Review

Waste Resin Derived Carbon Materials for Sodium-Ion Batteries

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ABSTRACT: As the environmental issues caused by waste resin become increasingly severe, there is an urgent need to develop ways to handle it in a high-value and harmless manner. Turning waste resin into functional carbon materials is a realizable and promising scheme, which could be a trigger to carry forward emerging sustainable battery technologies and applications. However, there are few review articles about the basics and research progress of the waste resin derived carbon materials for sodium-ion batteries. This review article provides a brief overview mainly about resin recycling and the potential usage of the resultant carbon materials for sodium-ion batteries. Specifically, we show the potential improvements in existing research, focusing on utilization of the waste as well as the significance of new routes for resin recycling. This work offers insights for the design of sustainable carbon materials for battery systems.

Keywords: Waste Resin; Waste Recycling; Carbon Material; Sodium-ion Battery; Negative Electrode Material

1. Introduction

Energy is an essential component of our daily lives and the sustained growth of economies. Over the last few decades, human beings' exploitation and utilization of fossil energy have produced many influences on the earth's atmosphere, hydrosphere and biosphere, on which we humans depend for survival. Scientists have been developing advanced materials for clean energy storage and conversion to address the issue in recent years [1]. This is the field where rechargeable batteries become increasingly important [2,3].

However, cost and availability issues of the battery electrode materials have been plaguing the researchers. The most popular lithium-ion batteries (LIBs) are also facing the challenge of the exhaustion of lithium on the earth [4]. Thus, low-cost sodium-ion batteries (SIBs) have drawn significant attention, which is particularly attractive for their cost advantages since they use widely available and affordable sodium resources instead of traditional LIBs [5]. Specifically, as discussed in our previous work [5], there are some reasons for the fact that SIBs are most likely to replace LIBs. Firstly, SIBs could use safer, more abundant, and environmentally benign active materials than LIBs, e.g., sodium salts are abundant in nature and inexpensive to obtain. Secondly, cheap aluminum current collector can be used as sodium does not readily form alloys with aluminum. Thirdly, SIBs are very similar to LIBs in architecture, working mechanisms, components, and fabrication steps, which makes it easy for battery manufacturers to convert the production line of LIBs to SIBs. Thus, developing a cheap and easily prepared material for high-performance SIBs is crucial.

Furthermore, based on the waste-derived electrode materials, the waste-to-treasure method can not only further reduce the overall cost but also alleviate the effect of contamination without generating new harmful substances. Similar concepts on waste plastics were also recently reported in the Journal [6]. With further developments, such a technology could pave the way towards achieving a "carbon emission peak" and "carbon neutrality" in the long term [7]. In this particular context, this mini-review provides a brief overview of the basics and research progress of the waste resin-derived carbon materials for SIBs, which is potentially beneficial work of both environmental protection and energy-storage material exploration. As humanity faces the significant challenge of the energy crisis, this research could help achieve a more sustainable future.

2. Brief Overview of SIBs

2.1. Working Principle

Although LIBs have dominated the recent rechargeable battery market, the high cost [8] anda low abundance of lithium have cast shadows on their wide use in the future. In the meantime, SIBs are considered alternatives or complements in some specific application scenarios, since Na has a similar atomic structure, comparable redox potential and, more importantly, it represents less cost and higher accessibility. In September 2021, Market.us reported that the global SIB market had reached US$ 542.4 Mn in 2018 and was projected to increase significantly at a CAGR of 23.5% from 2019 to 2028 [9]. Despite some debate on detailed sodium storage mechanisms [10], SIBs share similar energy storage mechanisms with LIBs.

Specifically, both rechargeable batteries consist of two electrodes, a separator, and electrolytes, which form a redox reaction (Figure 1). During charging, the positive electrode side undergoes an oxidation half-reaction, where a positively charged external power source extracts electrons from the positive electrode material and transfers them via an external circuit to the negative electrode side (typically with a layered and porous structure). This leads to an electrical imbalance in the battery system, forcing sodium ions to migrate towards the negative electrode through the internal circuit. The negative electrode promotes a reduction half-reaction that allows the storage of sodium ions within the layers, known as sodiation. This process continues until the negative electrode is entirely sodiated, achieving the maximum electrical potential between the electrodes, which is also referred to as the fully charged state. For discharging, on the other hand, represents an opposite reaction where ions migrating out of the negative electrode [11].

Therefore, during the charge-discharge cycles, due to the continuous movement of sodium ions into and out of the negative electrode, some fundamental properties of the negative electrode materials should be guaranteed to fulfil satisfactory performance, including a large capacity of sodium storage, enhanced structural stability [12], as well as high electrical conductivity [13]. Therefore, carbon-based material is a favorable option. Additionally, phosphorus is also a promising candidate, but its practical application is still challenging because of its poor electronic conductivity and large volume expansion during cycling [14,15].
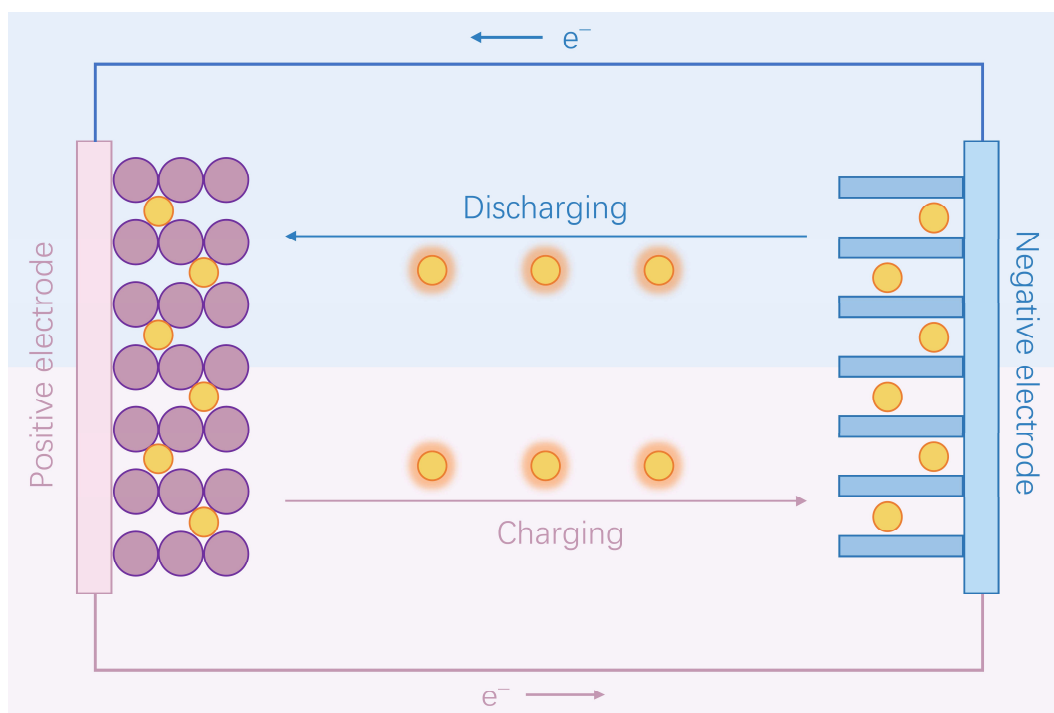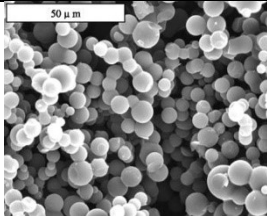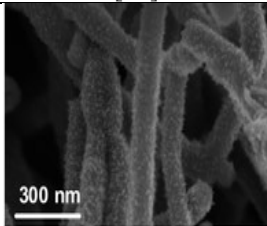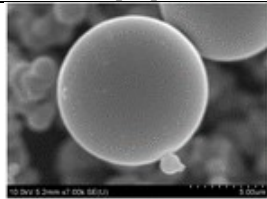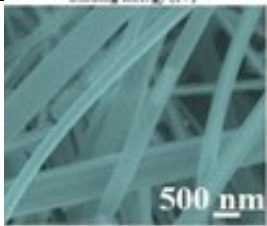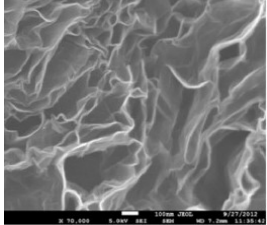

Figure 1. Schematic illustration of the operating mechanism of alkali-ion batteries. Adapted from ref. [5] with permission. Copyright 2023, Elsevier B.V.

2.2. Carbon-Based SIB Negative Electrodes

Over the past few decades, carbon materials for SIBs negative electrodes have been widely investigated [16,17]. Many sorts of them have been put into commercial use today. These materials and their relative properties are listed in Table 1. Graphite comes as the first material to be discussed, but the intercalation process of sodium within graphite is

thermodynamically unfavorable [5,18]. Soft and hard carbon are less graphitized, allowing pores and non-crystalline regions to trigger structural modification in the presence of interlayer sodium. This allows higher sodium storage capacity and structural stability. In fact, with both impressive capacity and initial coulombic efficiency (ICE), hard carbon has been considered the most advanced carbon material for years [19]. It has been one of the most popular commercial materials for scientific and industrial use [20,21]. Despite their excellent performance, these materials are difficult to obtain at a low pyrolysis temperature, which might bring about huge energy consumption in their production. High-cost graphene is sometimes even more alluring, some of which even approach the performance of commercial LIBs (>700 mAh/g) [22]. For instance, early in 2016, Zhang et al. reported a reversible high-rate performance of 809 mAh/g at a current density of 1500 mA/g and capacity retention of 85% over 350 cycles based on a nitrogen-doped graphene network [23]. Yet, there is still a long way for graphene to go because of the generally low ICE (<70%) and potentially low energy efficiency associated with voltage hysteresis.

Table 1. Summary of various types of carbon-based materials and their characteristics. Reproduced from ref. [24] with permission. Copyright 2016, Elsevier B.V.

Category	General Characteristics	Typical Representative	Microscopic Morphology
Hard carbon	Non-graphitic but contains a graphene structure consisting of numerous nanocrystal lines	Resin pyrysis-derived hard carbon	[25]
Nanostructured carbon or carbon composites	Having nanosized structures and a high surface area	Hollow carbon nanowires	[26]
Carbon-based composites	Various carbon materials coupled with alloy materials	Mesoporus carbon-based Se microcomposite	[27]
Carbon nanotubes	Hollow structure with walls formed by atomic layer of graphene	N-doped carbon nanotubes based material	[28]
Graphene	Having sp2-bonded atomic carbon monolayer	Hummers' method-derived rGO	[29]

Besides these above-mentioned carbon materials, other alternatives have also been drawing attention. Carbon nanotubes, or CNTs, are ideal candidates, especially the ones derived from waste. They are allotropes of carbon with nanostructures with a length-to-diameter ratio greater than one million. Thanks to their high specific surface area, good

chemical stability, high electronic conductivity, and non-toxicity, CNTs have received considerable concentration among energy storage applications [30].

CNTs alone can serve as excellent negative electrode materials with specific optimization techniques. Typically, by carefully increasing the interlayer distance of a few outer layers in multiwalled CNTs (MWCNTs), Soroja et al. successfully increased the density of active sites in it and reached 510 mAh/g of specific capacity under a current density of 20 mA/g [31].

More importantly, CNTs can serve as the matrix in negative electrode composite materials. Tao et al. summarized that, when combined with carbon nanotubes (CNTs), carbon-based materials, metal oxides, metal sulfides, and polyanionic compounds typically demonstrate enhanced electrochemical performance compared to their pristine forms [32]. Owing to the endowed high electric conductivity, physiochemical stability, and surface-to-mass ratio, CNTs can increase the ionic conductivity, inhibiting drastic volume expansion and aggregation of active materials, and promoting a stable interface between electrolyte and active materials [32]. For instance, Feng et al. reported a reduced graphene oxide/carbon nanotube (rGO/CNTs) hybrid material, which led to the increased structural stability of rGO and achieved 295 mAh/g of reversible capacity under 50 mA/g after 200 cycles [33].

In consideration of both cost, efficiency and performance, it is believed that carbon materials derived from waste resin through chemical processes could be intriguing for both separate and combined uses in SIBs.

2.3. Current Status of SIB Research

Despite the selection of the right electrodes, the final performance is highly dependent on the optimization of carbon materials, electrolytes and the battery assembly technique. Microstructure regulation can change the porosity, defect concentration, interlayer distance, and overall structure [34]. Thus, boosting the performance of nanocarbon negative electrodes is also important. Nearly all successful carbon-negative electrode materials refer to microstructure regulation, including doping, functionalizing, pore distribution regulation and so on. One primary task in the research community is to conduct systematic characterization and analysis of the composition and microstructure of the carbon materials in orderto project light on potential methods to improve their performance in the near future. Also, revealing the general principles of the optimization or modification methods will be highly important.

Meanwhile, electrolyte modulation also influences the performance of carbon-negative electrode materials. Similar to the LIBs, small molecule esters are the mainstream electrolyte in the SIBs [35]. Besides, the interaction between electrolytes and electrodes, for example, the formation of a solid electrolyte interface (SEI), also significantly contributes to the performance of the whole battery [36]. Early in 2012, Ponrouch et al. [37] studied various electrolytes on a hard carbon negative electrode and verified that high efficiency and capacity could be achieved with dual-solvent electrolyte systems like $NaPF_6$ in Ethylene Carbonate-Diethyl Ethylene Carbonate (EC:DEC) and $NaPF_6$ in Ethylene Carbonate- Propylene Carbonate (EC:PC). Nevertheless, it is also promising for future research to focus on electrolytes and make improvements.

The last key factor lies in the battery assembly. Experience from LIBs has shown the significant influence of mixing, coating, and rolling on their performance [38]; however, related details are always confidential in business. It is believed that SIBs encounter similar problems, and a robust and standardized preparation procedure is crucial for the fair assessment of novel electrode materials. Briefly speaking, the commercialization of SIBs is still underway, and the big challenges are mainly related to small markets, relatively low energy density and immature production lines.

In addition to these influencing factors, sustainable carbon materials from the recycling of organic wastes can definitely play a significant part in the unfolding energy revolution. Small molecules like hydrocarbons and hydrogen could also be obtained during the recycling processes, which leads to the enrichment of the feedstock pool for the synthesis of electrolytes.

3. Resin Derived Carbon Materials for SIBs

3.1. Direct Routes

Some interesting expeditions have been made regarding using resins as the starting materials to yield carbon materials for SIBs, but few of them share the same technical route. Some early works reported some low-capacity but stable carbon materials derived from synthesized epoxy resins [39] or similar-structured gel [40]. In recent years, Kamiyama et al. [41], Zhang et al. [42] and Li et al. [43] developed high-capacity carbon-negative electrode materials by direct pyrolysis of a mixture of phenolic resin (one kind of resin) and other polymers in an inert atmosphere. They investigated the effect of temperature and microstructures. It is worth noting that phenolic resins have also been

considered as significant precursor materials of hard carbon for SIBs owing to their high carbon yield, low cost, and potentially high-quality carbon production [44]. In some other related fields, some scholars used epoxy-matrix polymer composites [45] or some other bio-polymers with similar crosslinking structures (like polysaccharides [46] or sugarcane bagasse [47]) as the precursor and also employed the pyrolysis method to yield carbons. All these researches exhibit an intriguing reversible capacity of around 300 mAh/g and commendable cycling performance (Table 2). Since the crossinglinking structure in raw materials could function as native template during pyrolysis, direct routes facilitate transformation toward interconnected three-dimensional carbon networks, which turn out to be desired properties of SIB negative electrodes.

Table 2. Some existing researches of retrieving carbon electrode materials based on the pyrolysis of resins. Notes: [a] if there are multiple groups of experiments, data is extracted from the one with best results. [b] Usually the pyrolysis contains two parts: pre-heating (<500 °C) and carbonization (>1000 °C). This column only shows the carbonization temperature. [c] All capacities are obtained with half-cell devices. [d] n./a. in the table represents "not applicable".

| Precursor | Pyrolysis Conditions | | | Electrochemical Properties [a] | | | | | Ref. |
	Additives or Catalysts	Temp. [b] (°C)	Time (h)	Capacity [c] (mAh/g)	Current Density (mA/g)	ICE	Capacity Retention	Cycles	
Phenol-formaldehyde resin	n./a. [d]	1100	3	134	50	n./a.	95.5%	100	[39]
Resorcinol-formaldehyde (RF) gels	n./a.	>1200	>2	63	20	>90%	"stable", but no specific info.	Not given	[40]
Microporous phenolic resin	Formalin and maleic acid	1500	1	386	10	85%	96.4%	60	[41]
Mixture of sucrose and phenolic resin	n./a.	1400	1	319	30	87%	90%	150	[42]
Pitch and phenolic resin	aqueous sodium alginate	1400	2	284	30	88%	~94%	100	[43]
Polysaccharides (cellulose)	n./a.	1500	1	353	25	~87%	negligible decay	50	[45]
Biomass-derivative lignin and epoxy resin	n./a.	1400	1	316	30	82%	90%	150	[46]
Sugarcane bagasse (cellulose + lignin)	n./a.	950	6	290	30	70%	90%	300	[47]

However, despite these inspiring results, the consideration of energy consumption would perhaps prove futile for massive industrial use. Owing to the stable chemical bonds in the resin, pyrolysis does not happen until the temperature is raised to about 1000 °C, which is hardly acceptable for energy consumption. Moreover, these researches have neglected the diversity of real resins (including but limited to composition, crosslinking degree and additive) since the above-mentioned epoxy resins or their analogues are mostly synthesized in the lab with explicit structural requirements for pyrolysis, instead of from the waste. Therefore, There is still much to be done in retrieving carbon-negative electrodes from the pyrolysis of resins and further optimizing the resultant carbon materials (e.g., via precursor selection [48], preparation method [42] and surface engineering [49]).

In fact, based on the type of resins, it is more systematic and logical for some waste to undergo recycling [50] and another portion to be subjected to pyrolysis [51]. In doing so, we can acquire more valuable products, reduce energy consumption, restrict carbon emissions and increase the decomposition reaction rate. To be more specific, some resins can be recycled instead of undergoing energy-intensive pyrolysis.

3.2. Indirect Routes

Although researchers have discovered the potential of using resins to prepare SIB negative electrode materials, there is still significant room for improvement in terms of recycling in steps and energy consumption costs. Additionally, the methane-containing waste pyrolysis gas can be further utilized. In the new era of "carbon neutrality" and "emission peak", the reduction of carbon emission in the catalytic decomposition of methane (CDM) will also contribute to serving for the country's environmental protection. Under this condition, the products are typically carbon nanotubes or nanofibers [25]. Although raw CNTs usually exhibit lower capacity (around 300 mAh/g at 0.1C) than amorphous carbon, but their electrochemical performance can be further improved [32]. And to modify the nanocarbon's structure, work can be done separately during and after pyrolysis. Also, the cost of producing carbon from the waste will be significantly

reduced. Considering the energy saved during this process, the performance is still promising, let alone the potential electrode modification techniques (such as forming composites with metal oxides [52] or expanding the interlayer distance [31]).

The detailed mechanism of CDM and the influence of reaction conditions on the performance of nanocarbon are presented below. Despite subtle differences in specific catalysts, the mechanism of CDM can generally be divided into four parts: (1) Methane adsorption on the catalyst surface, (2) gradual breaking of the four C-H bonds in methane, forming hydrogen atoms and carbon-hydrogen radicals, (3) combination of hydrogen atoms and generation of hydrogen gas, (4) diffusion of radicals and carbon atoms inside the catalyst, accumulation and agglomeration of carbon atoms, and growth of carbon materials [53]. The carbon material encapsulation of the catalyst gradually reduces its efficiency, leading to complete deactivation of the catalyst [54]. Figure 2 below takes a common Fe-based catalyst [55] as an example and illustrates the process of CDM, from which the nanocarbon materials are obtained and the reaction temperature is lower at 500–800 °C.
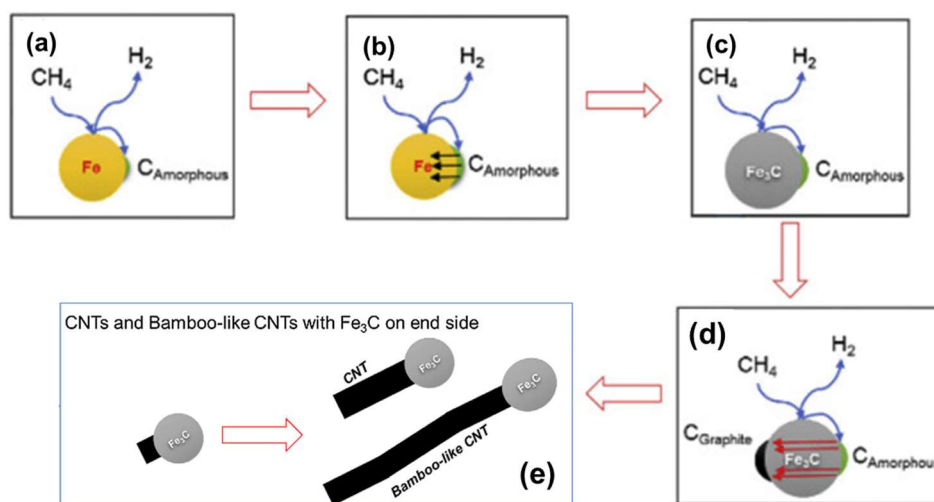

Figure 2. Illustration of bamboo-shaped CNTs growing on the surface of Fe catalyst. (**a**) Dehydrogenation of methane absorbed on the surface of Fe and the formation of amorphous carbon; (**b**) diffusion of carbon into Fe catalyst; (**c**) formation of ferrite (Fe_3C); (**d**) Formation of graphite on the other side of ferrite; (**e**) Growth of CNTs. Reproduced from ref. [55] with permission. Copyright 2017, Elsevier B.V.

Research showed that the type of catalyst and reaction conditions (temperature, other gases, etc.) have a significant impact on the morphology of CDM-derived carbons [56]. For a long time, the research focus of CDM has been on the efficiency of hydrogen production and catalyst lifespan [57], with less emphasis on regulating carbon morphology, optimizing its structures, and giving it a certain functionality. Actually, the as-obtained carbons are usually treated as the "by-product" of CDM (but its selling price matters a lot towards commercialization). Furthermore, research on using this type of carbon to manufacture rechargeable alkali-ion batteries is even rarer. The few existing papers suggest that optimizing the CDM process to alter carbon structure is a promising approach. For example, Ji et al. found that catalysts mixed with carbon material are conducive to producing disordered structures, which may reduce the conductivity of carbon [58]. We believe that potential optimization methods include selecting appropriate temperatures and catalysts to prevent the formation of amorphous carbon, regulating the degree of graphitization of carbon, and removing catalyst impurities present in nanocarbon. It is crucial to point out that this is an important direction for future researchers in this field.

For materials predominantly composed of carbon, defects [59] and dopants [60] can also be introduced to create moderate lattice distortion after pyrolysis, which improves the transfer kinetics of ions and electrons. Note that defect is an intrinsic property of nanocarbon deposits. Pei et al. also studied the interplay of these two factors, leading to a N- and-S co-dopant nanocarbon material with excellent efficiency and stability [61]. The structure can also be utilized to form composite materials that perform better than pristine ones.

4. Issues and Recycling of Waste Resin

Epoxy resin, a typical resin, has been widely used in various fields of production and life, such as machinery, electronics and the chemical industry, thanks to its excellent adhesive and stability performance [62]. However, as a

thermosetting polymer, the recycling of waste epoxy resin has become an intractable environmental problem, as the cured epoxy resin matrix has a three-dimensional crosslinking structure and is thus highly inert to most chemical interactions. Conventionally, both landfill disposal and incineration have been widely used for large amounts of waste epoxy resin in China; however, these methods have been proven even more harmful to both soil and air[63]. As such, the situation of other waste resins must also be staggering. Therefore, a comprehensive and circular economy technical route is urgently needed to alleviate the serious contamination by recycling instead of garbage disposal.

In order to retrieve chemical or value-added materials from the waste without excess pollution and energy consumption, today's researchers turn their eyes to resource recycling techniques [64], including pyrolysis [65] and chemical recycling [66]. However, most recoveries end up with low-end industrial products, such as simple aromatic compounds (e.g., benzene, toluene, ethylbenzene, etc.), metal oxides and mixed gases [67,68], which limits the economic profits and cannot reduce pollution effectively. Therefore, it is urgent and beneficial to propose novel recycling routes for waste epoxy resin, through which environmental pollution can be effectively reduced while transforming waste into treasure by preparing materials of high added value simultaneously. Additionally, as illustrated in Figure 3, a question still remains: Is it better to go with the direct or indirect route?
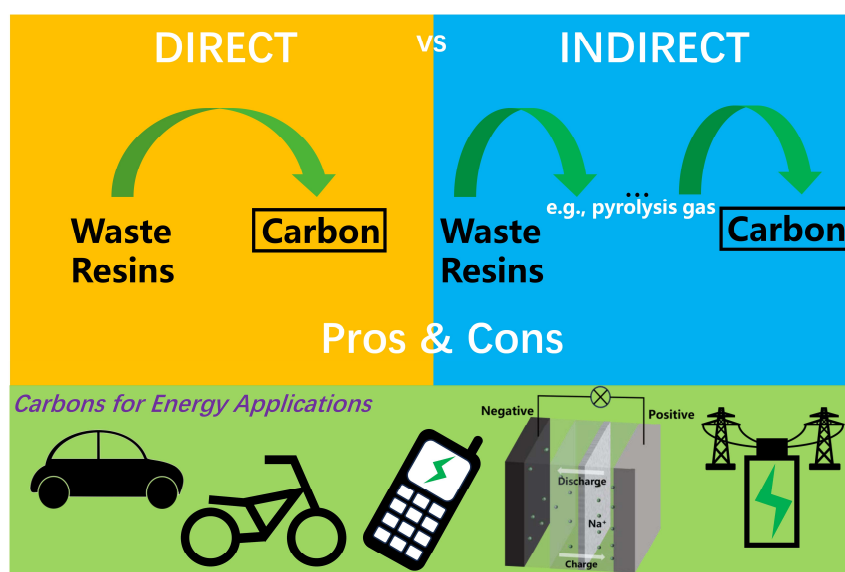

Figure 3. A simple drawing of the comparison between direct and indirect waste resin-to-carbon routes.

After analyzing the environmental crisis caused by the waste epoxy resin mentioned above, we will now discuss the advances in preparing SIB nanocarbon-negative electrodes using resin as a raw material. As mentioned earlier, some easily degradable epoxy resins can be broken down via a novel thermal solvent treatment and then recycled, while other "stubborn" epoxy resins undergo a pyrolysis procedure, which ends up with retrieving nanocarbon and hydrogen based on the catalytic decomposition of methane-containing waste pyrolysis gas. Therefore, this unique strategy could enable value-added recycling of resins and their composites towards a more "circular" resin industry.

5. Limitations and Potential Solutions

For carbon materials, generally, a large surface area (up to hundreds of square meters per gram) does not necessarily result in low ICE, as the key factor to reversibility is the structural stability of carbon and the quality of SEI. The effect of the surface area on the electrochemical performance remains unclear. Due to the growth mechanism of carbon on the surface of the Fe-based catalyst, the resultant carbon will encase Fe-based compounds and room-temperature acid treatment cannot fully remove it. Thus, concentrated nitric acid, for example, can be used to wash the mixture under heating conditions. However, this process will result in the uncontrolled introduction of impurity atoms due to the chemical reaction between carbon and acids and damage to the nanocarbon structure from the acid.

For further researchers either need to develop a method that can fully remove nickel without destroying the surface structure of carbon, such as base treatment, or replace the catalyst with a material that is not easy to mix with nanocarbon, such as molten metal or salts [69]. As relevant research is still lacking, extensive exploration is needed through independent studies.Besides, currently, it is still uncommon to use CNTs (obtained from CDM) alone as negative electrode material for sodium-ion batteries, as their unique framework is highly conducive to producing materials with

better performance. A more extensive literature survey should be conducted to identify the common structural characteristics of successful CNT-based electrode materials, such as SnS/CNT [70], Si/CNT [71], and N/S-doped CNT [72] materials. Based on this, optimization can be carried out on the resin-derived carbon to achieve potentially better performance.

Given that the authors have recently used mature catalysts within a conventional fixed-bed setup, it is understandable that this approach may result in lower performance levels compared to research specifically focused on producing high-performance carbons. However, in the near future, both the catalysts and reaction equipment will be further independently redesigned and improved. This is the reason for the limited production in a single day. For one thing, the CDM equipment is a custom-made, small device; secondly, the catalysts become completely inactivated within just 120 minutes and are not reusable, as carbon forms on the catalyst surface and encapsulates it. It is believed that large-scale industrialization is possible only after these two improvements are made.

In terms of the catalyst itself, since the Fe-based or Ni-based catalyst stands as a conventional material, whose typical limitations can be solved in later research, including but not limited to reusability [73] and longer lifetime (>200 min) [74] by means of structural regulation, additives and other methods. Also, the remaining catalyst left in the carbon influences both characterization (especially SEM). It may probably influence the electrochemical performance, which should also be solved, either by developing an efficient removal method or replacing the catalyst into a non-magnetic and easily-removed material.

6. Outlooks

To sum up, a brief review of epoxy resin recycling and carbon material-based SIBs was given. Here, we pointed out the potential improvements in existing researches, focusing on utilization of the waste as well as the significance of a new route for epoxy resin pyrolysis. Therefore, further researches can be done to (i) develop high-performance catalysts for the designed waste-to-carbon conversion; (ii) explore more effective ways for the structural and functional optimizations of the resultant carbons; (iii) utilize advanced characterization techniques for unveiling the sodium storage mechanism; (iv) design better reactors toward large-scale industrialization and so on.

The environmental pollution and waste caused by waste resins have become a severe environmental, social, and economic problem in China and around the world. For instance, the annual production of waste epoxy resin from electronic waste alone has exceeded 20 million tons. Returning to the concept of waste resin-to-treasure, it is worth pointing out its social, economic, and ecological benefits beyond electrochemistry. If the problems of production efficiency can be solved, the scaling of the technology would be alluring.

No doubt that considerable amounts of value-added hydrogen and carbon can be produced annually from this route, and the revenue is enormous. For electrochemical use, the profit can be even higher. The hydrogen produced can replace a portion of methane steam reforming (SMR) for hydrogen production [75]. With the same hydrogen produced, carbon dioxide emissions can also be reduced [76]. If the proposed technologies become sufficiently mature in the future to enable global recycling of waste resins, they could generate substantial economic, social, and carbon dioxide emission reduction benefits. Moreover, in addition to large amounts of waste resin, there are also vast amounts of biomass waste such as wet garbage, agricultural waste, food processing waste, livestock manure, municipal sludge, etc. The key technology proposed here can also be applied to the circular recycling of various types of waste and has broad application prospects.

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Author Contributions

Writing—Original Draft Preparation, J.T.; Writing—Review & Editing, Z.W.; Writing—Review & Editing, G.-M.W.; Supervision, G.-M.W.; Funding Acquisition, G.-M.W.

Ethics Statement

Not applicable.

Informed Consent Statement

Not applicable.

Declaration of 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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