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Reutilization of the expired tetracycline for lithium ion battery anode

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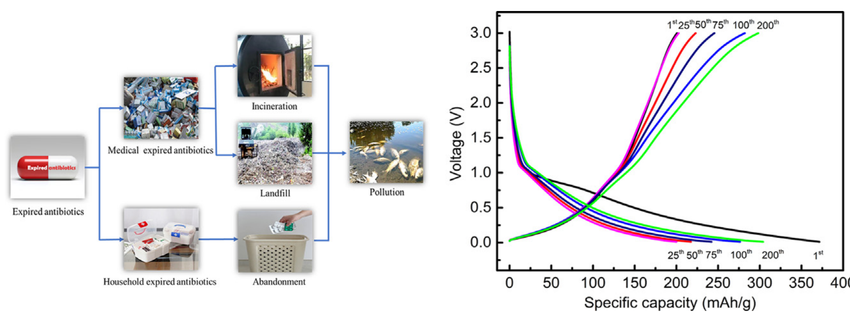
HIGHLIGHTS

- The expired tetracycline was reutilized as LIB anode for the first time.
- The expired tetracycline anode delivered the satisfactory performances.
- The potential Li-storage mechanism was also explored by CV and FTIR.
- A new application of the expired tetracycline was launched.

GRAPHICAL ABSTRACT



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ABSTRACT

Waste antibiotics into the natural environment are the large challenges to the environmental protection and the human health, and the unreasonable disposal of the expired antibiotics is a major pollution source. Herein, to achieve the innocent treatment and the resource recovery, the expired tetracycline was tried to be reutilized as the electrode active material in lithium ion battery (LIB) for the first time. The micro-structure and element component of the expired tetracycline were characterized by scanning electron microscope (SEM), energy dispersive X-ray spectroscopy (EDX), X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectroscopy (FTIR). Furthermore, the corresponding electrochemical performances were also investigated by galvanostatic charge/discharge and cyclic voltammetry (CV). To be satisfactory, the expired-tetracycline-based electrode delivered the initial specific discharge capacity of 371.6 mAh/g and the reversible specific capacity of 304.1 mAh/g after 200 cycles. The decent results will not only offer an effective strategy to recycle the expired tetracycline, but also shed a new light on the cyclic economy and the sustainable development.

1. Introduction

Since the advent of the penicillin in 1929, various antibiotics have been widely used for the therapy of many infectious diseases because of high efficiency and easy availability. Meanwhile, with the increase of the antibiotics' demands, the continuous release into the environment and the resultant negative impacts to the ecosystem were more and more serious (Luo et al., 2011; Zhang et al., 2015). It was reported that the concentrations of the antibiotics ranged from several ng/L to hundreds of μg/L in the various environmental compartments, such as hospital sewage, municipal sewage, surface waters, groundwater and seawater (Yan et al., 2013). These waste antibiotics into the environment can not only result in the direct toxicity to the animals and the plants, but also induce the generation of the antibiotic-resistant bacteria, which would be a serious threat to the human health (Pan and Chu, 2016; Zhang et al., 2016). Especially, it's hard to degrade the antibiotics once they are released into the natural ecosystem. Even in the waste water treatment plants, these antibiotics can't be completely eliminated (Fenech et al., 2013). Seemingly, how to effectively recycle

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the expired antibiotics and control their emission is very important and necessary.

Besides the metabolic antibiotic residue, another main emission is from the unreasonable disposal of the expired medicines. Once the supply of the antibiotics exceeds the demand, these medicines will easily become overdue and useless, which would pose a potential threat to the natural environment. According to the management status of the expired antibiotics in Fig. 1, these expired antibiotics mainly come from two resources: one is from the pharmacies or the hospitals, and the other is from the family reserve. In the case of the former, the expired medicines can be collected by the special organizations or manufactures (Voudrias et al., 2012). After being gathered, these expired drugs would be subject to the incineration or the landfill, in which the expired antibiotics can be destroyed *via* high temperature incineration or biological degradation. The resultant consequences would waste the useful resources and bring out the second pollution, because some antibiotic residues may persist and leak into the rivers and the groundwater. As for the latter, the expired medicines in the families would be randomly discarded together with other common solid wastes because of the wide dispersity, low social consciousness and high collection cost (Bound and Voulvoulis, 2005). Seemingly, there hasn't yet been an effective friendly strategy to recycle and reutilize the expired medicines for the moment. Therefore, in order to build the circular economy mode of the expired medicines and reduce the environment pollution and emission, it is urgent to actively carry out the investigations on the cyclic utilization of the expired medicines, particularly on the expired antibiotics.

Since being commercialized by Sony corporation in 1991, LIB has rapidly dominated the market of the portable electronic devices. Recently, LIB has been also demonstrated in other areas such as the electric vehicles and the smart grids (Armand and Tarascon, 2008; Dai et al., 2016; Hou et al., 2017; Li et al., 2014; Zhu and Chen, 2015). Besides the common inorganic materials, the organic electrode materials have also become one of the hot subjects in LIB due to their many advantages such as low cost, easily processing, lightness, softness and resource renewability (Liang et al., 2012). Generally, these organic electrode materials can be divided into carbonyl compounds, organosulfur compounds, organic free radical compounds and conducting polymers (Renault et al., 2016). Besides the traditional chemical synthesis, the organic electrode materials can be also extracted from the biomass resources or recovered from the waste resources (Chen et al., 2008; Milczarek and Inganäs, 2012). For example, as a natural antioxidant, ellagic acid can be extracted from the pomegranate husk and act as the electrode active material in LIB with high reversible capacities of 450 mAh/g at 0.1C and 200 mAh/g at 0.4C, (Goriparti et al., 2013). Another natural organic compound, humic acid can be derived from the wood, the soil or the coal, and the humic-acid-based anode in LIB delivered 180 mAh/g after 200 cycles at 40 mA/g (Zhu et al., 2015). Seemingly, to recycle the organic electrode materials from the biomass or the waste resources is becoming a promising strategy for the development of LIB electrode material. In principle, considering that the main effective ingredients of many antibiotics are the organic compounds with O-containing functional groups, the expired antibiotics may be also suitable for the electrode active materials in LIB.

Tetracycline is well known as an inexpensive broad-spectrum antibiotic with the efficient inhibitions to most of aerobic, anaerobic, Gram-negative, and Gram-positive bacteria. Therefore, it is the common clinic drug to cure some infectious diseases of the human beings and the veterinaries. Its chemical structural formula consists of four linearly annelated six-membered rings and several functional groups on the rings such as one methyl group, one tertiary amine group, one amide group, two carbonyl groups and five hydroxyl groups, as shown in Scheme 1. The skeleton and these functional groups are the important prerequisites for its biological activity. More interestingly, hydroxyl and carbonyl groups within the tetracycline molecules may be also active to storage Li^+, while the polycyclic naphthacene structure may facilitate the structural stability. However, to the best of our knowledges, the reports about the tetracycline-based anode for LIB are still scarce, especially for the expired tetracycline. Herein, in order to avoid the secondary pollution and reduce the recovery cost as much as possible during the recycling of the waste materials, the expired tetracycline was tried to be directly used as the anode material for the first time and investigated by methods of SEM, EDX, XPS, FTIR, galvanostatic charge/discharge and CV.

Fig. 1. The management status of the expired antibiotics.