, 2013; Ma et al., 2014; Zhao et al., 2011; Zhong et al., 2012).

For a suitable CO₂ sorbent, the kinetics of adsorption should be fast. The CO₂ adsorption kinetic performance of LCM-550-2 was inspected at 25°C. As demonstrated in Fig. 8b, within roughly 3 min, 90% of the saturated adsorption capacity has been reached, signifying its fast CO₂ adsorption rate. On a practical view, the fast kinetic rate for lotus leaf-derived sorbents can shorten the adsorption cycle times and improve the working efficiency.

The isosteric heats of adsorption (Qₑₒₜ) is a critical parameter to describe the interactions between adsorbents and gas molecules, which can govern the energy penalty for adsorbent regeneration. Clausius-Clapeyron equation (Kaye and Long, 2005) was applied to calculate the Qₑₒₜ of an adsorbent based on the CO₂ adsorption data at 25 and 0°C. Fig. 8c shows the Qₑₒₜ vs. CO₂ loading amount curves for several chosen samples. At near zero loading, Qₑₒₜ of 22 to 26 kJ/mol were found for these sorbents. With the further increasing of CO₂ loading amount, Qₑₒₜ decreases to around 19 kJ/mol suggesting the surface heterogeneity for these adsorbents. Judging from the value of Qₑₒₜ shown here, physisorption occurs during the CO₂ adsorption process, which means the limited energy consumed during the regeneration of the spent adsorbents.
On top of all above excellent properties, an adsorbent should have high dynamic CO₂ capture capacity, since the flue gas flows in a continuous way during the realistic application. Thus, the dynamic CO₂ capture capacity of a potential adsorbent must be assessed. Herein, dynamic breakthrough experiment was carried out with a binary CO₂-N₂ mixture (10:90 V/V) at 25°C. From Fig. 8d, the breakthrough happens at ca. 5 min for LCM-550-2 under the tested conditions. By integration of the breakthrough curve, the dynamic CO₂ adsorption capacity attained 0.96 mmol/g, which indicates the high working capacity in the application of CO₂ capture.

3. Conclusions

In summary, waster biomass lotus leaf was converted into the highly effective N-doped porous carbonaceous CO₂ adsorbents. Due to the melamine modification and KOH activation, the as-obtained sorbents possess high N content and advanced porosity. High CO₂ uptake was found for this series of sorbents, up to 3.87 and 5.89 mmol/g at 25 and 0°C under 1 bar, respectively. Detailed investigation revealed that the combined interplay of N content and narrow microporous volume are responsible for the CO₂ adsorption abilities of these sorbents. In addition, other excellent CO₂ adsorption properties can be possessed by these sorbents, such as excellent reversibility, high CO₂/N₂ selectivity, applicable heat of adsorption, fast CO₂ adsorption kinetics and good dynamic CO₂ adsorption capacity. Herein, a universal strategy to make N-doped carbons was provided using biomass of leaves as the raw material. The low cost of precursor together with easy synthesis procedure make leaves-based carbons great promising in CO₂ capture and many other applications.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material associated with this article can be found in the online version at doi:10.1016/j.jes.2020.11.008.

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