

Valorising Waste PET Bottles into Carbon Anodes for Li-ion Battery using Ionothermal Carbonisation: A Preliminary Study

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Abstract

Waste PET bottles (WPB) is fast becoming an environmental nuisance and its valorization to carbon anode could be a sustainable method to manage this waste and also develop cheap and high-performance carbon materials for Li-ion batteries (LIBs). Carbonaceous materials derived from WPB were prepared using an ionothermal carbonization (ITC) method in choline chloride urea-deep eutectic solvent system. The ITC-derived materials were subsequently annealed in air to obtain carbonaceous materials. The ITC-derived carbon displayed ultra-high nitrogen doping but lesser carbonization and graphitic ordering compared to the reference carbon material obtained using hydrothermal carbonization (HTC). Therefore, higher temperature annealing/pyrolysis was recommended for the ITC-derived carbon. The HTC-derived carbon was investigated as anode material in LIB with promising electrochemical performance. The LIB displayed stable reversible capacity of about 130 mAh/g at a current density of 0.1 mA/g after 20 cycles and an increasing Coulombic efficiency that reached 98% after the 50th cycle. This work shows that a facile and sustainable synthesis method could be used to produce cheap activated carbons with potential applications in energy storage systems such as LIBs.

Introduction

The rapid population growth and depletion of fossil fuels have led to unprecedented increase in renewable energy demands globally. Since these renewable energies are intermittent sources of energy, there is a need for a backup system that will serve as excess energy storage that will make it available when needed for an uninterrupted energy supply. Presently, the most effective energy storage system (ESS) is a lithium-ion battery (LIB); which has become a key component in portable electronic devices and electric vehicles owing to its unparalleled high energy density and power density¹⁻³. Currently, many ESS technologies use electrode materials obtained from unsustainable sources such as fossil-based carbon sources making them costly and environmentally unsustainable⁴. As a result, the research community has been exploring carbon materials derived from cheap renewable resources using eco-friendly approaches as electrode material in ESS including LIBs^{1,5-8}.

Post-consumer products such as waste PET is a threat to the environment hence a sustainable waste management that involves waste PET valorization to functional materials for various applications including energy storage is highly needed to achieve a circular economy. Waste plastic can serve as cheap and sustainable starting-materials for the synthesis of porous activated carbons (ACs)⁹⁻¹². Also, the high amounts of carbon coupled with the small amounts of impurities in PET make it an attractive precursor for the preparation of ACs¹⁰. The traditional method of preparing ACs from fossil-based sources is through direct pyrolysis which is not eco-friendly^{10,12,13}. Also, this method involves the use of expensive and toxic precursors which often yield ACs with limited functional groups thereby limiting their performance in many applications such as electro-catalysis. Other disadvantages with this method is the low yields of carbon products and often excessive washing process to remove residual chemicals/poisons and to prevent corrosion of the reaction system.

In quest for new eco-friendly methods for the synthesis of carbon materials from bio-resources, green solvents are being explored. In this regard, ionic liquids (ILs) which has low-vapour pressure, non-flammable, recyclable, and non-volatile have been widely used ^{6,7}. However, the high cost, complex preparation and separation, low biodegradability have been the major drawbacks to the large-scale application of ILs ¹⁴. As a result, deep eutectic solvents (DES), analogues of ILs, have been used as alternative green solvent to ILs in preparing carbon materials obtained from bio-resources due to their higher biodegradability, low-costs, eco-friendliness, facile methods of synthesis and recycling ¹⁵. Furthermore, the hydrothermal carbonization (HTC) has been developed as an alternative carbonization method for the preparation of carbonaceous materials under mild conditions (i.e., < 10 bars and 180-220 °C). HTC offers a facile, high yield and energy-intensive method capable of producing functional carbon materials which can be achieved through doping of heteroatoms by the addition of non-aqueous solvents like DES ^{6,7,16}. This thermochemical method which is referred to as ionothermal carbonization (ITC), analogous to HTC, could also facilitate the production of carbonaceous materials with high specific surface areas and pore volumes ⁷.

Therefore, it is plausible to use waste PET pre-treated with choline chloride-urea DES as precursors for ionothermal carbonisation not only to obtain ACs but to reduce the activation temperature used, thereby reinforcing the green credentials of the process. Moreover, the process allows heteroatomic doping of nitrogen element into the carbon material surface which has been shown to improve electrochemical performance ⁷. In this communication, the potentials of hydro- (iono-) thermal carbonization in preparing activated carbons from waste PET and its subsequent use as anode in LIBs are presented.

Experimental Section

Ionothermal Carbonization (ITC)

Waste PET bottles were shredded to particle sizes (between 4 and 5 mm), washed with water, air dried and labelled PET-P. ACs were prepared by ITC of the PET-P following the modified procedures of Baccour et al. ⁷. Initially, a DES made from choline chloride urea (CU) was prepared and appropriate amount of PET-P was dispersed in 20 mL CU-DES at room temperature. The mixture was loaded into a Teflon lined stainless steel autoclave (250 mL volume), placed in an oven and was heated at 200°C for 3 hr. The as-obtained carbon material, called ionochars, was filtered, washed several times with de-ionised water until a neutral pH was obtained, and then dried in an oven overnight at 80°C. The dried ionochar was ball-milled at 500 rpm for 3 hr and labelled PET-CU-ITC3. After this, PET-CU-ITC3 was annealed at 400°C for 2 hr in air. The obtained materials were ball-milled into fine powders at 500 rpm for 3 hr and it was labelled PET-CU-ITC-A. As a comparison, ITC synthesis was undertaken following the same procedure above except that water (instead of CU-DES) was used as the solvent. Hydrochar (PET-H₂O-ITC3) was obtained which was subjected to annealing and ball-milling to obtain a black carbonaceous material labelled PET-H₂O-ITC-A.

Characterisation

The product yield of the ionochars, hydrochars and carbon materials were determined using the equation below:

$$\text{Activation yield} = \frac{\text{mass of carbonised material (g)}}{\text{mass of raw material (g)}} \times 100$$

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The elemental analysis of C, H, N and S were done on the obtained products using a Vario micro cube elemental analyser in which the C, H, N and S were determined using the thermal conductivity detector while the value of S was detected by infrared detector. The crystal structure of the products was measured by STOE laboratory diffractometer equipped with a Cu source and with transmission geometry. In addition, a HORIBA LabRam Evolution HR HeNe-Laser (633 nm, 17 mW) Raman microscope was used to determine the degree of order of the carbons in these materials. The functional groups in the final AC material was determined using a Bruker Tensor 27 FT-IR spectrometer attached to STA and operated under Ar-atmosphere. Scanning electron microscopy (SEM) images were acquired on a SEM-EDX Merlin microscope (Zeiss GmbH).

Electrochemical Tests

The electrochemical measurements were carried out with a VMP3 multichannel potentiostat (Biologic, France) operating at 25°C. The anode was prepared by coating a mixture of the AC material (80%), carbon black (10%), and polyvinylidene fluoride binder (10%) in 1-methyl-2-pyrrolidone solvent on a Cu foil, which was dried in the vacuum oven at 80°C overnight. This coated electrode film, with a thickness of 150 µm, was cut into a disc (12 mm in diameter) which was used as the working electrode (anode). The electrochemical properties of the materials were tested using the 2032 coin-type cells which were assembled in a glovebox (MBraun) flushed with argon gas. To assemble half cells, the as-prepared anode was used as the working electrode and lithium metal foil as the counter electrode which were separated by two Celgard 2325 (16 mm), while BASF LP30 selectylite (1 M LiPF₆ in a 1:1 volume ratio of ethylene carbonate and dimethyl carbonate) was used as the electrolyte. The galvanostatic charge-discharge tests and cycling performance were performed in the voltage range of 0.01 and 2.8 V (vs Li/Li⁺) using a current density of 0.1 A/g.

Results And Discussion

Iono- (-hydro) thermal carbonization of waste PET bottles

The scheme of the ionothermal carbonisation (ITC) for the synthesis of activated carbon (AC) from waste PET bottles is presented in Fig. 1.

Most reports have used chemical activation method for the preparation of AC from waste PET bottles which is usually undertaken in a single-step with pyrolysis^{10-12, 17}. This method is not sustainable and eco-friendly, hence a green and a more cost-effective synthesis route was adopted in this research. In the ITC process (as shown in Fig. 1), a low temperature carbonisation of 200°C was used with the duration varied between 3–24 hr. A viscous liquid (ionochar) was formed after this low temperature carbonisation was applied to CU-DES impregnated PET-P under autogenous pressure. When the reaction was done in water (HTC) in longer durations than 3 hr, white crystals were obtained which were found to be pure terephthalic acid crystals. Furthermore, AC material were obtained when the ionochar (PET-CU-ITC3) and hydrochar (PET-H₂O-ITC3) carbonised for 3 hr were subjected to annealing at 400°C for 2 hr. It was found out that when these same precursors were pyrolysed at similar and higher temperatures, they sublimed. Therefore, this observation reinforced the need to either stabilise PET using low temperature annealing before pyrolysis; or anneal it in air at a lower temperature for a longer duration to obtain turbostatic carbons as implemented in this work. This low temperature annealing method in air to produce AC from waste PET could offer a cheaper and a more facile method compared to the traditional pyrolysis method in N₂/He gases. Generally, the ITC methods presented a very facile and low-cost method of obtaining nitrogen-rich ACs from PET that also avoids the generation of toxic waste. Also, since the CU-DES contained no inorganic specie, no post-synthesis washing of the products was undertaken unlike ionothermal pyrolysis method which use toxic chemicals as activating agents^{10,18}.

Carbonization Conditions, Product Yield And Elemental Analysis

Table 1 gives the carbonization conditions, product yield and elemental analysis of obtained carbonaceous materials from the iono- (-hydro) thermal carbonization of waste PET bottles. The product yields of the ITC obtained samples were much lower than those obtained from the HTC method. For example, PET-H₂O-ITC6 and PET-CU-ITC6 samples gave product yields of 75% and 36%, respectively. This substantial reduction in product yields of the ITC samples could be due to the use of CU-DES solvent. The hydrolysis of the PET samples is expected to be more vigorous with CU-DES compared to water resulting in lesser product yields. This is evident in the decreasing product yields of the ITC samples with increase in carbonization times while the product yields of the HTC samples increased linearly with carbonization times. PET-CU-ITC3 and PET-CU-ITC24 samples gave product yields of 56% and 27% while PET-H₂O-ITC3 and PET-H₂O-ITC24 gave product yields of 68% and 83%, respectively. Furthermore, the product yields of PET-H₂O-ITC3 and PET-H₂O-ITC-A were 68% and 17%, respectively. Expectedly, annealing of PET-H₂O-ITC3 in air further reduced the yields of the final material probably due to structural rearrangement in the materials with the loss of volatiles and gases such CO, CO₂, NO₂, CH₄, etc.¹⁹. However, it is important to note that the PET-H₂O-ITC3 showed no formation of activated or pseudo-graphitic carbons as seen with PET-H₂O-ITC-A product.

The elemental analysis of the samples obtained from the iono- (hydro-) thermal carbonization of waste PET are also given in Table 1. Generally, increasing the carbonization times resulted in slight increase in

the carbon content and corresponding decrease in the hydrogen content. PET-H₂O-ITC3 gave 58.01 wt% C and 3.76 wt% H while PET-H₂O-ITC24 gave 59.17 wt% C and 3.29 wt% H. Similar trend has also been reported in previous study on the evolution of C and H contents with carbonisation of lignocellulosic materials ¹⁹. The elemental results show that the carbon content of the obtained materials varied marginally between 58.01 and 61.78 wt% with no significant increment in C between 3–24 hr carbonization time. Therefore, 3 hr carbonisation time was used for the ITC method before annealing. PET-H₂O-ITC-A gave 72.06 wt% C compared to the 58.01 wt% C that was found in its precursor (PET-H₂O-ITC3) which shows the additional carbonization of the material using annealing in air. Annealing at this same temperature did not show any effect on the carbon content of the PET-CU-ITC-A sample compared with PET-CU-ITC3 sample. This shows that the CU-DES impregnated PET samples might require higher heat treatments. In addition, the ITC samples showed ultra-high amounts of nitrogen in the final products compared to the HTC samples. The high nitrogen content is due to the CU-DES with urea component facilitating the doping of nitrogen into the carbon structure ²⁰.

Table 1

Carbonization conditions, product yield and elemental analysis of obtained carbonaceous materials from the hydro- (iono) thermal carbonization of waste PET bottles.

Sample	Temperature (°C)		Yield (%)	Ultimate analysis (weight %)			
	Hydro- (iono-) thermal carbonisation	Annealing (A)		C	H	N	S
PET-H ₂ O-ITC3	200	-	68	58.01	3.76	0.24	0.00
PET-CU-ITC3	200	-	56	58.68	5.63	16.89	0.08
PET-H ₂ O-ITC6	200	-	75	58.51	3.58	0.23	0.10
PET-CU-ITC6	200	-	36	59.50	5.21	16.04	0.05
PET-H ₂ O-ITC24	200	-	83	59.17	3.29	0.17	0.10
PET-CU-ITC24	200	-	27	61.78	5.91	15.15	0.04
PET-H ₂ O-ITC-A	200	400	17	72.06	2.72	0.00	0.00
PET-CU-ITC-A	200	400	13	58.77	2.58	11.87	0.00

Note: x in PET-H₂O-ITCx means the duration (in hr) of iono- (hydro-) thermal carbonization. The ITC gave viscuous brownish liquid products while the HTC gave solid particles which had whitish to milkish colouration. The yield of the samples that were subjected to ITC and then annealing (i.e. PET-H₂O-ITC-A and PET-CU-ITC-A) were calculated from the yield obtained after ITC and not from the starting material (PET-P).

Xrd Analysis Of Products Obtained From Itc Of Waste Pet

The XRD analysis was done to investigate the degree of order and evolution to a turbostratic-type carbon in the final materials. Figure 2 gives the XRD diffraction patterns obtained for some of the iono- (hydro-) thermal carbonization products (a) and the further annealed products (b). The XRD of PET-H₂O-ITC3 (Fig. 2a) shows an evolution to a terephthalic acid diffraction peaks which was fully crystallised in the PET-H₂O-ITC24 material. This means the HTC of waste PET bottles resulted in the depolymerisation of the material into its main precursor namely terephthalic acid. The PET-CU-ITC24 gave diffraction peaks that were neither that of terephthalic acid nor of the desired activated carbon material. However, when PET-H₂O-ITC3 material was subjected to annealing at 400°C (PET-H₂O-ITC-A), the XRD peaks (Fig. 2b) show two characteristic broad and weak peaks i.e. the 002 and 100, typical of amorphous, disordered

turbostratic-type carbons²¹. The broad 002 peak can be attributed to interlayer reflection and it corresponds to the amorphous structure in this carbon²². The weak 100 peak can be attributed to in-plane scattering which is typical of a turbostratic-type carbon²³.

Similarly, when PET-CU-ITC3 material was subjected to annealing at 400°C (PET-CU-ITC-A), it displayed same XRD peaks as PET-H₂O-ITC-A carbon but with lots of impure phases as shown in Fig. 2b. This impure phases might be due to the presence of CU-DES in the material which might require higher temperature treatment for better crystallinity. Also, the PET-H₂O-ITC-A carbon show a more pronounced 101 peak, indicating an increased structural order in this carbon⁶. Therefore, PET-H₂O-ITC-A carbon was the only carbon used for further characterization in this work while the CU-DES impregnated samples will be subjected to higher temperature treatments and pyrolysis in future studies.

Morphology, Raman And Ftir Spectra Of Pet-ho-itc-a Activated Carbon

The Raman spectrum of the PET-H₂O-ITC-A activated carbon is shown in Fig. 3a. The spectrum shows two characteristic carbon D and G peaks around 1350 and 1589 cm⁻¹ which can be assigned to the disordered and graphitic carbon bands, respectively⁵⁻⁷. The small I_D/I_G ratio (0.80) of the PET-H₂O-ITC-A carbon indicates the presence of an amorphous carbon structure with some defects, which agrees well with previous results^{6,7}. The FTIR spectrum of the PET-H₂O-ITC-A is shown in Fig. 3b. It shows the presence of many functional groups on the carbon surface which could aid the electrochemical performance of this carbon anode in LIBs. The bands in the range of 1000–1200 cm⁻¹ indicate the presence of C-O stretching vibration associated with oxygen containing functional groups in alcohols, ester or ether groups. The band at 1600 cm⁻¹ is due to the aromatic C = C stretching vibrations while the one at 1711 cm⁻¹ is due to the existence of carbonyl/carboxyl groups^{6,24}. The weak band at 2858 cm⁻¹ and the strong band at 2970 cm⁻¹ can be assigned to aliphatic C-H stretching vibrations. The SEM images in Fig. 3c and 3d show particles of varying shapes and having a wide particle size distribution.

Electrochemical Performance Of Pet-ho-itc-a Anode In Lib

The PET-H₂O-ITC-A was used as an active material to prepare the anode in a LIB. The galvanostatic charge-discharge profiles of the PET-H₂O-ITC-A anode for the 1st, 2nd, 10th and 50th cycles are shown in Fig. 4a. The LIB was cycled between 0.01–2.8 V (vs. Li/Li⁺) at 25°C using a current density of 0.1 A/g with no initial activation applied at a lower current density. The quantitative charge-discharge capacities and Coulombic Efficiency (CE) data for the 1st, 2nd, 30th, and 100th cycles are given in Table 2. The PET-H₂O-ITC-A anode give a 1st cycle discharge capacity and CE of 919 mAh/g and 16%, respectively. This poor initial CE represents a large irreversible capacity loss which is characteristic of carbon anodes which is due to the irreversible degradation of the electrolyte and subsequent formation of SEI⁷. In addition, the

disordering in these amorphous carbons can also facilitate the trapping of Li⁺ in the voids, thus leading to the large capacity loss in the 1st cycle²⁵. After the 1st cycle, there is a gradual stabilization of the carbon anode which can be seen in the reduction of the reversible capacity and capacity loss along with a concomitant increase in the CE with subsequent cycling. For example, the 50th cycle display a discharge capacity and CE of 115 mAh/g and 98%, respectively. The corresponding galvanostatic cycling performance of PET-H₂O-ITC-A anode is given in Fig. 4b. The discharge capacities are significantly higher than the charge capacities in about the first 20 cycles which is evident in the poorer CE for these cycles. In the initial cycle at 0.1 A/g, capacities of 919 and 150 mAh/g were obtained during lithium uptake and release, respectively. On subsequent cycling, the electrode shows stable capacity with better capacity retention.

Table 2

Charge-Discharge capacities and Coulombic Efficiency at the 1st, 2nd, 10th and 50th cycles for PET-H₂O-ITC-A anode in LIB

Discharge capacity at 1st cycle (mAh/g)	Reversible capacity (mAh/g)			Coulombic Efficiency (%)			
	2nd cycle (mAh/g)	10th cycle (mAh/g)	50th cycle (mAh/g)	1st cycle (mAh/g)	2nd cycle (mAh/g)	10th cycle (mAh/g)	50th cycle (mAh/g)
919	211	140	115	16	67	90	98

Conclusion

The valorization of waste PET bottles into activated carbon using the sustainable and facile hydro-(iono-) thermal carbonization method has been demonstrated in this study. Even though annealing in air at a low temperature was applied to treat the ITC and HTC derived carbons, yet they showed pseudo-graphitic carbon from the XRD analysis. The ITC method using CU-DES resulted in poorly carbonized products compared to the carbons obtained from HTC, however the ITC-derived carbons had high amounts of doped nitrogen. This result shows that higher temperature annealing or pyrolysis in a controlled atmosphere could help to increase graphitic ordering in these carbons and also increase their yields. The HTC derived carbons showed good potentials as anode in LIB. Further optimization of the ITC method for the valorization of waste PET bottles is recommended. Several experimental parameters should be evaluated such as post-synthesis washing of the as-prepared carbons, the use of varying ratios of the DES components and other DES systems, and the type of heat treatment on the carbonaceous material which could influence the chemical, textural, structural and electrochemical performance of the carbon materials.

Declarations

Conflicts of Interest

There are no conflicts of interest to declare.

Data Availability

All data generated or analyzed during this study are included in this published article

Acknowledgments

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Figures

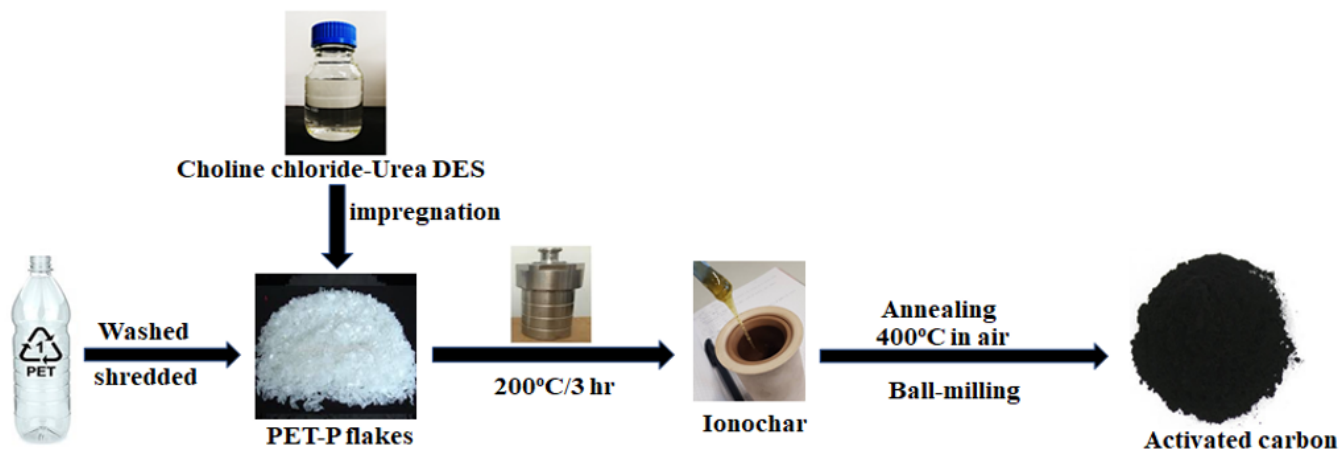


Figure 1

Schematic of the ionothermal carbonisation waste PET to obtain activated carbons.

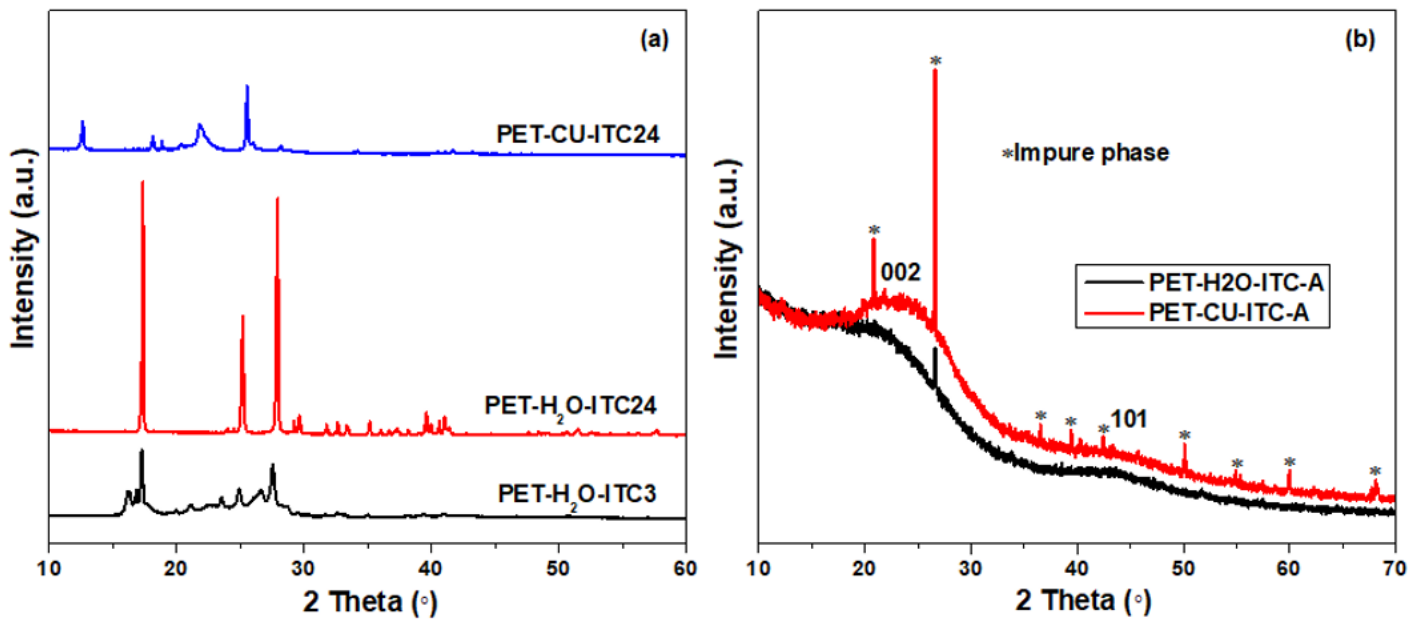


Figure 2

XRD (a) Hydro- (iono-) thermal carbonization products (b) annealed products

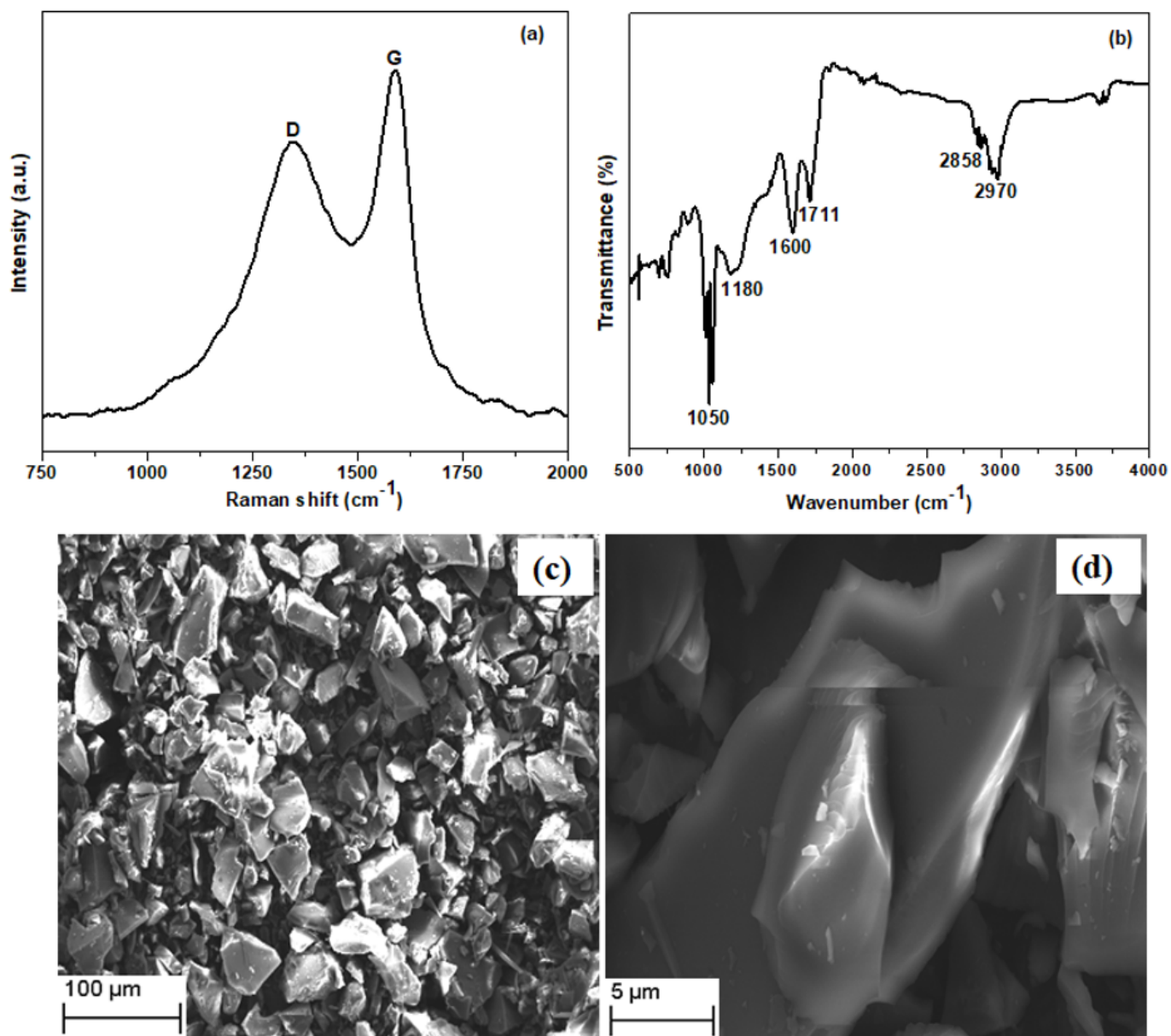


Figure 3

PET-H₂O-ITC-A activated carbon (a) Raman and (b) FTIR spectra; SEM images at a magnification of (c) 109 x (d) 2k x

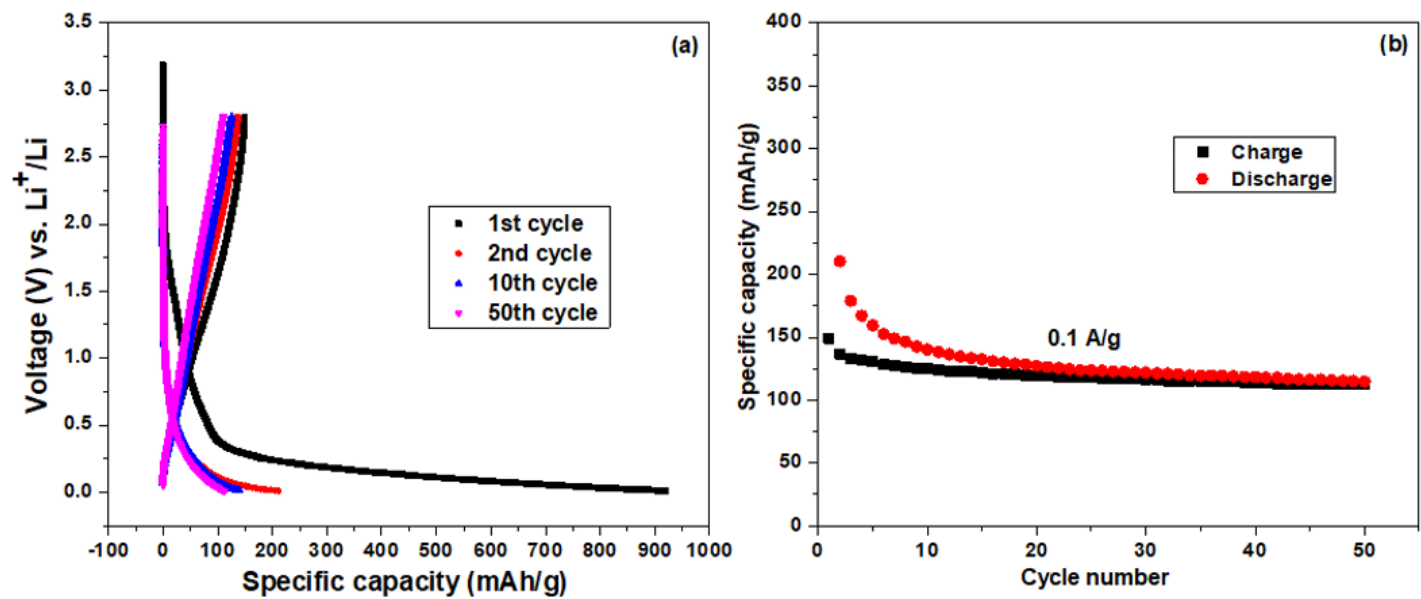


Figure 4

Electrochemical performance of PET-H₂O-ITC-A carbon anode (a) Galvanostatic charge-discharge profiles (b) cycling performance