Synthesis and Characterizations of Nanoporous Carbon Derived from Lapsi (Choerospondias axillaris) Seed: Effect of Carbonization Conditions

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Abstract

We report a facile synthesis of nanoporous activated carbons (NACs) derived from Lapsi (*Choerospondias axillaris*) seed powder (LSP). LSP and sodium hydroxide (NaOH) are **mixed in 1:1 ratio by wt. and** carbonized at 400 °C for different time (3, 4, and 5 hours) and at different temperature (400, 500, 600, and 700 °C) for 3 hours under a constant flow of ultrapure nitrogen gas. The effect of the conditions during the preparation on the structures and properties of NACs is investigated carefully. Fourier transform infrared spectroscopy (FT-IR) show the presence of several functional groups (*e.g.*, hydroxyl, carbonyl, and carboxyl groups) on the carbon surface. Irrespective of the carbonization conditions, both *D* and *G* bands are clearly observed in Raman scattering spectra. It is found that the increase in carbonization time significantly increases the G/D bands ratio, indicating an enhancement in the graphitic character. The NACs prepared by carbonization at 400 °C for 3 hours have the highest surface area (over 1000 m²·g⁻¹) and show high adsorption efficiency in removing methylene blue (MB) from aqueous solution (around 200 mg·g⁻¹).

Keywords: nanoporous material; nanoporous carbon; activated carbon; Lapsi (*Choerospondias Axillaris*) seed; carbonization; mesoporous material

1. Introduction

Porous materials have [been significantly developed because of](#) their fundamental [properties](#) and potential applications in many research areas including adsorption, storage, separation, and sensing.¹⁻⁶ [Porous materials have been classified according to their pore diameters; micropores are those with diameters less than 2 nm, mesopores range from 2 nm to 50 nm, and macropores are larger than 50 nm in diameter.](#) The growing impacts of industrial pollutants on the environment have led to the search for new relevant [treatment](#) methods [along with](#) the development of new materials that can potentially reduce these environmental problems. Among various types of existing treatment, water treatment using activated carbon is quite promising.⁷

Activated carbons (ACs) have been widely utilized as effective adsorbents in water treatment. The ACs with porous structures have high surface area and high adsorption capacity.⁸ They can remove a variety of pollutants, such as dyes, heavy metals, and gases. [The preparation of](#) ACs from cheap and renewable precursors is in [high](#) demand. Many researchers have reported the production of ACs from different low-cost precursors, such as bamboo,⁹ coconut shell,¹⁰ flamboyant pod,¹¹ olive stone,¹² olive husk,¹³ cotton stalk,¹⁴ coffee endocarp,¹⁵ plum kernel,¹⁶ pistachio shell,¹⁷ and fir wood.¹⁸ [Previous reports have demonstrated that it is critical to obtain](#) [the-a](#) high surface area of ACs to maximize the adsorption capacity.¹⁹ Tailoring both pore size (*e.g.*, bimodal micropore and mesopore structures) and sites [functionalization](#) on the pore surface is also effective.²⁰

Generally, the ACs can be prepared by physical [or](#) chemical activations. The physical activation can be carried out by direct carbonization of precursor at different temperatures, generally from 800 °C to 1100 °C under inert atmosphere. On the other hand, the chemical activation is carried out by using different activating agents followed by the carbonization of [the](#) mixtures (precursors and activating agents) [at](#) temperature [ranging](#) from 400 °C to 900 °C.²¹ After carbonization, [the samples need to be washed carefully in order](#) to remove the remaining chemical agents.²²

The chemical activation method is more advantageous due to its low carbonization temperature, and high product yield.²³ For the preparation of ACs from carbonaceous precursors, several chemical activating agents (*e.g.*, sulphuric acid (H₂SO₄) potassium hydroxide (KOH), NaOH, calcium chloride (CaCl₂), magnesium

chloride (MgCl_2), phosphoric acid (H_3PO_4), zinc chloride (ZnCl_2), and ferric chloride (FeCl_3)) have been utilized so far.²⁴⁻²⁸ [Activation through NaOH](#)²⁹⁻³¹ is better than KOH because it can be used at lower dosage.

In this paper, we report a facile [route for the](#) synthesis of nanoporous activated carbons (NACs) derived from Lapsi (*Choerospondias axillaris*) seed powder (LSP). LSP can be used as low-cost precursor, because Lapsi is a wild, large, deciduous and dioecious fruit tree under the Anacardiaceae. The LSP was [mixed](#) with sodium hydroxide (NaOH) [in 1:1 wt. ratio](#) and carbonized at 400 °C for different time (3, 4, and 5 hours) and at different temperature (400, 500, 600, and 700 °C) for 3 hours under a constant flow of ultrapure nitrogen gas. The obtained LSP-derived nanoporous activated carbons (NACs) [were](#) carefully characterized and the effect of the conditions [during preparation](#) on the structural properties [was](#) also investigated.

2. Experimental

Materials. LSP was collected from local food Industry of Kathmandu, Nepal. NaOH and hydrochloric acid (HCl) (purity > 99.99%) were purchased from Qualigen, India. A continuous flow of ultra-pure nitrogen was set up during carbonization.

Preparation of NACs. LSP was carefully washed with distilled water three times and [then](#) dried at 110 °C for 24 hours. Dried LSP was crushed in an electric grinder. Then, NaOH was directly mixed with LSP (The weight ratio of LSP:NaOH was 1:1.). After that, NaOH-loaded LSP was carbonized for different times (3, 4, and 5 hours) at 400 °C under a constant flow (100 ml·min⁻¹) of N₂ gas. Similarly, NaOH-loaded LSP was carbonized at different temperatures (400, 500, 600, and 700 °C) for 3 hours. After carbonization, as-prepared samples were washed with 1M HCl solution to remove impurities, and washed several times with warm distilled water (around 40 °C) to remove the chlorides. The obtained materials were then dried at 110 °C for 24 hours, [followed by another grinding process](#). The obtained [final products](#) are noted as NAC_*x*_*y*, where *x* and *y* indicate the applied carbonization temperatures and times, respectively.

Characterization. Surface functional groups of NACs were determined by FT-IR spectra (Thermo Electron Corporation, Nicolet 4700). Wide-angle X-ray diffraction (XRD) patterns of NACs were measured by Rigaku X-ray diffractometer (RINT) under operation of 40 kV and 40 mA with Cu-K α radiation. Raman scattering spectra were recorded on a Jobin-Yvon T64000 by exciting samples with green laser (514.5 nm). For Raman scattering measurements, the samples were put on clean glass substrate. [The](#) surface morphology and [the](#) porous structure of NACs were investigated by scanning electron microscope (SEM) (Hitachi S-4800). For preparation of SEM samples, the [materials](#) were put on carbon tapes. Nitrogen adsorption-desorption isotherms were recorded at 77 K on an automatic adsorption instrument (Quantachrome Autosorb-1). Before the measurement, the samples were degassed for 24 h at 120 °C. To measure the concentration of remaining MB, the [optical absorption of the](#) suspensions were measured at λ_{max} of 660 nm using the UV-Vis spectrophotometer (UV-CECIL-CE-100).

3. Results and discussion

Figure 1 shows SEM images of all the samples at different magnifications. LSP-derived NACs in the powder form have a wide range of particle size. The surface morphology and porosity seems apparently similar for all the samples, although they were carbonized in different conditions. The surface structures of NACs are in irregular shape. On the sample surface, many macropores in the micrometer length scale were clearly observed. From high resolution SEM images, it was clearly confirmed that mesoscale pores in the range from 10 nm to 50 nm were also formed. Thus, the carbonization of LSP using NaOH induced the formation of well-developed porous structures with different pore sizes.

To further investigate the porosity, the surface areas of the obtained NACs was measured through Brunauer-Emmett-Teller (BET) method using nitrogen adsorption-desorption isotherms. Nitrogen adsorption-desorption isotherms of NACs carbonized at 400 and 700 °C for 3 hours, and NAC carbonized at 400 °C for 5 hours are shown in **Figure 2a**. For comparison, NAC prepared without NaOH is also shown. As clearly seen in **Figure 2a**, the adsorption isotherms are strictly dependent on the preparation conditions, demonstrating that carbonization time and temperature is very critical. In the case of NAC_400_3, a clear nitrogen uptake was observed a low relative pressure (P/P_0) (< 0.1) region, showing the presence of micropores (less than 2 nm in diameter). On the other hand, the further nitrogen uptake slightly increased in the higher relative pressure region. A small hysteresis loop was also observed at higher relative pressure region. These situations have been often seen in nanoporous materials with randomly arranged pores having various sizes. It is additionally noted that the nitrogen uptake decreased significantly after increasing the carbonization temperature and time. The sample carbonized without NaOH shows low nitrogen uptake, indicating that the chemical treatment with NaOH plays an important role for developing nanoporous structures. **Figure 2b** summarizes the calculated BET surface areas of NACs prepared at different carbonization times and temperatures. Large drop in the surface area following an increase in carbonization time and temperature is thought to be caused by the collapsing of the original nanopores (The details are discussed in later section).

Wide-angle XRD patterns for the obtained NACs prepared at different carbonization time and temperature are shown in **Figure 3**. All the NACs show two broad diffraction peaks which can be attributed to the (002) and (100) planes of graphitic structures. The peak intensities are relatively weak, indicating that the graphitic structures are not fully developed. This situation is also common to commercially available ACs.³² The graphitic degrees were further considered by using Raman scattering spectra (**Figure 4**). Two strong Raman peaks were observed at approximately 1349 and 1596 cm⁻¹. The vibrational bands at 1350 cm⁻¹ and 1600 cm⁻¹ are well known as *D* (disordered) and *G* (graphitic) bands, respectively. The relative intensity ratio of *G* and *D* bands (I_G/I_D) indicates the graphitization degree.³³⁻³⁵ For NACs, this graphitization degree depends on the applied carbonization time and temperature, although the wide-angle XRD measurement cannot detect the difference of graphitization degree (**Figure 3**). Interestingly, a relationship between the surface areas and graphitic degrees could be established, as shown in **Figure 5**. The samples with higher I_G/I_D value (*i.e.*, higher graphitic degrees) exhibit smaller surface area. In the case of NaOH treatment, the pore formation is caused by the introduction of sodium ions in the carbon structure.³⁶ The increase in carbonization time and temperature is expected to promote the reaction kinetics, thereby developing the nanopore structure inside the carbon matrix. As mentioned above, however, at higher temperature (> 400 °C), the nanoporous structure may coalesce or break, due to a well-documented gasification process ((i) $\text{NaOH} \rightarrow \text{Na}_2\text{O} + \text{H}_2\text{O}$, and (ii) $\text{H}_2\text{O} + \text{C} \rightarrow \text{CO} + \text{H}_2$) and/or the gradual graphitization (as shown in **Figure 5**).³⁷

Infrared spectroscopy is one of the key tools to elucidate the surface functional groups of many organic substances. **Figure S1** (supporting information) shows FT-IR spectra of the selected two NACs prepared at different carbonization temperature (NAC_400_3, NAC_700_3). The appearance of several sharp peaks confirms that the obtained NACs contain different surface functional groups. For example, a broad band in the range of 3200-3600 cm⁻¹ (centered at around 3429 cm⁻¹) is attributed to the stretching of OH bonds. For the samples carbonized above 600 °C, this band disappear, as seen in the spectrum for NAC_700_3 (**Figure S1**), indicating that higher carbonization temperature breaks the OH functional group.³⁸ The bands present at 2923 and 2852 cm⁻¹ correspond to asymmetric and symmetric stretching of the C-H bond of the CH₂ group,

1 respectively. A small band observed at 1700 cm^{-1} can be assigned to C=O stretching vibrations of ketones,
2 aldehydes, lactones or carboxyl groups. Similarly, the two bands around at 1441 and 1599 cm^{-1} correspond to
3 C-C -stretching of aromatic C-C bond and the weak bands that are observed in the range of $755\text{-}875\text{ cm}^{-1}$ can be
4 assigned to C-H bending mode of aromatic ring.³⁹ Thus, it is revealed that the FT-IR spectra of our samples
5 showed a clear evidence of the presence of the various surface functional groups.^{38,39}

6 The performance of NACs is usually expressed in terms of iodine number. Higher iodine number means
7 higher activation. The iodine number has been also known as one of [the](#) important tools for measuring
8 porosity.⁴⁰ According to [a](#) previous report,⁴¹ we measured the iodine numbers of the obtained NACs. NAC
9 samples (0.1 g) was mixed with 5% HCl solution (5 mL). The mixture was boiled for a few hours and then
10 cooled down to room temperature ($25\text{ }^{\circ}\text{C}$). A 0.05 M iodine aqueous solution (10 mL) was added into the
11 mixture [which](#) was vigorously shaken for several minutes [and then](#) filtered. A 10 mL of filtrate was titrated
12 against 0.1 M hypo solution ($\text{Na}_2\text{S}_2\text{O}_3$) using starch as an indicator. The concentration of iodine adsorbed on
13 ACs was calculated according to the previous report.⁴¹ **Figure S2** shows the variation of iodine number of the
14 obtained NACs at different carbonation conditions. As seen in **Figure S2a**, when the carbonization temperature
15 was fixed to be $400\text{ }^{\circ}\text{C}$, the iodine number is [reduced by](#) increasing the carbonization time from 3 hours to 5
16 hours. The carbonization temperature also showed a similar impact on the pore structure (**Figure S2b**). The
17 iodine number is decreased [by increasing the](#) temperature from $400\text{ }^{\circ}\text{C}$ to $700\text{ }^{\circ}\text{C}$. These trends are similar [to](#)
18 the variation of surface area, as shown in **Figure 5**. The highest iodine number ($758\text{ mg}\cdot\text{g}^{-1}$) [corresponds to the](#)
19 NAC with the highest surface area (carbonized at $400\text{ }^{\circ}\text{C}$ for 3 hours). [This iodine number is comparable to the](#)
20 [commercially available activated carbon. The iodine number of commercial activated carbon \(Qualigen, India\)](#)
21 [was ca. \$782\text{ mg}\cdot\text{g}^{-1}\$.](#) This result shows that LSP-based NACs can be utilized as a new family of efficient AC in
22 water treatment.

23 As mentioned above, AC materials have been utilized as an efficient adsorbent for [the](#) removal of
24 several organic dyes [from water](#), such as MB (basic) and methyl orange (acidic).⁴² [To](#) study [the adsorption](#) of
25 MB, multi-points isotherm adsorption tests were carried out using Langmuir model. NAC samples (0.1 g) were

mixed with 100 mL of MB solutions with different concentrations (25, 50, 100, 150, 200, 500, and 1000 mg·L⁻¹). The mixtures were shaken for 24 hours on a mechanical shaker. The suspensions were filtered out and the concentration of remaining MB was [deduced from optical absorption measurements](#) at $\lambda_{\max}$ of 660 nm using the UV-Vis spectrophotometer. The maximum adsorption capacity of MB on NAC (q_e , mg·g⁻¹) was determined using the following Langmuir isotherm.⁴³

$$q_e = \frac{(C_o - C_e)V}{M}$$

where C_o (mg·L⁻¹) is the initial concentration of MB, C_e (mg·L⁻¹) is the equilibrium concentration of MB, V (L) is the volume of MB solution, and M (g) is the mass of NAC sample. **Figure 6** shows the variation of MB number of ACs prepared at different carbonization times and temperatures. Our LSP-based NACs showed a good MB adsorption capacity and the maximum MB value of *ca.* 201 mg·g⁻¹ was [obtained](#) by using NAC sample having maximum BET surface area of 1002 mg·g⁻¹. [This value is comparable with the commercial granular activated carbons \(100-300 mg·g⁻¹\),⁴² demonstrating that our AC derived from low-cost precursor \[can be used in dye adsorption application with equal potential\]\(#\).](#) However, the adsorption capacity was not correlated well with the BET surface area. For example, NAC_400_3 and NAC_400_4 exhibit almost the same MB absorption ability, although there is a drastic difference in their surface areas (*i.e.* NAC_400_3 is 1002 m²·g⁻¹, while NAC_400_4 is only 40 m²·g⁻¹). The ability of MB adsorption is determined by several factors. The high nanoporosity and open pore network of the carbon matrix can facilitate fast molecular diffusion as well as promote accessibility to the entire pore surface. The presence of highly graphitized walls are more critical. The MB dye more effectively interacted with the carbon surface due to the π - π interaction⁴⁴ between the MB molecule and the graphitic carbon (*sp*²-carbon).

4. Conclusion

We have prepared NACs from LSP with NaOH and have carefully studied the effect of carbonization time and temperature on the structures and properties. The maximum surface area of approximately $1000 \text{ m}^2 \cdot \text{g}^{-1}$ was obtained from NAC carbonized at 400°C for 3 hours. With the increase of carbonization time and temperature, the graphitic structure was gradually developed and the surface area was drastically reduced. Our NAC with the highest surface area appears to be an efficient adsorbent for the removal of MB from aqueous solution. Therefore, we believe that our approach is quite applicable to large-scale production of NAC.

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4

5 **Figure 1** SEM images of LSP-based NACs (a) carbonized at 400 °C for different times (3, 4, and 5 hours), and (b)
6 carbonized for 3 hours at different temperatures (500, 600, and 700 °C).

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1 **Figure 2**

2

3 **Figure 2** (a) N₂ adsorption-desorption isotherms of LSP-based NACs (NAC_400_3, NAC_700_3, NAC_400_5,
4 and NAC_400_3 prepared without NaOH), and (b) Summary on the BET surface areas of NACs.

5

1 **Figure 3**

2
3 **Figure 3** Wide-angle XRD patterns of LSP-based NACs (a) carbonized at 400 °C for different times (3, 4, and 5
4 hours), and (b) carbonized for 3 hours at different temperatures (500, 600, and 700 °C).
5

1 **Figure 4**

2

3 **Figure 4** Raman scattering spectra of LSP-based NACs (a) carbonized at 400 °C for different times (3, 4, and 5
4 hours), and (b) carbonized for 3 hours at different temperatures (500, 600, and 700 °C).

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6

1 **Figure 5**

2

3 **Figure 5** Relationship between the surface areas and the I_G/I_D values of LSP-based NACs (a) carbonized at
4 400 °C for different times (3, 4, and 5 hours), and (b) carbonized for 3 hours at different temperatures (400,
5 500, 600, and 700 °C).

6

1 **Figure 6**

2
3 **Figure 6** Variation of MB number (*i.e.*, adsorption amount of MB) of LSP-based NACs (a) carbonized at 400 $^{\circ}\text{C}$
4 for different times (3, 4, and 5 hours), and (b) carbonized for 3 hours at different temperatures (400, 500, 600,
5 and 700 $^{\circ}\text{C}$).

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