Supportive Information

Highly Efficient Multisubstrate Agricultural Waste-Derived Activated Carbon for Enhanced CO₂ Capture

Mardikios Maja Bade ¹, Amare Aregahegn Dubale ¹,*, Dawit Firemichael Bebizuh ¹ and Minaleshewa Atlabachew ²

¹ Department of Chemistry, College of Natural and Computational Science, Energy and Environment Research Center, Dilla University, PO. Box 419, Dill, Ethiopia
² Department of Chemistry, College of Science, Bahir Dar University, PO. Box 79, Bahir Dar Ethiopia

*Corresponding authors
Email: amare2122@gmail.com

Chemicals and Reagents
All the reagents were commercially available and were used as received without further purification. Hydrochloric acid (HCl) was used to remove inorganic impurity from the surface of activated carbon, distilled water < 5.0 µS, potassium iodide (KI), Iodine (I₂), sodium thiosulphate (Na₂S₂O₄), and starch were used for conducting iodine test, sodium carbonate (Na₂CO₃), sodium hydroxide (NaOH) and potassium hydroxide (KOH) were used as activation of the powder composite precursor of activated carbon and acts as dehydration reagent during chemical activation, nitric acid (HNO₃), sulfuric acid (H₂SO₄), glacial acetic acid (CH₃COOH), D-glucose as well as anthrone reagent were used for conducting cellulose estimation.

Apparatus and Instruments
Drying oven, model DHG-9030, made in Japan was used for sample drying purpose, electronic balance model ESJ-200-4 made in Japan was used to measure sample weight, mortar and pestle for grinding, 250 µm mesh size sieve, electrical muffle furnace, model 91E, made in England was used for carbonization, graduated cylinders (100, 500, 1000 and 2000 mL) to measure volume, Mercury thermometer to measure temperature, heating mantle with a stirrer (Horst),
water bath model W14, centrifuge, test tubes, cubit 1 cm, UV-visible spectrophotometry NV203 used for cellulose estimation purpose, pH meter, Platinum crucible, Burette digital model 10E31273 made in Germany was used for iodine test, Air compress was used to water displacement test, 1 mL 30G× 80 mm syringe was used as a column, Gas analyzer GeoTech UK Model used to analyze the gas composition, and airtight syringe was used to measure gas volume.

**Cellulose content and proximate analysis of agricultural wastes**

The composition of the raw material is an important factor in selecting precursor for activated carbon production. The chemical composition of the precursor materials, mainly the percentages of the cellulose in agricultural wastes were set up by means of standard procedures.

**a. Estimation of Cellulose**

Cellulose is a major structural polysaccharide in plants and the most rich organic compound in nature, as well as is composed of glucose units joined together in the form of the repeating units of the disaccharide with several cross associations. The percentages of the cellulose in agricultural wastes were estimated by standard methods [1]. The ratio 3 mL of acetic acid and nitric acid mixture (150 mL of 80 % acetic acid + 15 mL of concentrated nitric acid) was mixed with 1.0 g of powdered sample and placed in a water bath at 100 °C for 30 min. After centrifugation, the collected residue was washed with distilled water, added 1 mL of 67 % H₂SO₄ and left for 1 h. Then 1 mL of the solution was diluted to 100 mL, and 10 mL of anthrone reagent was added to 1 mL of this solution and mixed well. The tubes were heated in a water bath for 10 min and measured the absorbance at 630 nm after cooling. The amount of cellulose was determined from the standard graph.

**b. Proximate Analysis**

Proximate analysis was done to determine the average of the percentage volatile matter content, ash content, moisture content and content of fixed carbon of the biomass obtained from three repeats. The procedures of the ASTM standard D5373-02 [2] was adopted to obtain the percentage of ash, volatile matter and moisture contents in the peanut shell, coffee husk, corn cob and banana peels used as starting materials.
i) Percentage moisture content
Moisture analysis was carried out using the drying oven method. Porcelain crucibles were weighed and their masses recorded. About 2 g of each agricultural waste sample was weighed into pre-weighed crucibles, and it was dried in the drying oven at 105 °C for 3 h. Then it was cooled in desiccators for 1 h and reweighed. The percentage moisture content was determined by the following formula:

$$\text{percentage of moisture} = \frac{\text{initial mass (g)} - \text{mass after oven dried (g)}}{\text{mass of sample (g)}} \times 100$$

ii) Percentage ash content
Ash content of agricultural wastes were determined by heating 2 g weight of powdered sample into ceramic crucibles and it was dried and reweighed to obtain the dry carbon weight. The sample was heated in an electrical furnace at 650 ± 25 °C for 3 h. The crucible was cooled in a desiccator, and remaining solids was weighed. The percentage of ash is calculated by:

$$\text{Ash\%} = \frac{\text{remaining solids weight (g)}}{\text{original carbon weight (g)}} \times 100$$

iii) Percentage volatile matter
The percentage volatile matter (PVM) was determined by 2 g of powdered sample was placed into weighed crucible and set it in air drying oven until a constant weight was obtained. The portions are then kept in a furnace at a temperature of 900 °C for 7 min and weighed after cooling in a desiccator for 30 minutes. The PVM was calculated using the equation below:

$$\text{PVM} = \frac{A - B}{A} \times 100$$

where, A is the weight of the oven-dried sample and B is the weight of the sample after 7 min in the furnace at 900 °C.

Percentage of fixed carbon = 100 – (% ash + % moisture + % volatile matter)

Preparation of multi-substrate AC composite
a. The effect of chemical activating agents
The efficient or effective activating agent was obtained, the agricultural wastes composite substrate impregnated with different chemicals such as sodium hydroxide (NaOH), potassium...
hydroxide (KOH), and sodium carbonate (Na$_2$CO$_3$). Previously achieved agricultural wastes composite substrate impregnated with chemical activating agents ratio (w/w) 1:1 for 12 h and dried for 8 h in oven at 110 °C. The dried impregnated composite sample was carbonized in an electric muffle furnace at 400 °C for 90 minutes. After carbonization, the mixture was removed from the furnace and allowed to cool at room temperature. The pyrolyzed carbon was washed with 5% HCl for 2 to 3 times, then washed several times with distilled water until a neutral pH reached. Afterward, the carbon paste was dried in a drying oven at 110 °C for 24 h, then cooled at room temperature and sieved by 250 µm mesh size to attain a uniform particle size besides the iodine test performed.

b. **Optimization of activated carbon production parameters**

Optimization is carried out to make some parameters function at their best by varying one parameter while keeping other parameters constant. The effect of major parameters such as impregnation ratio, activation (impregnation) time, carbonization temperature, and holding (carbonization) time were optimized as follows.

i. **Optimization of chemical impregnation ratio**

The effect of impregnation ratio of chemical agents such as sodium carbonate (Na$_2$CO$_3$) on the porous characteristics was studied by varying the impregnation ratios of activating agent to agricultural wastes composite (w/w) from 0.2 to 1.5 (0.2, 0.25, 0.5, 1.0, and 1.5) and fixing the other parameters such as impregnation time 12 h, carbonization temperature 400 °C and holding time 90 minute. After 12 h activating time the excess solution was filtered and dried for 8 h in oven at 110 °C. The dried sample was carbonized in an electric muffle furnace at 400 °C for 90 minute. After carbonization, the mixture was removed from the furnace and allowed to cool at room temperature and sieved through 250 µm mesh size and also iodine test was achieved.

ii. **Optimization of activation/impregnation time**

The optimum impregnation time was attained, previously optimized impregnation ratio mean Na$_2$CO$_3$ to composite substrate impregnated for 6 h, 12 h, 24 h, 36 h and 48 h with fixed other parameters as previously well-defined for chemical impregnation ratio. After the impregnation time was achieved, the excess solution was filtrated and air-dried for 8 h in oven at 110 °C.
A dried sample was carbonized in an electric muffle furnace at 400 °C for 90 minutes. After carbonization, the mixture was removed from the furnace and allowed to cool at room temperature and sieved as well as iodine test performed.

iii. **Optimization of carbonization temperature**

Carbonization temperature is one of the most influencing factors for the development of porosity in the activated carbon. Effect of carbonization temperature on porous characteristics of produced activated carbon was studied in the range 350 – 550 °C (350, 400, 450, 500, and 550°C) with fixing other parameters as described above for chemical impregnation ratio and activation time.

iv. **Optimization of carbonization time**

Holding time is the duration of the sample kept at the final carbonization temperature. Previously optimized parameters such as impregnation ratio, impregnation time, and carbonization temperature are fixed, and holding time was varied in between 30 – 150 min (30, 60, 90, 120, and 150 min) to investigate its effect on porous characteristics.

**Iodine Test**

The iodine number is determined according to the ASTM D4607-94 [3] method. The iodine number is defined as the milligrams of iodine adsorbed by 1.0 g of carbon when the iodine concentration of the filtrate is 0.02 N. The experiment consists of 1 g activated carbon powdered sample treated with 10.0 mL of 5% HCl. This mixture is boiled for 30 s and then cooled. Before long then, 100.0 mL of 0.1 N iodine solutions was added to the mixture and stirred for 30 s. The resulting solution was filtered and 50.0 mL of the filtrate was titrated with 0.1 N sodium thiosulfate, used starch as indicator. The iodine number \( \frac{X}{M} \) is calculated as follow:

\[
\frac{X}{M} = \left\{ \frac{(N_1 \times 126.93 \times V_1) - \left[ \frac{(V_1 + V_{HCl})}{V_F} \right] \times (N_2 \times 126.93 \times V_2)}{M_C} \right\}
\]

where \( N_1 \) is the iodine solution normality, \( V_1 \) is the added volume of iodine solution, \( V_{HCl} \) is the added volume of 5% HCl, \( V_F \) is the filtrate volume used in titration, \( N_2 \) is the sodium thiosulfate
solution normality, \( V_2 \) is the consumed volume of sodium thiosulfate solution and \( M_C \) is the mass of activated carbon.

![Figure S1](image.png)

**Figure S1.** Adsorption experimental design (a) CO\(_2\) gas cylinder with flow rate controller, (b) gas analyzer and (c) air tight syringe

| AC       | Trial | AC (g) | \( V_{\text{HCl}} \) (mL) | \( V_{\text{Iodine}} \) (mL) | \( V_{\text{filtrate}} \) (mL) | \( V_{\text{Titrant}} \) (mL) | \( X/M \) (mg/g) | Mean \( X/M \) (mg/g) |
|-----------|-------|--------|----------------|----------------|----------------|----------------|----------------|-----------------|
| SH-CAC    | 1     | 1      | 10             | 100            | 50             | 25.3           | 564            |                 |
|           | 2     | 1      | 10             | 100            | 50             | 25.3           | 564            | 564 ± 0.6       |
|           | 3     | 1      | 10             | 100            | 50             | 25.2           | 565            |                 |
|           | 1     | 1      | 10             | 100            | 50             | 24.1           | 597            |                 |
| PH-CAC    | 2     | 1      | 10             | 100            | 50             | 24.1           | 596            | 597 ± 0.85      |
|           | 3     | 1      | 10             | 100            | 50             | 24.1           | 598            |                 |
| SC-CAC    | 1     | 1      | 10             | 100            | 50             | 23.0           | 627            |                 |
|           | 2     | 1      | 10             | 100            | 50             | 23.0           | 628            | 627 ± 0.55      |
|           | 3     | 1      | 10             | 100            | 50             | 23.0           | 626            |                 |

At the 0.05 level, the means are significantly different.
### Table S2: Optimum impregnation ratio

| Chemical to composite ratio (w/w) | IT (h) | CT (°C) | HT (min) | X/M (mg/g) |
|---------------------------------|--------|---------|---------|------------|
| 0.2                             | 12     | 400     | 90      | 691 ± 0.3  |
| 0.25                            | 12     | 400     | 90      | 721 ± 0.2  |
| 0.5                             | 12     | 400     | 90      | 664 ± 0.1  |
| 1                               | 12     | 400     | 90      | 627 ± 0.6  |
| 1.5                             | 12     | 400     | 90      | 533 ± 0.6  |

### Table S3: Optimum impregnation time

| Chemical to composite ratio (w/w) | IT (h) | CT (°C) | HT (min) | X/M (mg/g) |
|---------------------------------|--------|---------|---------|------------|
| 0.25                            | 6      | 400     | 90      | 654 ± 0.7  |
| 0.25                            | 12     | 400     | 90      | 721 ± 0.2  |
| 0.25                            | 24     | 400     | 90      | 741 ± 0.9  |
| 0.25                            | 36     | 400     | 90      | 679 ± 0.2  |
| 0.25                            | 48     | 400     | 90      | 637 ± 0.3  |

### Table S4: Optimum carbonization temperature

| Chemical to composite ratio (w/w) | IT (h) | CT (°C) | HT (min) | X/M (mg/g) |
|---------------------------------|--------|---------|---------|------------|
| 0.25                            | 24     | 350     | 90      | 678 ± 0.6  |
| 0.25                            | 24     | 400     | 90      | 741 ± 0.9  |
| 0.25                            | 24     | 450     | 90      | 774 ± 0.1  |
| 0.25                            | 24     | 500     | 90      | 695 ± 0.3  |
| 0.25                            | 24     | 550     | 90      | 590 ± 0.1  |
### Table S5: Optimum carbonization /holding time

| Chemical to composite ratio (w/w) | IT (h) | CT (°C) | HT (min) | X/M (mg/g) |
|----------------------------------|--------|---------|----------|------------|
| 0.25                             | 24     | 450     | 30       | 580 ± 0.6  |
| 0.25                             | 24     | 450     | 60       | 608 ± 0.4  |
| 0.25                             | 24     | 450     | 90       | 774 ± 0.1  |
| 0.25                             | 24     | 450     | 120      | 651 ± 0.1  |
| 0.25                             | 24     | 450     | 150      | 605 ± 0.1  |

### Table S6: Comparison of iodine adsorption capacities of various activated carbons.

| Adsorbent                                | X/M (mg/g) | Reference |
|------------------------------------------|------------|-----------|
| Almond shell activated carbon           | 737        | [4]       |
| Hazelnut shell activated carbon          | 731        | [4]       |
| Walnut shell activated carbon            | 679        | [4]       |
| Bio-waste activated carbon               | 704        | [5]       |
| SH-CAC                                   | 655        | This study|
| PH-CAC                                   | 680        | This study|
| SC-CHAC                                  | 706        | This study|
| SC-CAC                                   | 774        | This study|

### Table S7. Percentage yield of the as-prepared composite activated carbon

| Sample | Weight of Na₂CO₃ (g) | Activation parameters | Percentage yield (%) |
|--------|----------------------|-----------------------|----------------------|
|        |                      | Impregnation time (h) | Carbonization temperature (°C) | Carbonization time (min) |                     |
| AC-0   | 0                    | 24                    | 450                  | 90                    | 31.4                |
| AC-0.2 | 0.2                  | 24                    | 450                  | 90                    | 58.1                |
| AC-0.25| 0.25                 | 24                    | 450                  | 90                    | 92.3                |
| AC-1.1 | 1.1                  | 24                    | 450                  | 90                    | 41.7                |
| Sample name   | grain size(Å) |
|---------------|---------------|
| SH-CAC        | 16.2          |
| PH-CAC        | 15.0          |
| SC-CAC        | 0.477         |
| SC-CHAC       | 0.677         |

**Table S9:** BET information for the prepared composite AC

| Sample   | BET information |
|----------|-----------------|
|          | $S_{\text{BET}}$ (m$^2$ g$^{-1}$) | Pore size (nm) | Pore volume (cm$^3$ g$^{-1}$) |
| SH-CAC   | 275             | 1.63           | 0.014                           |
| PH-CAC   | 426             | 4.25           | 0.021                           |
| SC-CAC   | 1239            | 14.7           | 0.097                           |
| SC-CHAC  | 931             | 7.47           | 0.043                           |

**Table S10:** Comparison of surface area and total pore volume of various activated carbon.

| Adsorbent       | BET surface area (m$^2$ g$^{-1}$) | Total pore volume (cm$^3$ g$^{-1}$) | Reference |
|-----------------|------------------------------------|-------------------------------------|-----------|
| Spent Tea leave AC | 1044                          | 0.590                              | [6]       |
| Pumpkin seed shell AC | 1421                         | 0.908                              | [7]       |
| Bagasse AC       | 923                               | 0.530                              | [8]       |
| Rice husk AC     | 927                               | 0.560                              | [8]       |
| Olive stones AC  | 1173                              | -                                  | [9]       |
| Jute fibers AC   | 1224                              | 0.740                              | [10]      |
| SC-CAC           | 1239                              | 0.973                              | This study |
Figure S2. Carbon dioxide adsorption capacity at different residence time

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