Carbon Nanotube prepared by catalytic pyrolysis as the electrode for supercapacitors from polypropylene wasted face masks

Wei Yang · Lin Cao · Wei Li · Xusheng Du · Zhidan Lin · Peng Zhang

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Abstract
The massive global consumption and discarded face masks drove by the ongoing spread of COVID-19. Meantime, incineration and landfill discarded face masks would result in severe environmental pollution and infectious hazards. Herein a suggestion to recycle polypropylene waste masks into CNTs by an environmentally friendly and high-added value disposal process was proposed, and which was prepared as supercapacitor electrode materials for energy storage attempting. The CNTs were prepared from waste masks by catalysis pyrolysis with Ni–Fe bimetallic catalysts. Especially, the bamboo-like structure CNT was obtained with Ni/Fe molar ratio is 3. This structure owned a high specific capacitance compared to other standard CNTs. Its specific capacitance could reach 56.04 F/g (1 A/g) and has excellent cycling stability with a capacitance retention rate of the material is 85.41% after 10,000 cycles. Besides, the assembled capacitor possesses a good energy density of 4.78 Wh/kg at a power density of 900 W/kg. Thus, this work provides a sustainable and cost-effective strategy for disposing waste masks into high-value CNT, and their potential application for supercapacitors was also studied and exploited. It would provide a new idea for recycling and utilizing other polypropylene wastes such as medical devices.

KeywordsCarbon nanotubes · Wasted face masks · Catalytic pyrolysis · Electrode · Supercapacitors

Introduction
Currently, wearing face masks is the most effective method to prevent the respiratory transmission of the pandemic COVID-19 [1]. According to statistics, 129 billion masks are discarded globally every month, about 3 million per minute [2]. But it is infeasible for recycling due to the high biohazardous potential of face masks [3, 4]. Hence, the face masks disposal approaches are mainly centralized incineration and landfill to disinfection and prevent reuse [5]. These treatments have caused severe environmental pollution and damaged the sustainability of resources. On the other hand, due to the poor regulation of the disposal waste masks, old and used waste masks are discarded carelessly in the natural environment, which can also bring harmful microplastics to hazard marine and terrestrial ecosystems [6, 7]. Therefore, it is urgent to develop an efficient and environmentally friendly method for disposing of wasted masks with the ever-growing numbers.

Carbonizing of polymers wastes, to date and beyond, is a promising method to recycle wastes and exploit their full potential without excessive consumption of non-renewable resources [8]. Hence, there has been growing interest in carbonizing of polymer wastes for high-value-added products. Variety types of carbon materials, such as amorphous carbon [9], carbon fibers [10], graphene [11], and carbon nanotubes [12, 13], have been researched and disposed of by polymer carbonizing. Among these carbon materials, carbon nanotubes (CNTs) draw a lot of attention due to their unique structure and properties. It has received extensive attention for energy storage applications, also according to the large specific surface area, the unique electrochemical and mechanical properties of CNTs, as one of the most suitable supercapacitors electrode candidates [14, 15]. Wen [16] succeeded in synthesizing CNTs by carbonizing mixed polymers (PP/PE/PS) with CB/Ni2O3 catalysts for supercapacitor electrode materials. Feng [17] also prepared CNTs from the car bumper waste polymers by using the combined
Co(Ac)$_2$-ferrocene/CuCl catalysts. Another CNTs were made from the flexible plastic packaging waste also by carbonization, as a prospective electrode material for the application in supercapacitors [12].

Typically, the process of converting polymers into CNTs is divided into two steps. Firstly, the polymer is degraded into light hydrocarbons as the carbon sources, and then, light hydrocarbons are dehydrogenated and aromatized under catalyst to form CNTs. The carbon sources and catalyst composition are the key factors contributing to the preparation of CNTs from polymers. The light hydrocarbons and aromatic compounds produced by polypropylene waste masks could serve as carbon sources through rapid pyrolysis [18, 19]. In addition, the bimetallic catalysts composed of transition metals with different molar ratios could effectively increase productivity and control the morphology and diameter of the CNTs [19–23]. Generally, the morphology and the diameter of CNTs could greatly affect the specific surface area and other properties of CNTs electrodes. And as preceding reported that the Ni–Fe bimetallic catalyst shows a favorable performance by the synergistic effect of Ni and Fe, which would promote the growth of CNTs, the ratio and composite of Ni and Fe would also adjust the morphology and of CNTs [18, 24–26]. Moreover, a big specific surface area and bamboo-like carbon nanotubes (BCNTs) could be generated by the Ni–Fe bimetallic catalyst, the obtained nanostructure tubes are separated by several cavities in the whole length tubes, and it has benefit applied to electrochemical material [27, 28].

Inspired by the studies above, it is maybe efficient to dispose of the polypropylene face mask wastes and mitigate environmental pollution by carbonizing wasted face masks into CNTs. Meanwhile, the unique properties of waste face masks CNTs could also be applied in supercapacitors to obtain additional economic benefits. Thereupon, herein is proposed that use waste face masks to produce CNTs by cheap Ni–Fe bimetallic catalysts. Different Ni/Fe molar ratios of catalysts would gain the unequal quality, morphology, and the specific surface area of CNTs and thence exhibit various electrochemical properties. Then, the waste face masks CNTs purified by acid are fabrication into electrodes to assembly the symmetrical capacitor, and the electrodes and capacitor electrochemical performance was systematically investigated.

**Materials and methods**

Waste polypropylene face masks were collected from mask recycling sites at communities. Concentrated H$_2$SO$_4$ (98%), HNO$_3$(68%), and Ni(NO$_3$)$_2$·6H$_2$O were obtained from Guangzhou Chemical Reagent Factory (Guangzhou, China). Fe(NO$_3$)$_3$·9H$_2$O, Al(NO$_3$)$_3$·9H$_2$O, NH$_3$·H$_2$O, and the potassium hydroxide (KOH) were delivered from Damao Chemical Reagent Factory (Tianjin, China).

### Synthesis of Ni–Fe-Al$_2$O$_3$ catalysts

Bimetallic catalysts with different Ni/Fe molar ratios were prepared by the co-precipitation method. Different molar ratios of Ni(NO$_3$)$_2$·6H$_2$O, Fe(NO$_3$)$_3$·9H$_2$O, and Al(NO$_3$)$_3$·9H$_2$O were dissolved in deionized water. Wherein the Ni/Fe molar ratio was changed from 1 to 5, and NiFe/Al molar ratio was 1. The metal nitrates solution was stirred at 40 °C, slowly adding 1 M ammonia water dropwise, adjusting the pH to about 8. Then centrifuge to wash the residue to obtain the catalyst precursor, and place the catalyst precursor in an oven at 105 °C to dry overnight. The dried solid was ground into particles smaller than 0.01 mm and calcined at 800 °C for 3 h at a heating rate of 10 °C/min in an air atmosphere. The prepared species catalysts were denoted as NiFeX (X is the molar ratio of Ni/Fe, from 1 to 5, and NiFe/Al = 1).

### CNT preparation

The pyrolysis–catalysis process of wasted masks is carried out in a tube furnace. Before preparation, the waste masks were cut into small pieces. The 10 wt% catalyst was mixed with the 90 wt% mask pieces and then placed in a quartz crucible. In the furnace, the temperature was rapidly raised to the reaction temperature at 20 °C/min from 25 °C to 900 °C. The reaction was kept for 2 h and then gradually cooled to room temperature to obtain CNTs, and the products with different catalysts were named CNT-NiFeX.

### Characterization of CNT-NiFeX

All of the samples were weighed by an analytical balance. And the yield of CNTs is calculated by the following formula [18]:

$$Y_C = \frac{m_y - m_c}{m_m} \times 100\%$$  

(1)

where $Y_C$ is the yield of carbonaceous product, $m_y$ is the weight of CNT-NiFe composite powder after pyrolysis, the $m_c$ is the weight of catalysts, and $m_m$ is the weight of masks. The morphology and microstructures of CNTs were observed by field-emission scanning microscope (FESEM, Zeiss ULTRATM 55) and field-emission transmission electron microscopy (FETEM, JEOL JEM-2100F). Thermogravimetric analysis (TGA, Mettler Toledo TGA/DSC 3 +) was conducted to characterize the decomposition process of samples. The phase structure of CNTs networks was detected by X-ray diffractionmetry (XRD, Rigaku Ultima IV).
instrument) using Cu Kα radiation at 40 kV and 20 mA and Raman spectrometer (HORIBA LabRAM HR Evolution) with an excitation beam wavelength at 532 nm. Element valence was conducted with X-ray photoelectron spectroscopy (XPS, Escalab250Xi, Thermo-VG Scientific, MA, USA). The carbon materials' N2 adsorption/desorption isotherms were acquired at liquid nitrogen temperature (77 K) on a Micromeritics ASAP 2460 instrument.

**Electrochemical measurement**

Before measuring electrochemistry, the sample was purified through acid. Add 200 mg of CNTs to 32 mL mixed acid (H2SO4 and HNO3 of 3:1 by volume ratio) ultrasonically disperse for 1 h and let stand for 24 h. Subsequently, the mixture was washed until a pH of 7 by deionized water and dried the wet CNTs at 70 °C for 12 h to obtain purified CNTs.

The CNTs electrode materials, acetylene black, and poly(tetrafluoroethylene) were mixed in a mass ratio of 80:10:10. Then, the mixture was coated onto a piece of foamed Ni grid and dried at 80 °C for 8 h in a vacuum oven and pressed to obtain a working electrode. Meanwhile, the electrochemical characteristics of CNTs working electrode and supercapacitors were studied via electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), and chronopotentiometry (CP) using CHI 760E workstation (CH Instruments). The cycle stability of the samples was characterized by specific capacitance retention of over 10,000 times of repeated charging and discharging. All electrochemical measurements were performed under a three-electrode cell at room temperature, 6 M KOH solution as the electrolyte, Pt sheet of 1 cm² as a counter electrode, and a Hg/HgO electrode as a reference electrode.

The CP analysis measures the potential difference across the capacitor as a function of time.

The calculation formula for the specific capacitance of the working electrode was based on CP analysis of the change in voltage across time during charge or discharge, and it can be calculated using the following Equation [9]:

\[ C = \frac{I \Delta t}{m \Delta V} \]  

(2)

where C (F/g) indicates the capacitance, I (A) designates the constant discharge current, \( \Delta t \) (s) shows the discharge time, \( \Delta V \) (V) is the discharge voltage range excluding the IR drop, and m is the mass of active material.

The specific capacitance of the symmetric capacitor was calculated as follows [12]:

\[ C_s = \frac{I \Delta t}{m \Delta V} \]  

(3)

where \( C_s \) (F/g) indicates the capacitance, I (A) designates the constant discharge current, \( \Delta t \) (s) shows the discharge time, \( \Delta V \) (V) is the discharge voltage range, and m is the mass of active material.

**Results and discussions**

**Analysis of CNTs quality**

**The yield of carbon**

The effect of Ni–Fe bimetallic catalyst on the carbonization of waste masks was initially investigated. The Ni/Fe molar ratios of the catalyst have a significant effect on the yield of CNTs. Table 1 shows the same mass fraction of catalyst with different Ni/Fe molar ratios; the yield of CNTs has increased from 10.80 wt% (NiFe1) to 26.75 wt% (NiFe4). But when the Ni/Fe molar ratios reached 5, the yield of carbon dropped to 8.94 wt%. The influence of the Ni/Fe molar ratios on the yield is mainly due to the different effects and the interaction of Ni and Fe. Carbon can diffuse along the C/Ni interface to afford whisker carbon [29]. However, the growth ceased if the graphene layers eventually encapsulated the Ni particle completely. Fe can effectively inhibit the catalyst surface from being covered by graphite carbon to maintain catalytic activity via enhancing the oxygen affinity of the catalyst surface and promoting the generation of hydrogen during pyrolysis of the polymer [18, 25]. A proper Ni/Fe molar ratio can promote CNTs’ growth and vice versa. It will inhibit the growth of CNTs, like NiFe1 and NiFe5.

| Sample       | Wasted masks(wt%) | NiFe1(wt%) | NiFe2(wt%) | NiFe3(wt%) | NiFe4(wt%) | NiFe5(wt%) | yield of carbon(wt%) |
|--------------|------------------|------------|------------|------------|------------|------------|---------------------|
| CNT-NiFe1    | 90.0             | 10.0       |            |            |            |            | 10.80               |
| CNT-NiFe2    | 90.0             | 10.0       |            |            |            |            | 22.29               |
| CNT-NiFe3    | 90.0             |            | 10.0       |            |            |            | 26.11               |
| CNT-NiFe4    | 90.0             |            |            | 10.0       |            |            | 26.75               |
| CNT-NiFe5    | 90.0             |            |            |            | 10.0       |            | 8.94                |

Table 1: Carbon yield of catalysts with different Ni/Fe molar ratios
The thermal stability of carbon

TGA curves with oxygen atmosphere are presented in Fig. 1a. The result of samples’ loss weight ratio after oxidation was consistent with the previous carbon yield. The metal residues decreased from 49.53% to 29.20% when Ni/Fe molar ratios increased from 1 to 4, and then, for the CNT-NiFe5, the metal residues reached 54.15%.

The weight loss temperature range of carbon products obtained by pyrolysis catalysis was mainly above 500 °C, and this thermal decomposition temperature connected with filamentous carbon closely [30]. In contrast, the thermal decomposition temperature of amorphous carbon was around 350–500 °C. Hence, the products were mainly filamentous carbon. DTG curve and the amorphous and filamentous carbon content of different samples are shown in Fig. 1b and c. The maximum weight loss temperature of CNT-NiFe3 takes place at 594 °C, and CNTs content was about 92.42%. These indicate that NiFe3 had the most positive effect on the catalytic growth of CNTs and higher thermal stability.

Analysis of CNTs morphology

SEM

Figure 2 shows the SEM morphology of synthesized nanomaterials produced with different Ni/Fe catalysts, indicating a strong correlation between the regulation of the catalyst and the morphology of CNTs. The CNTs’ length can reach tens of microns, as shown in Fig. 2a. And all the CNTs prepared with different Ni/Fe molar ratios display a crimped and entangled filamentous morphology. Nevertheless, the formed CNTs with each catalyst are different in length and size. When the Ni/Fe molar ratios got 3, the diameter of the CNTs was the most uniform. When the ratio deviates significantly from 3, the catalyst will generate large diameters of filamentous carbon, such as CNT-NiFe1 and CNT-NiFe5. The large diameters of filamentous could be related to the weak interaction between the active metal and the catalysts carrier. The weak interaction between them would increase the size of catalysis metal particles and decrease the catalyst activity, resulting in various diameters and shorter lengths of CNTs [31].

TEM

TEM further verified that the filamentous carbon was in a multi-walled carbon nanotube structure. From Fig. 3d, the
catalyst particles could be observed at the tip of the CNTs, which indicates that the growth mechanical of CNTs was tip growth mode [32]. And the interlayer spacing between the graphite layers of CNTs is about 0.34–0.35 nm, which is consistent with the ideal graphite interlayer spacing [33], indicating that the CNTs had high lattice integrity.

Thick filamentous carbon fibers without tubular structure can be observed in Fig. 3a, b, and f. Therefore, the CNT-NiFe1, CNT-NiFe2, and CNT-NiFe5 structures were almost like carbon nanofibers instead of carbon nanotubes. This structure may be caused by the large active metal particles and the low catalytic effect of the catalyst. Besides, the carbon fiber may further reduce the specific surface area of the carbon material. Figure 3g shows the diameter distribution of CNTs produced from different samples. When the molar ratio of Ni/Fe is 3, the diameter CNTs were more uniform, about 10–20 nm.

CNT-NiFe3 has a bamboo-like carbon nanotubes (BCNTs) structure shown in Fig. 3c, unlike the common CNTs with an integral hollow cylindrical structure. Its interior is separated by curved graphene sheets, forming many separated cavities. This structure was considered to have a higher specific surface area and defect density and faster electron transfer rate than common CNTs. It had potential and advantages applied to electrochemical material [27, 34].

**Analysis of CNTs structure**

**XRD**

The crystal structures are shown in Fig. 4a. There were similar crystal compositions between catalysts with different Ni/Fe molar ratios. The diffraction peaks of all as-prepared catalysts could be indexed to Fe-Al and Ni–Al spinel phases. The spinel structures indicated an interaction between the active metals and the alumina carrier. It favored the elevation of active sites on the CNTs tips along with effective diffusion of carbon atoms, promoted CNTs growth, and was conducive to the generation of finer CNTs [31, 35].

The CNT-NiFeX crystalline structure is shown in Fig. 4b. After the pyrolysis catalytic reaction, a broad diffraction peak appeared near 26° (20), which was
graphitic carbon (002) (JCPDS card No. 75–1621) diffraction peak. Meanwhile, two diffraction peaks can be observed at 44.2° and 51.8°, which belong to FeNi$_3$ (111) (JCPDS card No. 88–1715). It was shown that the Fe-Al and Ni–Al spinel in the original catalyst had been deoxidated and converted to FeNi$_3$ alloy. And this alloy could enhance the higher quality CNTs growth [19]. The full width at half maximum (FWHM) of the graphite diffraction peak could reflect the degree of graphitization of CNTs [36]. Well-resolved peaks with a small FWHM indicate high graphitization degree structures in CNTs. By comparing the FWHM of the diffraction peak of graphite (002) from Fig. 4b, the FWHM value of CNT-NiFe3 was 2.104°, the lowest of samples, which means CNT-NiFe3 had the highest graphitization degree and could get better electrochemical performance.
Raman spectroscopy analysis can indicate the degrees of graphitic ordering. For CNTs, the peak at wavelength 1580 cm\(^{-1}\) is called the G band, which associates with the vibration of \(sp^2\) bonded carbon atoms in the graphite layer. The D band, around wavelength 1340 cm\(^{-1}\), originates from the vibration of carbon atoms of disordered graphite or amorphous. The G’ band observed at around wavelength 2700 cm\(^{-1}\) is associated with the process of two-photon elastic scattering [37]. The peak intensity ratios \(I_G/I_D\) and \(I_G'/I_G\) give important information about the graphitization degree of the carbon materials.

All the samples show two Raman peaks at 1580 cm\(^{-1}\) and 1340 cm\(^{-1}\) from Fig. 5. The \(I_G/I_D\) ratios of the CNT-NiFe1 sample to the CNT-NiFe5 sample were 0.91, 1.07, 1.13, 1.08, and 0.76. These results indicated that CNT-NiFe3 exhibits a higher \(I_G/I_D\) ratio with higher degrees of graphitic ordering that could lead to better electronic conductivity and superior electrochemical performance [12]. Meanwhile, CNT-NiFe3 had the highest \(I_G'/I_G\) ratio. The higher ratio of \(I_G'/I_G\) also implies higher CNTs purity [38]. This result is consistent with the morphology results from Figs. 2, 3, and the XRD results from Fig. 4b.

The Application of Synthesized CNTs electrode for Supercapacitors

At room temperature, cyclic voltammetry (CV) was used to characterize the electrochemical performance of CNT samples in a 6 M KOH electrolyte. It can be observed that all samples present approximately rectangular patterns with broad redox peaks at low potentials. The broad redox peaks may originate from the introduction of nitrogen/oxygen-containing functional groups on the surface of carbon nanotubes during mixed acid purification. And the reactions of redox can contribute additional pseudocapacitance. The larger the area enclosed by the CV curve, the higher the specific capacitance of the electrode material [39]. It can be seen from Fig. 6a that the CNT-NiFe3 has a higher response current at the same scanning speed, indicating that it has a higher ratio capacitance.

Capacitance performance was analyzed by the CP curve in Fig. 6b. The CP curve of the CNTs electrode material at a current density of 1 A/g shows an isosceles-like triangle between -0.8 V and 0 V, which proves the electric double-layer capacitance characteristics of the CNTs sample. The specific capacitance performance of the material depends on the discharge time interval. The results show that the CNT-NiFe3 has the largest discharge time, indicating that it has the largest specific capacitance.

To further explore the electrochemical performance of CNT-NiFe3 samples, CNT-NiFe3 samples were analyzed by cyclic voltammetry at a scan rate of 10–200 mV/s. The results showed that the CNT-NiFe3 sample had similar rectangular shapes at different scan rates in Fig. 6c. The rectangular shapes mean that the CNT-NiFe3 sample has excellent magnification capability. Constant current charge and discharge curves of CNT-NiFe3 samples at different current densities are in Fig. 6d. The CNT-NiFe3 working electrode has 59.57 F/g, 56.04 F/g, 51.50 F/g, 49.88 F/g, 48.97 F/g, 47.50 F/g specific capacitance at the current densities of 0.5 A/g, 1 A/g, 2 A/g, 3 A/g, 4 A/g, 5 A/g, respectively; the rate capability was 79.75%.

EIS analyzed the electrochemical performance of the CNT-NiFe3 sample can be seen from Fig. 6e that a typical Nyquist chart consists of a high-frequency area and a low-frequency area. The value of the first intercept of the Nyquist diagram in the high-frequency region and the X-axis indicates that CNT-NiFe3 had a low intrinsic ohmic resistance (\(R_s\)) which is only 0.98 \(\Omega\). The semicircular region corresponding to the ion charge transfer resistance (\(R_{ct}\)), which was formed between the electrolyte and the electrode, was 0.18 \(\Omega\). The Nyquist plot in the low-frequency region was approximately a vertical line, indicating that the CNT-NiFe3 would perform a low equivalent series resistance, good capacitance, and excellent electrochemical performance. At a current density of 4 A/g, the electrode was subjected to 10,000 charge–discharge cycles to test the long-term stability of the material. After 10,000 charge and discharge cycles, the capacitance retention rate of the material was 85.41%, and the Coulomb efficiency was 93.83%. This result shows that the CNT-NiFe3 electrode has good cycle stability.

To study the practical application of CNT-NiFe3 electrodes in energy storage, we composed CNT-NiFe3 electrodes with KOH electrolyte into a symmetric supercapacitor. The CV curve of a capacitor device still
maintains a rectangular shape at different scanning speeds of 10–200 mV/s, which was a characteristic of a typical electric double-layer capacitor. In the Nyquist diagram of the capacitor, the shape was close to vertical, indicating low impedance. The straight-line slope of this related to the diffusion limit of electrolyte ions, and the radius of the semicircle was the charge transfer resistance ($R_{ct}$) of the capacitor. The Nyquist plot in the low-frequency region was approximated by a vertical line and $R_{ct}$ value, indicating the best ion diffusion and fast charge transfer rate performance. After assembling the capacitor device, the charge transfer resistance between the electrode and the electrolyte increases, thereby reducing the specific capacitance of the capacitor. Under different current densities, the capacitor’s constant current charge and discharge curves are shown in Fig. 7c. The assembled specific capacitance is 9.24 F/g (current density 1 A/g). Energy and power density was calculated according to the constant current charge and discharge curve (Fig. 7d). When the power density is 900 W/kg, the energy density is 4.78 Wh/kg. When the power density is 9347 W/kg, the energy density is 3.64 Wh/kg. The long-term stability of the capacitor was tested by 10,000 cycles of charge and discharge at a current density of 1 A/g. The capacitor electrode declined very fast during the first 250 cycles. It may cause by irreversible redox reactions in the charge and discharge curve of the capacitor. After 10,000 cycles, the charge and discharge curve still maintains an isosceles triangle-like shape. The capacitance retention rate of the capacitor is 71.40%, and the Coulomb efficiency is 95.22%.

Why does the CNT-NiFe3 exhibit higher specific capacitance than other CNTs after purification? Ahn [40] found that smaller diameter CNTs have a larger surface area. Thus,
the higher specific capacitance of CNT-NiFe3 can be attributed to its larger specific surface area caused by a smaller diameter. The nitrogen adsorption–desorption of various CNTs was performed using the BET method to determine the specific surface area. Figure 8a shows CNT samples BET isotherms. All of the CNTs showed a hysteresis loop at P/

Fig. 7 a) CV curves of capacitors at different voltage sweep speeds of CNT-NiFe3 capacitor; b) GCD curves of capacitors at different current densities of CNT-NiFe3 capacitor; c) specific capacitance of CNT-NiFe3 capacitor at different current densities; d) energy and power density of capacitors e) EIS curves of capacitors of CNT-NiFe3 capacitor; f) capacitance retention and coulombic efficiency of capacitors after charging and discharging cycles curve of CNT-NiFe3 capacitor

Fig. 8 a) N₂ adsorption/desorption isotherms and pore size distributions of CNTs samples; b) pore size distributions of CNT-NiFe3
In the range of 0.45 ~ 1.0, demonstrating the presence of mesopores (2 ~ 50 nm) structures. The pore size distributions of CNT-NiFe3 shown in Fig. 8b indicate that the product owns well-developed mesopores. This interconnected mesoporous structure is essential for fast electrolyte wettability and ion diffusion. The specific surface areas of different CNT, with Ni/Fe molar ratio from 1 to 5, were calculated to be 118.33, 135.45, 152.29, and 150.78 and 113.92 m²/g, respectively. CNT-NiFe3 did has the largest specific surface area, which was a benefit for constructing conductive networks [41].

Generally, electrode material’s excellent electrochemical performance was owing to higher surface area and functional groups. To further verify the structure of the CNT samples after purification, we analyzed the chemical composition and valence state of CNT-NiFe3 through the XPS spectrum. From Fig. 9, the total XPS spectrum of CNT shows two peaks at 284 eV and 532 eV, corresponding to C1s and O1s. And the peak around 401 eV corresponds to N1s. It indicates that N and O have been introduced on the surface of the CNTs in pickling purification by concentrated sulfuric acid and concentrated nitric acid.

In the C1s high-resolution spectrum, the 284.83 eV peak corresponds to the sp² hybridized graphitic carbon structure, and the 285.53 eV and 289.17 eV correspond to the C–OH and O=C=O structures, respectively. In the O1s high-resolution spectrum, the peak of 532.04 eV corresponds to the C=O structure, 533.70 eV corresponds to the C–O=C–OH structure, and 535.02 eV corresponds to the chemically adsorbed oxygen structure [16]. Two peaks derived from the high-resolution N1s spectrum, two peaks at 400.53 eV and 401.58 eV, were associated with pyrrole and graphite nitrogen [12]. These nitrogen/oxygen-containing functional groups can improve the capacitor’s specific capacitance by contributing to pseudocapacitance and increasing the wettability of carbon materials. Table 2 shows the elemental composition of different CNTs samples analyzed by XPS. Nitrogen-containing functional groups were believed to have the most pronounced influence on capacitance due to their pseudocapacitive contributions [42]. CNT-NiFe3 has

![Fig. 9](image)

**Table 2** Elemental composition of CNTs samples

| Elements | CNT-NiFe1 | CNT-NiFe2 | CNT-NiFe3 | CNT-NiFe4 | CNT-NiFe5 |
|----------|-----------|-----------|-----------|-----------|-----------|
| C (at%)  | 53.26     | 60.1      | 62.57     | 52.18     | 49.23     |
| N (at%)  | 1.29      | 2.94      | 3.31      | 2.78      | 1.65      |
| O (at%)  | 45.45     | 36.52     | 34.13     | 45.04     | 49.12     |
the highest N content of materials, and it may be due to its BCNTs structure. And the trend of N content in the CNTs sample was similar to the specific capacitance, proving that the higher specific surface area and the BCNTs structure together to make CNT-NiFe3 electrochemical performance optimal.

Conclusions
In summary, CNTs were successfully prepared by carboning polypropylene wasted face masks and applied as electrode material in supercapacitors. The results reveal that using Ni–Fe bimetallic catalysts could adjust the yield and morphology of CNTs with different Ni/Fe molar ratios. Significantly, when the catalyst’s Ni/Fe molar ratio reaches 3, it can catalyze waste masks into BCNTs structures with a uniform diameter around 10~20 nm. And the yield of CNT could get 26.11 wt% with high purity and degree of graphitic ordering.

CNT-NiFe3 electrodes can achieve a specific capacitance of 56.04 F/g when the current density is 1 A/g, also has excellent cycling stability with a capacitance retention rate of the material is 85.41% after 10,000 cycles. The symmetrical capacitor assembled from CNT-NiFe3 and KOH has a specific capacitance of 9.24 F/g at a current density of 1 A/g and possesses a good energy density of 4.78 Wh/kg at a power density of 900 W/kg. Due to the small-diameter bamboo-like carbon nanotube structure, allow CNT-NiFe3 to obtain the highest specific surface area and N-doped content after purified. This method obtained a high yield of CNT and excellent capacitance performance of CNTs electrode materials. It provided a novel idea of treatment of polymer medical waste and high value-added recycling of resources.

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Declarations

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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