

FULL PAPER

# Bio-Based carbon nanomaterials synthesis from waste tea

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Carbon nanoflakes were fabricated from domestic waste tea leaves as a bio-alternative source of carbon in one step. The fabricated nanoflakes based-carbon were characterized by field-emission scanning electron microscope, Fourier transform infrared, energy dispersive X-ray analysis, and Raman spectroscopy. In addition, thermogravimetric analysis was investigated to examine the thermal stability of the products. The morphology of the products showed the observation of carbon-flakes nanomaterials having thin graphitic outer mono shells with a diameter of about 35 nm and different monolith sizes of about 15 to 180 nm. The as-obtained materials demonstrated good thermal stability with a yield of 48% due to the high temperature used in the deposition stage. The fabrication showed that the temperature range of the deposition step in addition to the cooling step plays a major role in the nanoflakes formation.

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**KEYWORDS**

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## Introduction

Tea is one of the largest consumed drinks in the world. In fact, it is the second drink (water is the first) with a consumption of about 6.3 billion kilograms in 2020 and estimated to reach 7.4 billion kilograms by 2025 [1]. The global tea production in 2021 was estimated at 6.5 million tons. Many countries are in the lead existed in tea consumption and production. For example, in 2016, Turkey was the largest tea-consuming country in the world, with a per-person tea consumption of approximately 6.96 pounds per year while, China had an estimated consumption of 1.25 pounds per capita [1,2].

With such an increase in production and consumption, the waste of both plants and consumed tea was increased. Tea wastes are one of the most world waste accumulations

due to the extensive use of the tea plant as a beverage and in bakeries [3]. These accumulations generated yearly additional issues to the growing environmental problems since most methods of de-eliminating these wastes are either burning or burying them. Both processes do not solve the problem as they give certain damages to the land and the air. Therefore, converting tea wastes into a valuable material is an effective way to decrease the load on our environment. As such tea wastes have been used in the manufacturing of particleboard, fertilizer, and for energy by producing liquid and solid (briquet) fuels [4-6].

In addition, tea wastes have been used in the production of the adsorbent material namely activated carbon, and for the removal of heavy metals from industrial water, supporting catalyst materials in some

industrial processes, and fabrication of nanomaterials-based carbon [4,7].

The activated carbon has been prepared from the tea woody part which points out the major tea-wastes after harvesting using phosphoric acid under different charring temperatures. The prepared active carbon has been used as a catalyst supporter of cobalt, nickel, and copper for hydrogen generation [4].

Others have used spent tea leaves to prepare activated carbon using potassium carbonate, zinc chloride, and phosphoric acid as activators and used the prepared active carbon to remove aspirin from aqueous media and the results were interesting in the particular [7]. The microwave was used in the carbonization of waste tea with  $H_3PO_4$  activation and found that the microwave energy has been affected the surface area and the BET surface area reasonably [8]. Waste tea was used to synthesize activated carbon through chemical activation using potassium acetate for the sorption of Acid Blue 25 (AB25) dye [9]. Another study uses physical activation and steam in the preparation of activated carbon where it was demonstrated that the activation temperature affected mesopore and micropore volumes and the specific surface area of the active carbon significantly [10]. Recently, nanoparticles have attracted researchers for their uniqueness and wonderful characteristics to alter them with the old products. At this point, waste tea has been used as a carbon source and carbonized at different temperatures from 400-800 °C and different times as well (30-120, 240, 360, 720, and 1440) minutes to obtain biochar subjected to a pulverizing process to obtain a mixture of nanotubes, nano-onions, and amorphous carbon. It was also reported that the highest temperature used the highest carbon and the lowest sulfur ratios obtained [3]. Others have pyrolyzed waste tea in controlled conditions to obtain water-soluble quantum dots and found them potentially interesting for hypochlorite detection as one

of the water qualities references [11]. Another team converted tea wastes into biochar via high-temperature carbonization and used the latter to produce graphene oxide compromising the method as an alternative for graphene oxide synthesis [12]. Many researchers used waste tea as a template to synthesize different metallic nanoparticles (like silver and iron) or to produce bimetallic nanocomposites (like silver-gold) structures using extraction method [13-16]. More or less, most of these particles are not precisely at the nanoscale nor with clear structure. This work aims to synthesize nanocarbons from biowaste materials through a fast method of two stages. In the first stage, domestic waste tea was dried and grounded into the powder then dried again before using it in the second stage, where the bio-powder was converted into nanocarbon directly in a high-temperature rotary furnace using metallic catalyst at different times. The results were carbon nanoflakes and some amorphous-based carbon structures.

## Materials and methods

### Chemicals

- 1- Waste tea has been collected from home after use, washed many times to remove any traces of sugar, and left to dry naturally. The dried washed waste tea was then ground using a kitchen grinder and again dried using a lab oven at 60 °C for one day.
- 2- Ferrocene,  $Fe(C_2H_5)_2$  (98%, Acros Organics) have been used as catalysts without further treatments.
- 3- Alumina boat of size 10 cm<sup>3</sup> has been used as substrate.

### Synthesis of nano-carbons

A certain amount of waste tea powder with ferrocene was loaded on the substrate (the ratio was 1:4 of ferrocene to waste tea, respectively); the substrate with its load was placed inside a quartz tube (1 m long and id=

2 cm) which was then mounted in a tubular furnace. The run started from ambient to 900 °C at a rate of 10 °C/min and was kept at that temperature for 1 hour in argon media, and then the quartz tube was removed from the furnace and the product was collected and put in a secured vial after cooling naturally. The yield was 48%.

### Testing

#### Scanning electron microscopy

The SEM images were obtained using a high-resolution field emission scanning electron microscope (FESEM) through JEOL FESEM JSM- 7500F.

#### Raman spectroscopy

Raman spectra were recorded on a Perkin-Elmer Raman Micro 200 dispersive Raman

spectrometer at the excitation wavelength of 785 nm, 350 mW.

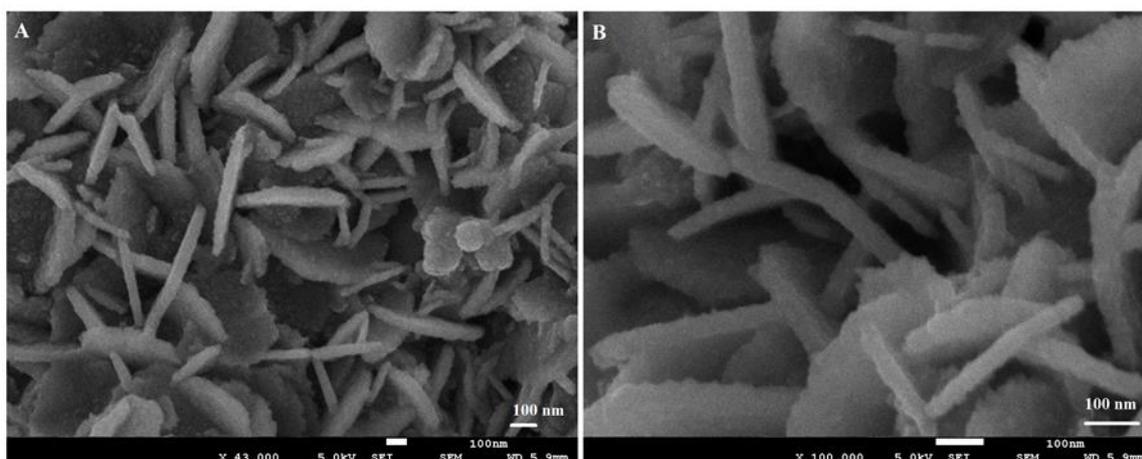
#### Thermal analysis

Thermo-gravimetric analysis (TGA) was recorded using Perkin-Elmer TGA 4000 and an inner nitrogen flow of 20 cm<sup>3</sup>/min with a heating rate of 20 °C per minute.

## Results and discussion

#### Electron microscopy

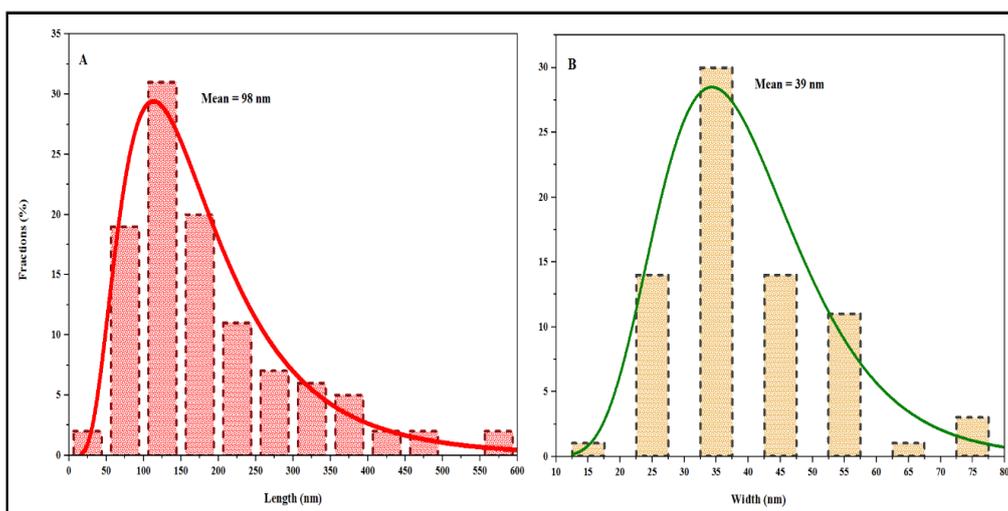
The FE-SEM images (Figure 1) revealed extensively the formation of CNFs spanning in nanometer scales, consisting mostly of potato fingers-like carbon structures, held together. It can be seen that flakes particles were distributed having a length between 27 to 240 nm, while there are some higher length flakes i.e. between 300-570 nm and the length mean was 98 nm as depicted in Figure 2 (A).



**FIGURE 1** FE-SEM images of carbon nanoflakes; (A) magnification 43000x and the scale is 100 nm and (B) magnification x100K and the scale is 100 nm

In contrast, the width of these nanoflakes was ranging from 15- 55 nm with few of a range 65-75 nm and the width mean for the nano flake's particles was 39 nm (Figure 2(B)). The flakes appeared condensed solid carbon

with taper ends, while some of them especially the large flakes have squamous sides giving the impression that they are formed from the fusion of two or more (without swelling) of the taper ends flakes.



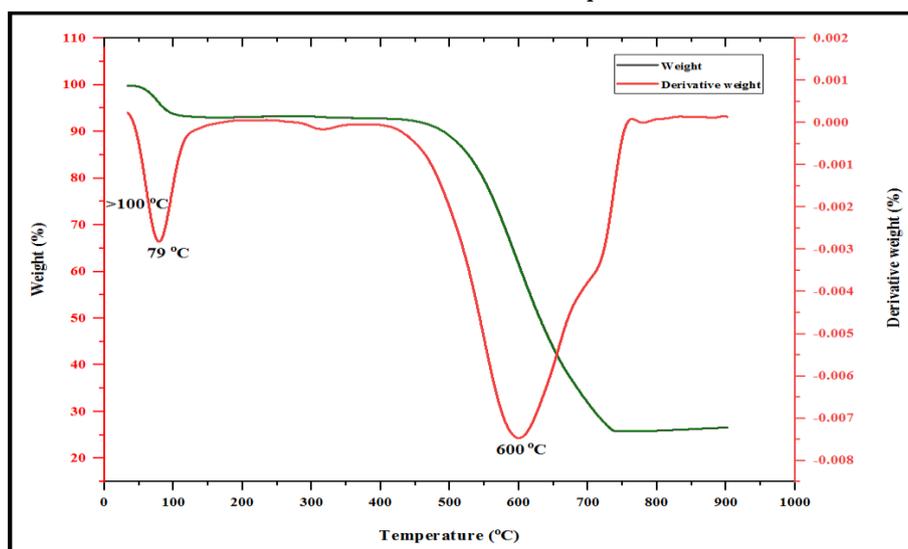
**FIGURE 2** Size distribution of nanoflakes based-carbon; (A) for the flake's length and (B) for the flake's width

### Thermal analysis

Figure 3 illustrates the TGA plot and its corresponding first-order derivative thermograph DTG.

The first draw in the weight is attributed to the dehydration of moist and surface water of the tea waste nanocarbon, where the loss of weight reached 6.84% at a temperature less than 100 °C (79 °C); the last and big draw in the weight happened in the range from 320 to

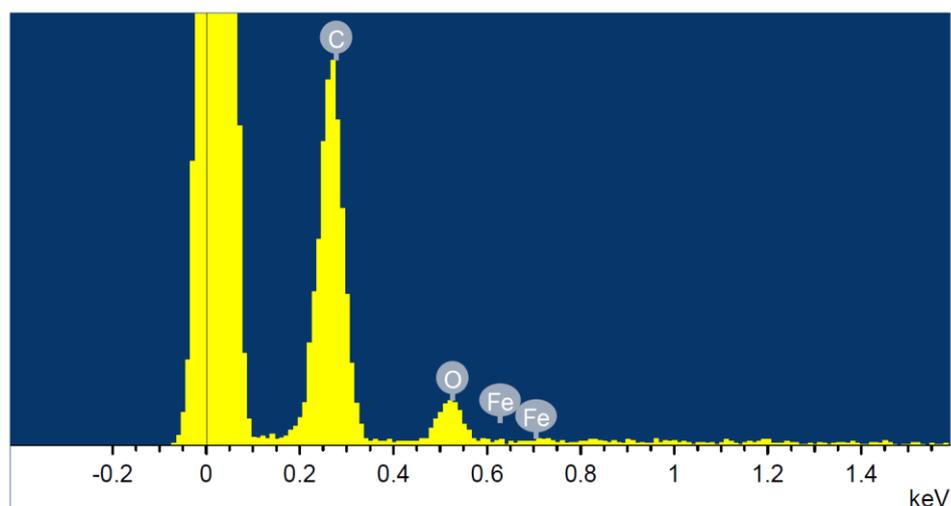
600 °C, where the loss in the weight was about 67%. The first and the second draw in the weight is attributed to the oxygenated and deoxygenated carbon, respectively, which is released during the mass degradation of the nanoflakes. The sample did not burn completely and the left residue (about 25%) possibly is because of the existence of the catalyst residue (ferrocene) since they are inorganic materials with higher resistance to the temperature.



**FIGURE 3** TGA plot and its derivative DTG of carbon nanoflakes

The EDAX analysis does support this verification where the results show the existence of iron (6.61% of the weight), while

carbon ratio was estimated (77.21%) and the oxygen ratio was (16.19 %), (Figure 4). This is agreed with the results from (6,17,18).



**FIGURE 4** The EDAX graph of carbon nanoflakes

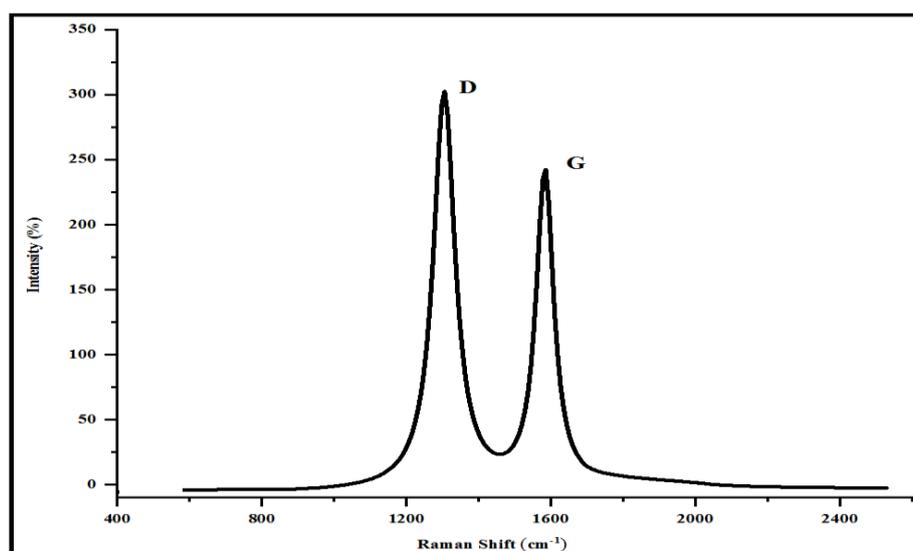
#### *Vibrational spectroscopic results*

#### *Raman spectroscopy*

One of the great tools for diagnosing nanomaterials, especially nano-based carbon is the Raman technique. It is simple and convenient in terms of the information; we can get form and the identity. We can observe the material being diagnosed. Raman spectra of carbon nanoflakes (Figure 5) show doublet at

1305.64 and 1585.88  $\text{cm}^{-1}$ , having the ratio of relative intensity which is relatively low (1.24), as presented in Table 1 suggesting that the nanoflakes are of low disordered since the intensity of the D band is not higher that much than that for the G band.

The in-plane crystalline was 3.52 nm revealing that the crystalline graphite is not of large size, while the distance between defects ( $L_D$ ) data was about 25 nm reflecting the low defected density of the prepared nanoflakes.



**FIGURE 5** Raman spectra of carbon nanoflakes

What supports this conclusion is the full width at half maximum D data found about 52

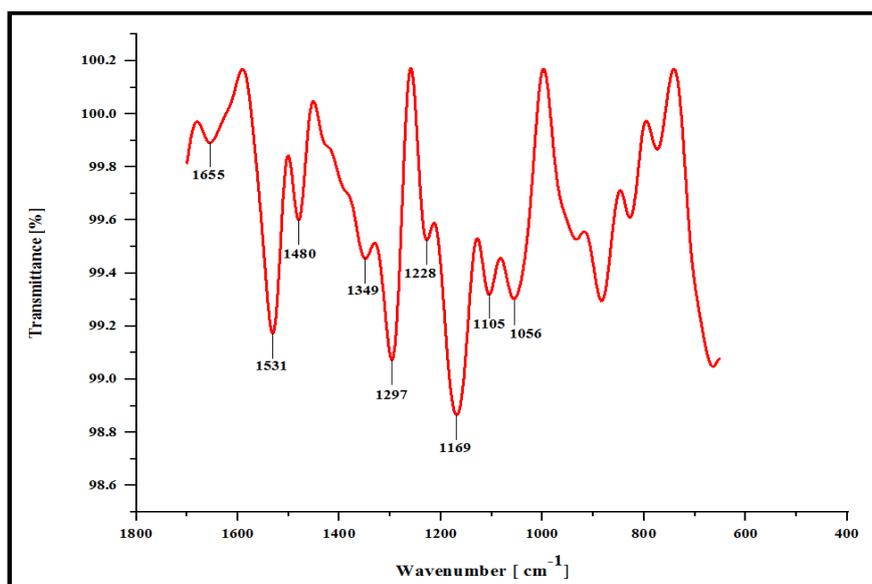
$\text{cm}^{-1}$  leaning on the fact that FWHM increases with the regime disorder and verse versa for

the G peak increased proportionally with FWHM. This is found a good agreement in the results from (17,19,20).

#### Fourier transform infrared

Figure 6 demonstrates the FTIR spectra of carbon nanoflakes. The Peaks 1349  $\text{cm}^{-1}$ , 1480  $\text{cm}^{-1}$ , 1531  $\text{cm}^{-1}$ , and 1665  $\text{cm}^{-1}$  attributed to the C=C stretching in the graphene-based materials [3,12]. In addition, the peaks at 1105

$\text{cm}^{-1}$ , 1169  $\text{cm}^{-1}$ , and 1297  $\text{cm}^{-1}$  represent the vibration mode of the C-O group [16,21], while the peak at 1228  $\text{cm}^{-1}$  corresponds to S=O stretching group [13]. Finally, the peak at 1056  $\text{cm}^{-1}$ , belongs to Fe-OH. These data revealed that the as-prepared carbons possessed a variety of the surface functional groups. Therefore, it can be asserted that the graphitization ratio obtained after the annealing process was high [3].



**FIGURE 6** The FTIR spectra of carbon nanoflakes

#### Conclusion

Carbon nanoflakes were synthesized in one step, with a reasonable yield (48%) from waste tea as a biomaterials source of carbon. Using biobased waste materials for nano-carbon production is a privilege to deduce the impact on the environment caused by the waste stacking. The fabricated nanoflakes have good thermal stability and the nanoflakes formation, therefore, happened as a result of the fusion of the nanoparticles.

The responsible condition of the formation leans on the temperature of decomposition, time of deposition, catalyst type, and the source of the starting material. We believe that the nanoflakes will find a basic application in water treatment after further development which will be done in the near future.

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#### Conflict of Interest

The authors declare that they have no conflict of interest.

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