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# Preparation of Super Activated Carbon from Various Parts of Nelumbo nucifera and its Application as Electrode Material in Supercapacitors

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In this study, five parts of *Nelumbo nucifera* (the lotus stem, lotus leaf, lotus seedpod, lotus seed and lotus root) were used to prepare super activated carbon with an ultra-high specific surface area. All five activated carbons were assembled into button-type supercapacitors as the electrode material. Constant current charge and discharge tests showed that under the same current density and charge/discharge voltage, the activated carbon prepared with the lotus stem has a relatively high specific capacitance of 238.52 F/g, a specific energy density of 13.7 Wh/kg and a voltage drop of less than 30 mV. After 1000 cycles, the specific capacitance remains at 89%, and the energy charge efficiency is 81%. Moreover, different MnO<sub>2</sub> contents (mass fractions) were loaded on the activated carbon of the lotus stem to fabricate electrode materials for a supercapacitor. The test revealed that among the tested supercapacitors, the supercapacitor with 10% MnO<sub>2</sub>-loaded activated carbon from the lotus stem as an electrode material has the highest specific capacitance (323.66 F/g), which is 46.23% higher than that of a pure carbon supercapacitor, and the material resistance of this load ratio is small. The electrode prepared with a lotus stem as the carbon source has good electrochemical performance and is expected to become a new source of carbon materials for supercapacitors and low-cost energy storage devices.

Keywords: supercapacitor; Nelumbo nucifera; specific energy, specific capacitance

# **1. INTRODUCTION**

Supercapacitors, which are also known as electrochemical capacitors, are a new type of energy storage device. They have a high power density, good reversibility, and long life cycles [1-5]. The selection of good electrode materials is a key factor to improve the capacitance of supercapacitors [6].

Carbon-based materials such as graphene [7], carbon nanotubes [8], and activated carbon have become the first choice for electric double-layer capacitor (EDLC) electrodes due to their good pore

size distribution, high specific surface area, and physical as well as chemical stability [9-10]. Choosing activated carbon as the electrode material for EDLCs has obvious advantages including its high porosity, large specific surface area, good corrosion resistance, low cost and accessibility for largescale commercial use [11].

At present, most commercial activated carbons are produced from petroleum and coal [12], which makes them expensive and harmful to the environment. However, biomass materials are becoming increasingly less expensive due to their low cost, renewability, ready availability, and porous structure, and they are beneficial to the environment [13]. In recent years, the use of biomass materials as the precursors of activated carbon to prepare EDLCs has satisfactory electrochemical performance and has attracted widespread attention. In particular, plant biomass, such as oil palm from empty fruit branches [14], coffee bean waste [15], sugar cane bagasse [16], willow catkin [17], tobacco [18], cotton stalks [19], pomelo peel [20], tea leaf waste [21], and broad beans, is used [22]. Plant biomass precursors are of great interest, because of their wide availability, low cost, and rich contents of various heteroatoms and functional groups, which can reduce the charge transfer resistance and improve the wettability. Therefore, these precursors can improve the capacitor performance [23].

*Nelumbo nucifera* is a renewable precursor of plant biomass material [24]. Lotus is a type of seed plant. Seed plants have many organs such as roots, leaves, stems, flowers, fruits and seeds. *Nelumbo nucifera* is an aquatic herb that has grown for many years. Each part of the lotus has a unique structure and rich porous structure, which is well developed for absorbing nutrients and ions. Using each part of a lotus as an activated carbon will have a high specific surface area and more pores for adsorbing ions [25].

Previous studies have found that the pore size distribution of biomass activated carbon highly depends on the original form of the plant organs [26], and activated carbon prepared with more micropores, mesopores or macropores as precursors has excellent electrochemical properties [27]. The five parts of the *Nelumbo nucifera* (the lotus root, lotus stem, lotus leaf, lotus seedpod, and lotus seeds) were directly pyrolysed, activated, and assembled into a button-type supercapacitor for electrochemical testing. The lotus stem was found to be the most excellent electrode material precursor among the five parts of *Nelumbo nucifera*.

## 2. EXPERIMENTAL

#### 2.1 Material and Preparation of Activated Carbon

*Nelumbo nucifera* was obtained from Jiangxi Province in China, along with hydrochloric acid (AR), potassium hydroxide (AR), and ethanol (AR). The battery shell and separator paper were provided by the Zhong Sheng (Chang Xing) Lithium Battery Material Co., Ltd., China. Nickel foam was provided by the Yong Chang Shuo (Xia Men) Electronic Technology Co., Ltd., China.

Activated carbon was prepared by carbonization and KOH activation. The dry, clean, and sheared biological materials were carbonized at 300°C. Then, solid potassium hydroxide was added to the carbonized biological material in a ratio of 1:3 and placed in a muffle furnace (840° C, 10

minutes), where it was activated [28]. It was washed with 1:9 HCl several times to remove inorganic salts and subsequently washed with distillation to a neutral pH of 6~7. Finally, the activated carbon was dried overnight in a vacuum drying oven at 120°C to obtain an activated carbon material using *Nelumbo nucifera* as the carbon source [29]. AC derived from different parts of Nelumbo nucifera were denoted as  $AC_{ls}$ ,  $AC_{ll}$ ,  $AC_{lsp}$ ,  $AC_{ls}$  and  $AC_{lr}$ , respectively. Finally, a certain amount of manganese dioxide is loaded into the best part of Nelumbo nucifera . MnO<sub>2</sub>-loaded activated carbon from the lotus stem is donated as  $MnO_2/AC_{ls}$ .

#### 2.2 Structure and Characterization

The microscopic morphology and structure were observed by a field-emission scanning electron microscope (FESEM, EVO-18). Nitrogen adsorption and desorption isotherms were obtained at 77 K on the micromeritics ASAP analyser. The Brunauer-Emmett-Teller model was performed to determine the specific surface area. The pore volume was estimated from the adsorbed amount at a relative pressure P/P0=0.99. The pore size distribution (PSD) was analysed by a non-local density functional theory (NLDFT) model assuming the cylindrical pore geometry of the adsorption data.

## 2.3 Preparation of the Supercapacitor and Electrochemical Measurements

The supercapacitor was prepared as follows: acetylene black, the activated carbon material and PTFE fine powder were mixed in a certain proportion and subsequently air dried on round nickel foam [29]. Then, the electrode plate preparation was complete. An appropriate amount of electrolyte (6 mol/L KOH) was added to the electrode plate, and the membrane was immersed in the electrolyte for several minutes. The excess electrolyte was then dried. The plate-membrane-plate was pressed into the battery box, and the tablet was pressed with a certain pressure to complete the preparation of the supercapacitor [28].

The electrochemical measurement of each supercapacitor was performed on an electrochemical workstation (CHI600d, Shanghai, China) using a 3-electrode system (the (Hg/HgO) electrode was the reference electrode, platinum foil was the counter electrode, and *Nelumbo nucifera* activated carbon in each part was the working electrode) in 6 mol/L KOH as the electrolyte at room temperature. The electrochemical performance was characterized by cyclic voltammetry (CV), constant-current charge and discharge and electrochemical impedance spectroscopy (EIS). The cyclic voltammetry curve was obtained at a scan rate of 5-10 mV/s, and the electrochemical impedance spectroscopy was originally measured at a frequency of 10 mHz - 10 kHz when the alternating current amplitude was 5 mV. The specific capacitance of the electrodes was calculated based on the constant-current charge/discharge test as follows:

$$C = \frac{I \cdot \Delta t}{m \cdot \Delta V} \quad (3 \ Electrodes) \qquad (1)$$
$$C = \frac{4I \cdot \Delta t}{m \cdot \Delta V} \quad (2 \ Electrodes) \qquad (2)$$

where m (g) is the mass of AC in both electrodes;  $\Delta V$  (V) is the voltage change excluding the IR drop (V) within the discharge process; I (A) is the current density; and  $\Delta t$  (s) is the discharge time [30].

The energy density (E, Wh/kg) and power density (P, W/kg) of the 3-electrode test were calculated as follows:

$$E = \frac{1}{2}C \cdot \Delta V^2 \cdot \frac{1}{3.6}$$
(3)  
$$P = \frac{E \cdot 3600}{\Delta t}$$
(4)

where E (Wh/kg) is the specific energy; C (F/g) is the specific capacitance based on each part of active material in both electrodes;  $\Delta V$  (V) is the voltage change excluding the IR drop (V) in the discharge process; and  $\Delta t$  (s) is the discharge time [31].

# **3. RESULTS AND DISCUSSION**

- 3.1 Selecting the Best Carbon Source in Nelumbo nucifera
- 3.1.1 Constant-current charge/discharge test



Figure 1. Constant current charge and discharge of supercapacitors made of each part of *Nelumbo nucifera* 

Fig. 1 shows the constant-current charge/discharge test for the following 5 parts: AC<sub>ls</sub>, AC<sub>ls</sub>, AC<sub>ls</sub>, AC<sub>ls</sub>, AC<sub>ls</sub>, AC<sub>ls</sub>, AC<sub>ls</sub> and AC<sub>lr</sub>. The constant-current charge discharge of the supercapacitors with 6 mol/L KOH as the electrolyte at current densities of 0.1A/g. The curve of each part of the electrode has a similar shape, which indicates a nearly symmetrical characteristic due to the nearly equal charging and discharging times [32]. However, the curves have different charge and discharge times; a wider bottom of the curve indicates that more electrons and electrolyte ions are charged and discharged, and a greater capacitance is generated [33]. The supercapacitors with the lotus stem as the carbon source has the widest bottom of the curve, longest discharge time, and largest specific capacitance [34].



**Figure 2.** (a) Specific capacitance of each part at 1.2 V and 0.1 mA; (b) schematic of the voltage drop of each part at 1.2 V and 0.1 mA; and (c) specific capacitance of each part at 1.2 V and 0.1 mA of the manganese dioxide/activated carbon composites.

Figure 2a shows the average specific capacitances of the five *Nelumbo nucifera* parts (AC<sub>ls</sub>, AC<sub>ll</sub>, AC<sub>ls</sub>, AC<sub>ls</sub> and AC<sub>lr</sub>), which are 278.316 F/g, 164.832 F/g, 217.146 F/g, 132.334 F/g, and 243.581 F/g; the supercapacitor with the lotus stem as the carbon source has the largest capacitance. Figure 2b shows the voltage drop graph of each part; the lotus stem has the lowest voltage drop, and the average pressure drop of the lotus stem is 28.125 mV, which is significantly better than those of the other parts. Compared with those prepared by other biomass materials, Yu et al. [35] prepared electrode materials from corn stalks, it only exhibited the maximum specific capacitance of 140F/g,

PAC4[36] exhibited specific capacitance value 185 F/g, the specific capacitance of the GGAC [37] reaches to 178 F/g, the specific capacitance of each part of Nelumbo nucifera is larger.

The specific energy should not be ignored in selecting the perfect electrode material. The formula for calculating the specific energy is as follows:

$$\mathbf{E} = \frac{1}{2}C \cdot \Delta V^2 \cdot \frac{1}{3.6}$$

where the E (Wh/kg) is the specific energy; C (F/g) is the specific capacitance based on each part of the active materials in both electrodes; and  $\Delta V$  (V) is the voltage change excluding the IR drop (V) in the discharge process [31]. Figure 2c shows that the supercapacitor composed of the lotus stem as the carbon source has the largest average specific energy density, which is 13.7 Wh/kg. L.et al synthesized HLPC exhibited a high specific capacitance of 342 F/g at 0.2 A/g, but HLPC delivered a maximum energy density of 9.4 Wh/kg in the same KOH electrolyte [20].

#### 3.1.2 Comparison with other biomass-derived electrodes

#### 3.1.2 Comparison with other biomass-derived electrodes

		CDDT	a : a			<b>C</b> 11	
Material	Electrolyte	SBET	Specific	Energy	Capacity retention (%)	Cell	Reference
		$(m^2/g)$	capacitance	density		configuration	
			(F/g)	(Wh/kg)			
waste tea	KOH	2245 ~	330	_	92% over 2000 cycles	3E	[21]
leaves		2841					
tobacco rods	KOH	$1866 \sim$	286.6	31.3	96% over 1000 cycles	3E	[18]
		2115					
sugar cane	KOH	$145 \sim 805$	270		Almost 100% over 1000	3E	[16]
bagasse					cycles		
waste stiff	KOH	2523	235	7.9	94.5% over 4000	2E	[33]
silkworm					cycles		
shiitake	KOH	2988	306		95.7% over 15000	3E	[39]
mushroom					cycles		
willow	KOH	$1533 \sim$	298	_	98% over 1000 cycles	3E	[17]
catkin		1936					
pomelo peel	KOH	2725	342		98% over 1000 cycles	3E	[20]
Camellia	KOH	1935	266.374		91.3% over 5000	3E	[25]
oleifera shell	H2SO4				cycles		
lotus stem	КОН	2389.7	278.316	13.7	89% over 1000 cycles	3E	This work

Table 1. Comparison of the electrochemical performance and S<sub>Bet</sub> of biomass-derived electrodes

Table 1 compares the electrochemical performances of biomass-derived electrodes in previous reports. Compared to other biomass electrode materials, the capacitance and energy density of the lotus stem biomass electrode are 278.316 F/g and 13.7Wh/kg, which are in the forefront. The lotus leaf [38] biomass material has been previously studied, but the capacitance density per unit volume of lotus leaf

is not as large as that of lotus stem, and the lotus leaf has a smaller energy density. *Nelumbo nucifera* is a perennial aquatic herbaceous flower native to China. Its entire body is treasured. The lotus seeds and lotus roots have edible value, and the lotus stems, lotus leaves and other parts have medicinal value. Thus, it is planted in large areas in China and has a long history of cultivation. As an excellent biomass activated carbon material, the lotus stem can reduce cost.

The specific surface area of the lotus stem activated carbon material was 2389.7 m<sup>2</sup>/g, which is higher than normal biomass activated carbon [17-25], the pore size was 2.0848 nm. The mesoporous pore size is in the range of  $2.0 \sim 3.3$  nm, it indicates that a large number of mesoporous, mesoporous and macroporous exist in the activated carbon. It is generally believed that mesopores and macropores provide ion transport channels and diffusion channels in electrolyte, and micropores provide space for ion storage energy. Therefore, the prepared activated carbon should have high specific capacitance and rate capacity when used as electrode material in supercapacitors.

In summary, the supercapacitor made with the lotus stem as the activated carbon source has the largest specific energy and the largest specific capacitance. The lotus stem is the most suitable part among the above five parts as the carbon source of activated carbon electrode materials.

# 3.2 SEM Test

# 3.2.1 SEM test of AC<sub>ls</sub>



Figure 3. SEM diagram of the AC<sub>ls</sub>

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Figure 3 shows the electron microscope pictures of  $AC_{ls}$  with magnifications of 5000, 2000, 500 and 100, from which we can clearly observe the structure and morphology of  $AC_{ls}$ . Macroscopically, the loading bar has a rich pore structure; after crushing, carbonization and activation, the micropore structure remains very developed. Therefore,  $AC_{ls}$  has many pore channels of different sizes, which leads to its large specific surface area and strong adsorption performance. It can be loaded with metal compounds such as manganese dioxide.

#### 3.2.2 SEM test of MnO<sub>2</sub>/AC<sub>ls</sub>



Figure 4. SEM diagram of MnO<sub>2</sub>/ AC<sub>1s</sub>

Figure 4 shows the electron microscope pictures of  $MnO_2/AC_{ls}$  composites with magnifications of 5000, 2000, 500 and 100. In Figure 4a, we clearly observe that manganese dioxide powder is successfully attached to the surface and pore channels of the rod-loaded activated carbon. In Figures 4b and 4c, the cross-section of the rod-loaded activated carbon is a bundle tube structure, and its surface is uneven, which increases the effective contact area. In Figure 4 d, the particle size of the material is small and relatively uniform. According to the above analysis, the AC prepared from the  $MnO_2/AC_{ls}$  has good adsorption, and the composite material will make an excellent electrode.

#### 3.3 GCD Test



3.3.1 GCD test of ACls

Figure 5. GCD diagram when the current density is (a) 0.1 A/g, (b) 0.3 A/g, and (c) 0.5 A/g

Figures 5a, b and c show the constant-current charge discharge diagrams of the supercapacitors with 6 mol/L KOH as the electrolyte at current densities of 0.1 A/g, 0.3 A/g and 0.5 A/g, respectively. After data processing, the specific capacitances in Figures 5a, 5b and 5c are 209.79 F/g, 200.80 F/g, and 186.03 F/g, respectively. The average specific capacitance of the pure carbon supercapacitor under different current densities is approximately 200.62 F/g. In addition, the data and images show that the specific capacitance decreases with the increase in current density, and the discharge voltage drop increases with the increase in current density. The reasons for this phenomenon are as follows: with the increase in current density, the charging and discharging time is shortened, and the electrochemical reaction rate lags behind the electron transmission rate. Therefore, the voltage reaches the set value

before the capacity value is reached; subsequently, the charging/discharging is stopped, and the next cycle is started. Then, the specific capacitance decreases, and the voltage drop increases.

## 3.3.2 GCD test of MnO<sub>2</sub>/AC<sub>ls</sub>

Figures 6a, b, c, d, e and f show the constant-current charge/discharge diagrams with 6 mol/L KOH solution as the electrolyte and manganese dioxide contents (mass fractions) of 5%, 10%, 20%, 30%, 40% and 50%, respectively.





Figure 6. GCD diagram of AC<sub>1s</sub> loaded with (a) 5% MnO<sub>2</sub>, (b) 10% MnO<sub>2</sub>, (c) 20% MnO<sub>2</sub>, (d) 30% MnO<sub>2</sub>, (e) 40% MnO<sub>2</sub>, and (f) 50% MnO<sub>2</sub>.

The 30% and 40%  $MnO_2$  samples have similar AC specific capacitances, and the 10%  $MnO_2$  sample has the highest AC specific capacitance (323.66 F/g, which is 46.23% higher than that of EDLC made from lotus stem activated carbon without  $MnO_2$ ), which greatly improves the capacity performance of pure-carbon-based EDLC. However, with the increase in the load ratio, the specific capacitance is not proportional to the load ratio.

The experimental results show that when the load ratio exceeds 40%, the specific capacitance of EDLC obviously and rapidly decreases, and the voltage drop sharply increases. In general, the specific capacitance first increases and subsequently decreases, which is related to the special energy storage principle of the pseudo capacitor: when the pseudo capacitor is charged and discharged, both a redox reaction and electric double-layer capacitance occur, which can increase the specific capacitance. However, when the load ratio exceeds a certain range, the specific surface area decreases, and the energy storage effect of the electric double-layer capacitor is weakened.

# 3.4 CLT Test

## 3.4.1CLT test of $AC_{ls}$

The specific capacitance retention rate is shown in Figure 7, the number of charge/discharge cycles is set to 1000, the energy efficiency is maintained above 81% at 10 A/g in 6 M KOH, and the specific capacitance retention rate is above 89%. These results indicate that the lotus stem-derived activated carbon has good cycle stability and high reversibility in KOH electrolytes [40]. Compared other similar biomass materials [41, 42], specific capacitances still is well retained. It indicates that

AC<sub>ls</sub> exhibit long-term cycle stability and good electrochemical reproducibility due to the excellent electrical conductivity.



Figure 7. Frequency distribution of the specific capacitance of variable cycles

## 3.4.2 CLT test of MnO<sub>2</sub>/AC<sub>ls</sub>

The assembled button battery was charged and discharged 1000 times with a high current on the tester (BTS4000).



Figure 8. Cycle life diagram of  $AC_{ls}$  with different contents of  $MnO_2$ 

Figure 8 shows that the 0%  $MnO_2$  samples have the best stability, and the specific capacitance retention rate reaches 95.7% after 1000 cycles at 10 A /g in 6 M KOH. The stabilities of the 5% and 10%  $MnO_2$  samples are also relatively good. After 1000 charge discharge cycles, the specific capacitance retention rate still exceeds 80%. When the load ratio is 40% or exceeds 50%, the stability of EDLC sharply decreases.

When the load ratio is 50%, the specific capacitance of EDLC is only 64.47%. It is very important to find the best load ratio that can improve the EDLC performance and save resources.

# 3.5 CV Test

In this test, 1 mol/L KOH solution was used as the electrolyte. The scanning rates were 5mV/s, 6 mV/s, 7 mV/s, 8 mV/s, 9 mV/s, and 10 mV/s, and the voltage range was -0.2 to 0.6 V.



Figure 9. Cyclic voltammetry diagrams of the electrode materials with (a)  $AC_{ls}$  as the carbon source, and (b) 10%  $MnO_2/AC_{ls}$  as the carbon source at different scan rates

The ability of the supercapacitor to store charge can be analysed by the cyclic voltammetry generated in a certain scan rate range. The results indicate the CV of the lotus stem electrode has an asymmetrical quasi-rectangular shape due to the charge resistance of the electrode in the potential range of -0.2 V to 0.6 V. Although the CV of the lotus stem electrode is asymmetric, its shape is well retained, which is consistent with the increase in the scan rate, and the electrode retains its considerable capacitance even at a high scan rate of 10 mV/s. This phenomenon is attributed to the ohmic effect resistance of electrolyte movement in porous carbon, where storage charge has been

confirmed as double layer formation mechanism [21, 43]. More importantly, the electrode exhibits typical capacitance characteristics with good reversibility and high stability.

In Figure 9b, the CV has a rectangular shape. With the increase in scanning rate, the shape of the figure is basically maintained. Due to the addition of manganese dioxide, there are two types of capacitance effects, and the symmetry of the measured figure is poor. However, the cyclic voltammetry curve of the 10%  $MnO_2/AC_{ls}$  does not show obvious oxidation and reduction peaks, and thus, the double-layer capacitance occupies the dominant position in the total capacitance value.

# 3.6 EIS Test

Using the 3-electrode system and 1 mol/L KOH aqueous solution as the electrolyte, we measured the impedance spectra and resistance of  $AC_{ls}$  and 10%  $MnO_2/AC_{ls}$ .



Figure 10. Impedance diagram of the electrode material with (a)  $AC_{ls}$  and (b) 10%  $MnO_2/AC_{ls}$ 

From the EIS data, the specific capacitance and impedance of the electrode are determined by the following formulas [36].

$$C = -\frac{1}{2\pi \cdot \mathbf{f} \cdot Z''m}$$
(5)  
$$Z = Z' + \mathbf{j} \cdot Z''$$
(6)

where m is the weight of the electrode; f is the frequency; Z' is the real impedance; and Z'' is the imaginary impedance.

The semicircle in the curve in Figure 11a is due to the ion migration resistance in the electrode micropores or the increase in capacitance, which may occur in the micropores. The intersection of the extension line of the arc portion and the horizontal axis is taken as the resistance of the electrode material. A smaller resistance corresponds to a better electrical performance of the electrode material, Also the vertical lines close to 90° at low frequencies indicate the pure capacitance and rapid transfer ability of electrolyte ions in the electrode material [44, 45]. The impedance of the electrode material in Figure 10a is 2.37 ohms, which is very small compared to the amount of electrode material in the 3-electrode system, which fully illustrates that the lotus stem is an ideal carbon source for the activated carbon electrode material. With manganese oxide, the impedance of the electrode material in Figure 11b is 2.21 ohms, the mass transfer and diffusion resistance of the material are small, and the conductivity is good. Therefore, the material presented herein is an ideal electrode material for EDLCs.

#### 4. CONCLUSIONS

By comparing the constant current charge/discharge test results of the supercapacitors made with the lotus stem, lotus leaf, lotus seedpod, lotus seed, and lotus root as the carbon sources, we found through calculations that the supercapacitors made with lotus stem had the smallest resistance and largest capacitance. The results of the cyclic voltammetry test, impedance test and cycle life test show that the supercapacitors with the lotus stem as the carbon source have excellent energy density, stability and other electrochemical performances. The BET and SEM results show that the activated carbon made with the lotus stem source has suitable physical properties such as surface junctions for an electrode material. In summary, among the five parts of *Nelumbo nucifera* (the lotus stem, lotus leaf, lotus seed and lotus root), the lotus stem is the most suitable part as a carbon source for the electrode material of supercapacitors.

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## References

- 1. J. Wang, P. Nie, B. Ding, X. Hao, S. Dong, H. Dou and X. Zhang, *J. Mater. Chem. A.*, 5(2017)2411. DOI: 10.1039/C6TA08742F.
- 2. Y.G. Wang, Y.F. Song and Y.Y. Xia, Chem. Soc. Rev., 45(2016)5925. DOI:10.1039/c5cs00580a
- 3. A. Muzaffara, M.B. Ahameda, K. Deshmukha and J. Thirumalai, *Renewable Sustainable Energy Rev.*, 101(2019) 123. DOI:10.1016/j.rser.2018.10.026
- 4. S. Herou1, P. Schlee1, A. B. Jorge and M. Titirici, *Curr. Opin. Green Sustainable Chem.*, 9(2018)18.DOI:10.1016/j.cogsc.2017.10.005
- 5. T. Selvaraja, V. Perumala, S. F. Khorc, L.S. Anthonya, S.B. Gopinathe, and N. M. Mohamed, *Mater. Res. Bull.*, 126 (2020) 110839. DOI:10.1016/j.materresbull.2020.110839
- 6. G.P. Wang, L. Zhang and J.J. Zhang, Chem. Soc. Rev., 41(2012)797. DOI: 10.1039/c1cs15060j

- 7. M. Sevilla and R. Mokaya, *Energy Environ. Sci.*, 7(2014)1250. DOI: 10.1039/c3ee43525c
- Y.L. Shao, M. El-Kady, L.J. Wang, Q.H. Zhang, M. Mousaviae and R. B. Kaner, *Chem. Soc. Rev.*, 44(2015) 3639. DOI: 10.1039/c4cs00316k
- 9. L. Wen, F. Li and H.M. Cheng, Adv. Mater., 28(2016)4306. DOI: 10.1002/adma.201504225
- 10. L.I. Liu, Z.Q. Niu and J. Chen, Chem. Soc. Rev., 45(2016)4340. DOI: 10.1039/c6cs00041j
- 11. A. Borenstein, O.I. Hanna, R. Attias, S. Luski, T. Brousse and D. Aurbach, J. Mater. Chem. A., 5(2017)12653. DOI: 10.1039/c7ta00863e
- 12. J. Yang, P. Xiong, C. Zheng, H.Qiuab and M.D. Wei, J. Mater. Chem. A., 2(2014)39. DOI: 10.1039/c4ta04140b
- 13. L, R. Jothiramalingam, H. A. Al-Lohedan, M. A. Munusamy, E. E and P. Merlin, *New J. Chem.*, 41(2017)3939, DOI:10.1039/C6NJ03867K
- R. Farma, M. Deraman, A. Awitdrus, I.A. Talib, E. Taer, N.H. Basria, J.G. Manjunatha, M.M.Ishak, B.N.M. Dollah and S.A. Hashmi, *Bioresour. Technol.*, 132(2013)254. DOI:10.1016/j.biortech.2013.01.044
- 15. T. E. Rufford, D. Hulicova, Z. Zhu and G. Q. Lu, *Electrochem. Commun.*, 10 (2008) 1594. DOI:10.1016/j.elecom.2008.08.022
- J.J.Liu, Y.F. Deng, X.H. Li and L.F.Wang, ACS Sustainable Chem. Eng., 4(2016)177. DOI:10.1021/acssuschemeng.5b00926
- Y.J. Li, G. Wang, T. Wei, Z.F. Fan and P. Yan, *Nano Energy.*, 19(2016): 165. DOI:10.1016/j.nanoen.2015.10.038
- 18. Y.Q. Zhao, M. Lu, P.Y. Tao, Y.J. Zhang, X.T. Gong, Z. Yang, G.Q. Zhang and H.L. Li, *J. Power Sources.*,307 (2016) 391. DOI:10.1016/j.jpowsour.2016.01.020
- 19. J.Z. Chen, K.L. Fang, Q.Y. Chen, J.L. Xu and C.P. Wong, *Nano Energy.*, 53(2018)337. DOI:10.1016/j.nanoen.2018.08.056
- 20. Q.H. Liang, L. Ye, Z.H. Huang, Q. Xu, Y. Bai, F.Y. Kang and Q.H. Yang, *Nanoscale.*, 6(2014)13831. DOI: 10.1039/c4nr04541f
- C. Peng, X. Yan, R. Wang, J.W. Lang, Y.J. Ou and Q.J. Xue, *Electrochim. Acta.*, 87 (2013) 401, DOI:10.1016/j.electacta.2012.09.082
- 22. G. Xu, J. Han, B.Ding, P. Nie, J. Pan, H. Dou, H. Li and X. Zhang, *Green Chem.*, 17(2015)1668.DOI: 10.1039/C4GC02185A.
- 23. A. M. Abioye and F.Nasir Ani, *Renewable Sustainable Energy Rev.*, 52 (2015) 1282. DOI:10.1016/j.rser.2015.07.129
- 24. C.P. Ruan, K.L. Aia and L.H.Lu, Adv., 4(2014)30887. DOI: 10.1039/c4ra04470c
- 25. J.T. Zhang , L.Y. Gong , K.Sun ,J.C. Jiang and X.G.Zhang, *J Solid State Electrochem.*, 16(2012) 2179. DOI :10.1007/s10008-012-1639-1
- 26. Y.Zhang, S.S. Liu, X.Y. Zheng, X. Wang, Y. Xu, H.Q.Tang, F.Y. Kang, Qu.H. Yang and J.Y. Luo, *Adv. Funct. Mater.*, 27(2017)1604687.DOI: 10.1002/adfm.201604687
- X.J. He, Pi.H. Ling, J.S. Qiu, M. Yu, X.Y. Zhang, C. Yu and M.D. Zheng, *J.Power Sources*, 240 (2013) 109. DOI:10.1016/j.jpowsour.2013.03.174
- 28. J. Wang, J.Q. Li, Q.J. Cui, G.X. Xu and L. Hu, 2018 2nd International Conference On Green Energy And Application, 978-1-5386-5236-7/18/\$31.00 ©2018 IEEE.
- 29. L. Hu, Y.T. Pan, S. Liu, etc, Int. J. Electrochem. Sci., 2020:8241-8252.
- 30. M.Inagaki, H. Konnoa and O. Tanaike, *J. Power Sources.*, 195 (2010) 7880. DOI:10.1016/j.jpowsour.2010.06.036
- 31. J.T. Zhang and X. S. Zhao, Chem. Sus. Chem., 5(2012)818. DOI: 10.1002/cssc.201100571
- 32. A. Allagui, T. J. Freeborn, Ahmed S. Elwakil and Brent J. Maundy, *Sci. Rep.*, 6(2016)38568. DOI:10.1038/srep38568
- C.C. Gong, X.Z. Wang, D.H. Ma, H.F. Chen, S.S. Zhang and Z.X. Liao , *Electrochemi. Acta.*, 220 (2016) 331. 30. DOI:10.1016/ j.electacta.2016.10.120
- 34. C. Bora, S. Dolui, J. Phys. Chem. C,118(2014)29688. DOI:10.1021/jp511095s

- K. F. Yu, H. Zhu, H. Qi and C. Liang, *Diamond Relat. Mater.*, 88 (2018) 18–22. DOI:10.1016/ j.diamond.2018.06.018
- 36. A. Belloa, N. Manyala, F. Barzegara, A. A. Khaleedab, D. Y. Momodua and J. K., *RSC. Adv.*, 6(2016) 1800-1809. DOI: 10.1039/C5RA21708C
- 37. L. Jiang, J. W. Yan , L. X. Hao , R. Xue, G.Q. Sun and B. L. Yi, *Carbon.*, 56 (2013) 146 154, DOI:10.1016/j.carbon.2012.12.085
- P. Cheng, S.Y. Gao, P.Y. Zang, X.F. Yang, Y.L. Bai, H.Xu, Z.H. Liu and Z.b.Lei, *Carbon.*, 93( (2015)315. DOI: 10.1016/j.carbon.2015.05.056
- 39. D. R. Patil, B. Koteswararao, K.Begari, A. Yogi, M. Moussa and Deepak P. Dubal, ACS Appl. Energy Mater., 2(2019) 2972.
- 40. J. Li, G. Zan and Q. Wu, RSC Adv., 6(2016)57464. DOI: 10.1039/C6RA08428A.
- 41. G. Zhang, Y. Chen, Y. Chen, H. Guo, *Mater. Res. Bull.*, 102(2018)391-398. DOI:10.1016/j.materresbull.2018.03.006
- 42. H. Chen, D. Liu, Z. Shen, B. Bao, S. Zhao and L.Wu, *Electrochimi. Acta.*, 180 (2015) 241–251 DOI: 10.1016/j.electacta.2015.08.133
- 43. L.H. Wang, M. Toyoda, M. Inagaki, *New Carbon Mater.*, 23 (2008) 111. DOI: 10.1016/S1872-5805(08)60015-3
- 44. M. Ghaemi, F. Ataherian, A. Zolfaghari, S.M. Jafari, *Electrochimi. Acta.*, 53 (2008)4607. DOI:10.1016/j.electacta.2007.12.040
- 45. V.S. Jamadade, V.J. Fulari, C.D. Lokhande, *J. Alloys Compd.*, 509 (2011) 6257. DOI:10.1016/j.jallcom.2011.03.035

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