Preparation and environmental impact assessment of NI-N modified sludge activated carbon electrode materials

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Abstract. With the activated sludge in urban sewage treatment as the precursor material, the carbon material is prepared by high temperature carbonization, and then the nickel-nitrogen modified activated carbon material is prepared by potassium hydroxide activation, urea and nickel doping. Electrochemical experiments with activated carbon materials show that it has a good specific capacitance of 345.9 F/g at 1 A/g current density. After 2 000 cycles at a current density of 10 A/g, it still maintains 84.53% capacitance, shows found that the main source of influence is the power consumption in the process of sludge carbonization.

Keywords: Activated Sludge; Electrode Materials; Full Life Cycle Assessment

1. Introduction

As a by-product of domestic sewage treatment, activated sludge will cause harm to the environment. The main means of sludge treatment are landfill and incineration, but it can not completely avoid the secondary pollution caused by it. Sludge contains a large number of carbon elements, preparing activated carbon electrode material through simple carbonization method, and solves the environmental pollution problem brought by sludge, but also provides a new solution for the resource treatment of sludge [1]. Zhang et al[2]prepared activated carbon electrode materials by sludge carbonization activation showed a specific capacitance of 287F/g. However, in recent studies, large logarithmic scholars have focused on the electrochemical properties of activated carbon electrode materials, and ignore the impact on the environment in the preparation process. Full life Cycle Assessment was used to evaluate the environmental impact of the preparation process and make suggestions for the preparation process [3].

2. Materials and Methods

2.1 Preparation of activated carbon

The dried sludge was carbonized in a tubular furnace, the carbonization temperature was 60℃ and the carbonization time was 2 h. The prepared carbon material, urea and KOH according to the mass ratio of 1:1:2, fully mixed and then placed in a tubular furnace for activation, activation temperature of 800℃, activation time of 2 h, get the nitrogen modified activated carbon material, named N-AC.

Take 0.1 g of N-AC material into 500 mg/L of Ni (NO_3) 2 solution, adsorb for 4h, transfer to 200℃ hydrothermal reaction for 12 h, filter and dry to get nickel and nitrogen modified Mars material, named Ni-N-AC-HR. At the same time, the material only through nickel ion adsorption doping as a control, named Ni-N-AC

2.2 Electrochemical testing

Electrochemical testing usually uses Cyclic Voltammetry (CV), Constant Current Charging and Discharge (GCD), and Electrochemical Impedance Spectroscopy (EIS) to react with their electrochemical properties. In this experiment, 6 M KOH solution is used as electrolyte, the working electrode uses activated carbon material prepared by nickel foam load, platinum sheet electrode for the electrode, and Hg/HgO electrode for the reference electrode.

2.3 Full-life-cycle impact assessment

Table 1. material and energy consumption during the preparation of electrode materials

process	matter and energy	measure	unit
Sludge pretreatment	electric energy	15.947	kJ
Carbon	electric energy	13.249	kJ
	N2	3.937	L
	HF	0.382	kg
	HCl	0.56	kg
	C2H6O	0.885	kg
	H2O	0.71	kg
Activation	electric energy	18.227	kJ
	N2	2.165	L
	KOH	1.193	kg
	urea	0.597	kg
	H2O	2.01	kg
Electrode material preparation	PDFE	0.0626	kg
	Carbon Black	0.0626	kg
	DMF	2. 005	kg

In this study, the life cycle of the sludge-based electrode material production process was evaluated using GaBi software.

System boundary considers the four main stages of preparation of activated carbon electrode material, respectively, sludge pretreatment stage, sludge carbonization stage, activation stage and electrode material preparation stage, functional unit is defined as 1 kg sludge powder material, input data (chemicals, energy and water) from the actual experimental data and Gabi database, as shown in Table 1.

3. results and discussion

3.1 Electrochemical testing

The electrochemical properties of N-AC, Ni-N-AC, and Ni-N-AC-HR were tested using cyclic voltammetry, constant current charge and discharge, and electrochemical impedance in the three-electrode system, and the experimental results are shown in Fig 1.

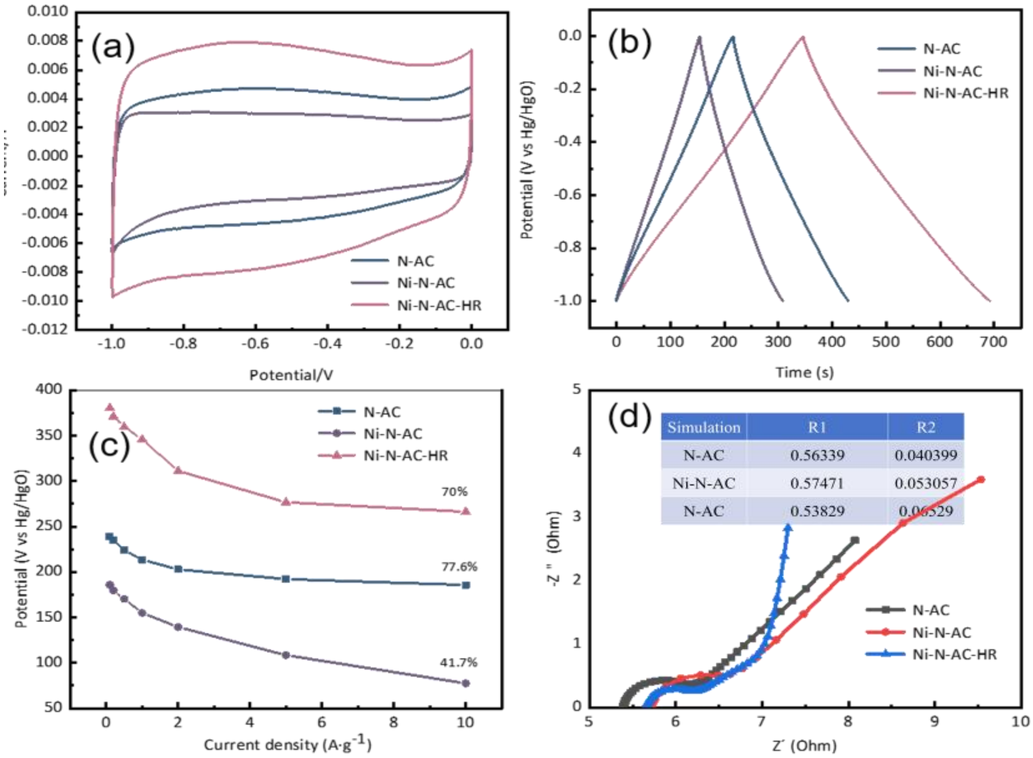

Fig.1 (a) Cyclic voltammetry curve; (b) constant current charge-discharge curve; (c) capacitance decay at different current densities; (d) electrochemical impedance

According to the CV and GCD curves of the electrode materials in Fig 1 (a) and Fig 1 (b), Ni-N-AC-HR has the highest specific capacitance of 345 F/g [4]. The N-AC specific capacitance without Ni ion modification is only 153.4 F/g. Fig 1 (c) shows the calculation results of the specific capacitance of the three materials at different current densities. It can be seen that the specific capacitance of the three materials decreases with the increase of the current. Ni-N-AC-HR reduces the specific capacitance to 70% at 10 A/g, while Ni-N-AC decreases to 40%, which indicates that the more stable load of Ni ion can be tested to be placed on the activated carbon material after hydrothermal reaction. Fig 1 (d) is the EIS diagram of the electrode material. In the equivalent circuit diagram, it can be clearly seen that Ni-N-AC-HR has the minimum electrochemical impedance [5].

Fig 2 shows the CV plots of Ni-N-AC-HR at different sweep velocities, the GCD map of current density and the cycle use experiments.

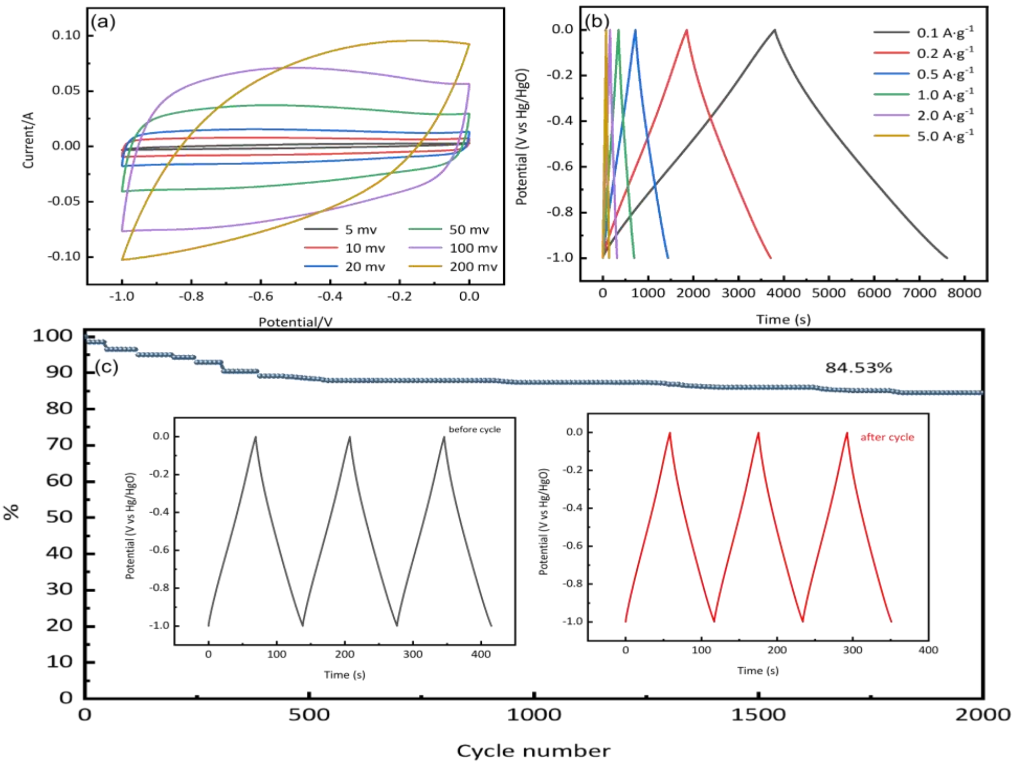

Fig 2 (a)5-200 mv/s scan speed constant current charge-discharge test;(b)0.1-5 A/g current density constant current charge-discharge test;(c) cycle life test

From Fig 2 (a), we can see that all the Ni-N-AC-HR5-100 mvs-1 have a quasi-rectangular shape, which indicates that the material has good multiplier performance [6]. Fig 2 (b) At the current density of 0.1-5 A/g, the constant current charge-discharge curve of Ni-N-AC-HR shows a relatively symmetrical isosceles triangle, while the slight deviation of the charge-discharge curve may be due to the redox reaction of oxygen, nitrogen functional groups and Ni2 + with the electrolyte ions in the material [7]. Fig 2 (c) shows the cycle performance diagram of Ni-N-AC-HR at a current density of 10 A/g. In the 2000 circle of the stability test, the capacitor value of the electrode gradually decays and finally stabilizes at 84.53% and no obvious deformation, indicating that the material has excellent electrochemical stability and highly reversibility.

3.2 Full life cycle environmental impacts

Gabi software calculated the environmental impact of the preparation of activated carbon electrode materials. According to the quantitative base value of CML 2001 evaluation method, the fossil consumption potential (ADP), Marine ecological toxicity potential (MAETP), acidification potential (AP), human carcinogenic toxicity (HTP), water eutrophication potential (EP) and global warming (GWP) were compared. The results are shown in Fig 3.

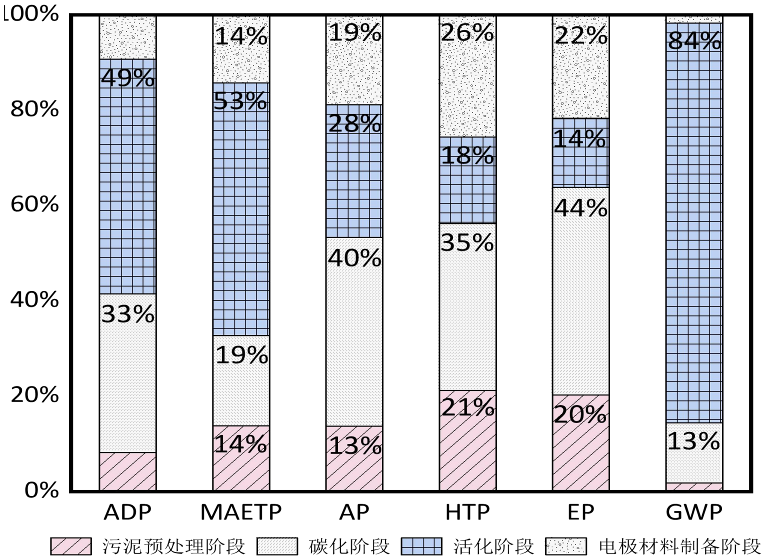

Fig 3 environmental impact contribution rate of each stage

As can be seen from Fig 3, in the whole life cycle, the activation stage has the largest proportion, contributing 84% to GWP, and ADP and MAETP contributing about 50%.GWP and ADP are mainly caused by the use of electric energy, while MARTP is caused by the use of activator and urea. The carbonization stage has the highest proportion, among the three indexes of AP, HTP and EP, 40%, 35% and 44% respectively. This is due to the use of a large number of chemical reagents HCl, HF, absolute ethanol to clean the metal impurities, SiO2, biological oil and so on in the activated carbon. The other two stages have a very small proportion, contributing less than 30% in MARTP, AP, HTP and EP, and no more than 10% in ADP and GWP

After comparison of each process know, can consider from carbonization, activation of these two stages to optimize the production process, in these two stages consumed a lot of electricity, can consider using clean renewable energy to replace carbonization, activation phase of electricity consumption, at the same time there are a lot of heat waste, in mass production, using steam turbine to recover lose heat can offset part of the electricity consumption. In addition, the carbonization process of sludge will produce a large number of bio-oil, which can be recycled.

4. Conclusion

The specific capacitance of Ni-N-AC-HR activated carbon material is greatly increased by the redox reaction of nickel ions. At the current density, the specific capacitance of 1 A/g is 345 F/g, which is about 2.25 times that of N-AC. At the same time, the stability of the material can be effectively improved after the hydrothermal reaction. 70% of the specific capacitance is still maintained at the current density of 10 A/g. After 2 000 cycles, the specific capacitance is 84.53% of the first cycle.

The environmental impact of activated carbon electrode materials by sludge activation was evaluated using LCA. The results show that GWP, ADP, MAETP, AP, HTP and EP are significant. Among them, the environmental impact of the carbonization and activation stage is the greatest.

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