Fish Industry Waste and Electrochemical Energy Systems

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Fish industry waste is attracting growing interest for the production of environmentally friendly materials for several different applications, due to the potential for reduced environmental impact and increased socioeconomic benefits. Recently, the application of fish industry waste for the synthesis of value-added materials and energy storage systems represents a feasible route to strengthen the overall sustainability of energy storage product lines.

fish industry waste activated carbon battery supercapacitors protein batteries carbon electrodes porous carbon fish waste valorization circular economy sustainability

1. Introduction

The energy crisis, climate change, increased energy consumption and growing awareness of environmental protection needs have imposed the challenge of sustainable development, pushing industrial and academic research toward efficient, clean, ecological and high-performance materials and equipment for energy storage and conversion [1]. The energy produced by renewable resources needs to be stored by electrochemical energy storage devices from which it can be extracted at a later time to perform necessary tasks [2]. These devices are required to have increasingly improved energy and power density. Moreover, electrochemical energy storage technology is crucial for the sustainable development of wearable electronics [3]. Therefore, it is essential to find high-performing, low-cost and environmentally friendly materials. Additionally, the developed materials should be able to be produced at a large-scale for usage in various industries [4][5][6][7][8].

One of the most invaluable, renewable and sustainable resources for the synthesis of high-performance materials for energy storage is biomass [9]. The term biomass indicates all renewable organic materials deriving from plants, algae, trees, crops, wood wastes, agricultural and forestry wastes, animal and poultry wastes, fishery and aquaculture waste and food processing waste [10][11]. In 2016, the total biomass waste in the world was approximately 550 gigatons of carbon, and is increasing every year [12]. This waste is either burnt or left in the ocean which leads to environmental pollution and the emission of greenhouse gas [13][14]. Biomass is exploited for energy production through thermochemical processes, including combustion, gasification and pyrolysis, and biochemical processes, including fermentation and anaerobic digestion [10][15][16].

Thanks to biomass’ recyclability, abundance and low cost, the application of biomass as a precursor to produce green carbon materials for energy storage is economically and technically sustainable [3][17][18][19]. Nowadays, many porous and nanostructured carbons derived from biomass present high conductivity, high tensile strength, low density and large aspect ratios, leading to enhanced energy storage capacity [20][21][22]. These carbon-based materials can be used in hydrogen storage [23], energy storage devices [24][25] carbon capture and storage [26][27], photovoltaics [28][29], dye degradation [30] and environmental remediation applications [31][32]. Essentially, the utilization of biomass not only helps to find inexpensive high-potential materials for different industries but also prevents or alleviates environmental pollution, providing opportunities for biomass-based industries [3][4][33][34].

Transformation of biomass into carbon-based materials can be done through carbonization, pyrolysis and activation techniques, producing enriched materials with high surface areas, vast pore volumes and small pore sizes [35][36][37][38][39]. Instrumental and methodological details regarding different synthetic strategies for biomass-derived carbon can be found in [40][41][42]. Due to a better electrolyte seepage and higher charge storage capabilities compared to conventional materials, porous and high-surface carbon materials are suitable as electrode materials for batteries and supercapacitors [43][44][45][46][47]. As a result, the research on preparation techniques and activator typologies has led to the concept of engineered biochar, wherein the physicochemical properties, performance and environmental benefits of pristine biochar can be tailored for specific applications [35][48]. For example, it is possible to derive a porous carbon with precise micropore size and large specific surface area (up to 3000 m^2/g) [4][33][49]. Additionally, chitin and chitosan, obtained from fish and crustacean shells, have been demonstrated to be effective as material for supercapacitors, LIBs, polymer electrolyte-based fuel cells and LSBs as polysulfide trapping agents [50][51][52].

Biowaste materials obtained from the fish industry have drawn significant attention as a novel raw material for various purposes. Around 50–75% of fish and seafood by-products, including viscera, skin, bones, scales, flesh, fins and shells are wasted during fish processing [53][54]. This waste occurs in huge quantities, considering that, in 2019, worldwide production of fish was estimated to be around 177.8 million metric tons (a number that will continually increase in the future) [55]. Of this amount, around 7.2–12 million tons are wasted yearly [56]. These waste products are discarded into the environment, in disposal areas or in the sea, with huge economic loss and detrimental effects on aquatic ecosystems, producing greenhouse gases and stench [57][58][59]. Fish waste disposal in the ocean increases organic matter content, leading to oxygen level reduction at the bottom of the ocean and endangering the lives of other oceanic inhabitants [60][61][62]. Discarding fish waste is a serious challenge that needs to be promptly overcome [53]. Consequently, the valorization of fish byproducts would be a great achievement, not only for the environment, but also for the fish and aquaculture industries [63][64].

Recent studies in literature reveal that fish industry waste can successfully be used as a low-cost precursor for the production of sustainable energy storage materials, since it is a rich source of carbon, nitrogen, oxygen, hydrogen and sulfur [60][65][66]. Moreover, biomass derived from fish waste includes a valuable amount of collagen, crude protein and amino acids, which are a great choice for preparing 3D and N-doped nanoporous carbon materials [39][67]. Fish scales, for example, contain organic and inorganic materials (collagen fibers and calcium-deficient hydroxyapatite, respectively) [68]. The organic parts of fish scales can be converted to carbon matrices; the inorganic parts may be a natural template to induce a chain porous structure after carbonization and activation [9][69][70][71]. Additionally, fins and fish skins contain valuable amounts of collagen fibers, including different amounts of carbon, oxygen, nitrogen, hydrogen and sulfur. Finally, the annual generation of around 0.5 million tons of crab shells makes crab shells another valuable source of material for energy storage devices. This amount of waste is much higher than the material produced for LIBs (about ten thousand tons for both anodes and cathodes) [72].

The increasing interest toward the valorization of fish waste into sustainable materials for electrochemical storage systems is highlighted by the growing number of scientific publications on this topic during the last decade and, in particular, in the last three years, as reported in **Figure 1**.

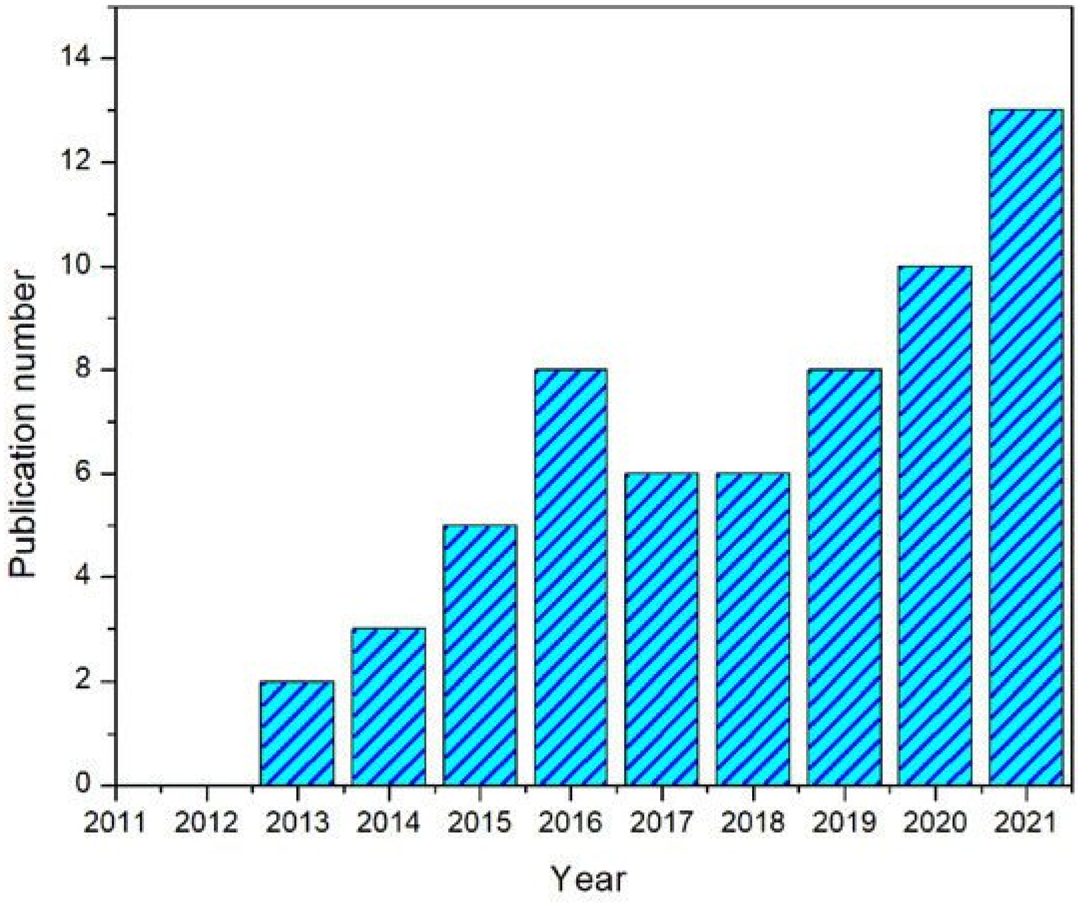

Figure 1. Distribution of scientific publications per year on sustainable materials derived from fish waste for electrochemical storage systems (Scopus database).

2. Applications in Lithium–Ion Batteries (LIBs)

To date, the most developed electrochemical energy storage devices are lithium-ion batteries (LIBs), which are currently applied in various fields including smartphones, laptops and electric vehicles, owing to their relatively high energy density and long cycle life [73][74][75]. Nevertheless, commercial graphite anodes cannot satisfy the increasing demand of the high energy density in LIBs. Predictions have claimed that the demand for lithium will be tripled by the year 2025 [76]. Another limitation is represented by the massive anode volume changes during Li^+ insertion and extraction, which leads to the pulverization of the lithium–alloy particles and fast capacity drop during charge-discharge cycles [77]. To overcome these limitations, research on alternatives for graphite anodes has focused on nanoporous carbons (NPCs). NPCs have drawn interest because of their potentially higher specific capacity and stability and their well-organized porous structure. These can prepare rapid ion diffusion channels, which is advantageous when attempting to obtain high Li^+ storage capacity [35]. A variety of NPCs have been investigated, such as carbon nanofibers [78][79] carbon nanocages [80][81], nitrogen-enriched nanocarbons [82][83], etc. Their porous structures can reduce the diffusion length of Li-ions, while their high specific surface area offers abundant active sites for Li^+ storage reactions [84].

Recent literature demonstrates that fish waste can successfully be used as a sustainable source for nanoporous carbon materials; it is enriched with elements such as nitrogen, oxygen, hydrogen and sulfur, and characterized by cost-effectiveness and thermal stability. Depending on the type of fish industry waste, different routes have been reported in recent studies for obtaining porous carbon electrodes for Li-ion batteries, as schematically reported in **Figure 2**.

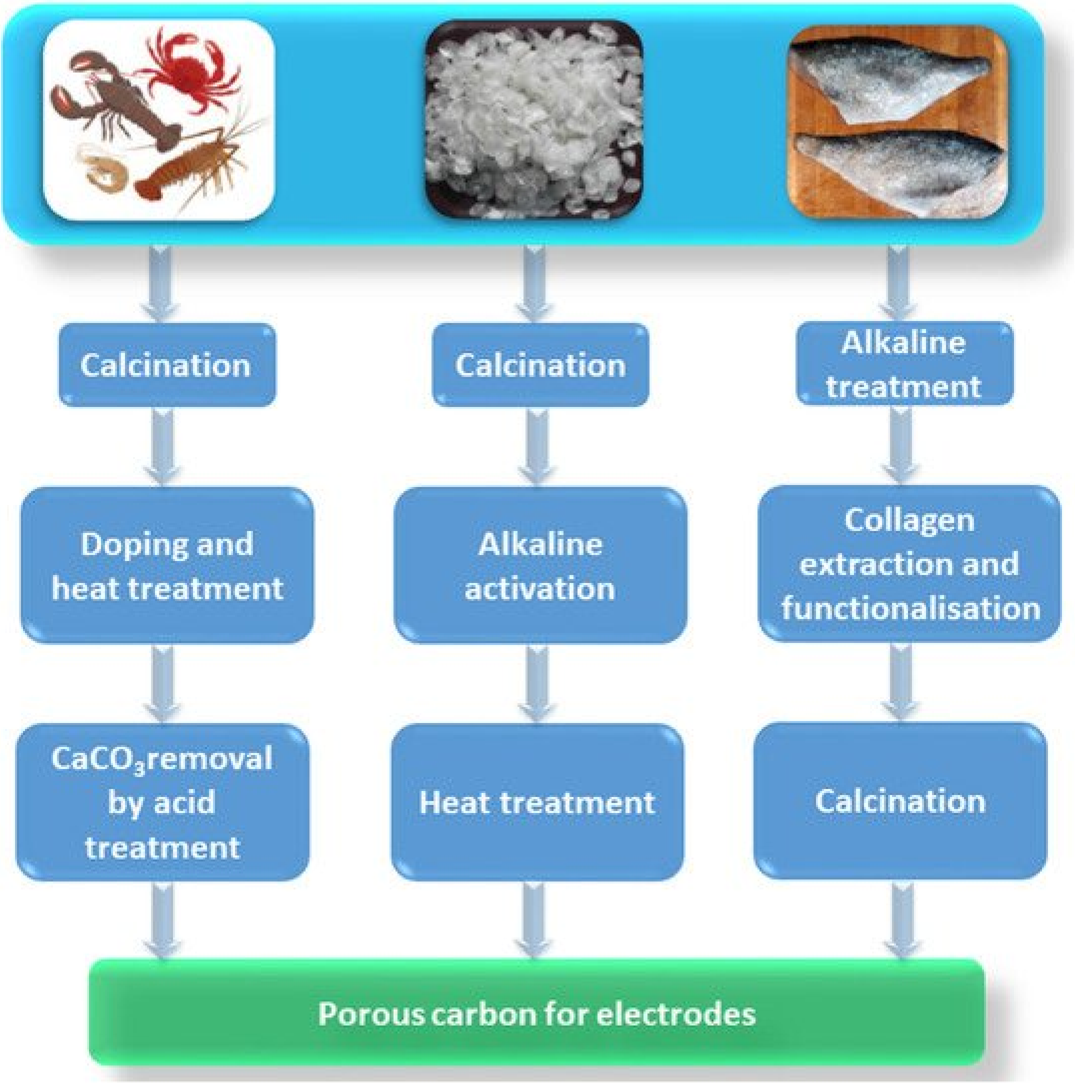

Figure 2. Schematic representation of the possible routes for obtaining porous carbon electrodes for Li-ion batteries from fish waste.

Crustacean shells are effective biotemplates for preparing nanostructured anodes for rechargeable Li-ion batteries, as demonstrated by Yao et al. [72]. They obtained hollow carbon nanofibers from crab shells encapsulating sulfur and silicon. The processing route involved several steps, as schematized in **Figure 2**. After air calcination of the crab shells, the organic components were removed and $CaCO_3$ templates containing twisted hollow nanometric channels were obtained, with diameters close to those of commonly anodized aluminum oxide templates. The $CaCO_3$ framework was coated with a thin layer of carbon via heat treatment in nitrogen. Then, the obtained active electrodes were inserted into the nanochannels, where they were treated by sulfur and silicon through thermal infusion and chemical vapor deposition, respectively. After dissolving the $CaCO_3$ framework by acid treatment, the researchers obtained hollow carbon nanofiber arrays encapsulating sulfur or silicon. The hollow nanostructures provided sufficient space for the volume expansion of sulfur/silicon during the discharge/charge processes and the thin walls of the hollow carbon nanofibers allowed rapid lithium-ion transport from the electrolyte to sulfur/silicon. As reported in **Table 1**, the Li-ion battery prepared with this crab shell-templated carbon/silicon anode showed high specific capacity (1580 mAh/g at 1C) and high cycling performance [72].

The mechanism responsible for the excellent electrochemical performance of fish waste-derived porous carbon materials in LIBs occurs due to their uniform interconnected porous structure, which is beneficial for the rapid penetration of electrolytes, fast Li^+ diffusion and the provision of active sites for the storage of Li^+ ions [85]. The electrochemical properties of electrode materials can be improved by heteroatom doping, which induces defects and increases available active sites [86]. Nitrogen-doped porous carbons derived from crawfish shells were prepared by Wang et al. [87] by modifying the initial calcination treatment in a nitrogen atmosphere, followed by acid treatment to eliminate $CaCO_3$. Then, N-doped porous carbon underwent a thermal treatment with cobalt acetate tetrahydrate to become nano-filled with nanometric cobalt oxide (Co_3O_4) nanoparticles. The N-doped porous carbon and Co_3O_4-N-doped porous carbon were used to prepare a working electrode via a slurry coating procedure with high electrochemical lithium storage performance. N-doped porous carbon had a capacity of about 400 mAh/g after 100 cycles, which was greater than that of commercial graphite (372 mAh/g). This demonstrated that N-doped porous carbon could potentially replace graphite in industrial production. More interestingly, as reported in **Table 1**, the N-doped PC-Co_3O_4 nanocomposite with 10 nm Co-based nanofiller presented a high reversible capacity of 1060 mAh/g after 100 cycles, acceptable rate capability, superior cyclic performance and excellent primary Coulombic efficiency (86.7%) [39][67].

In addition to the use of prawn shells (PSC), prawn meat (PMC) was used by Lian et al. [85] to prepare porous carbon materials to be applied as anodes in lithium-ion batteries. After calcination under an inert atmosphere, the obtained carbon structure was washed, centrifuged, dried and then combined with polyvinylidene fluoride (PVDF), acetylene black (AC) and N-methyl-2-pyrrolidone solvent. The initial discharge/charge capacities of PSC and PMC materials for the first 3 cycles at the current density of 30 mA/g were 1803/910 and 1200/694 mAh/g with the coulombic efficiency of 50.4% and 57.8%, respectively (see **Table 1**). Solid electrolyte interface formation was cited as the reason for the low initial coulombic efficiencies. For PSC and PMC, coulombic efficiency reached 91% and 93% after the first cycle and 94% and 95% after the third cycle, respectively [85]. The best performance was obtained by PSC due to the presence of a more uniform nanoporous structure compared to PMC and a higher level of N-doping.

The steps for the preparation of porous carbons from fish scales are schematized in **Figure 2**. As reported by Selvamani et al. [22], after air calcination, activation in alkaline solution and heat treatment, the obtained carbon was characterized by a high specific surface area and excellent electrochemical behavior, even under high charge/discharge situations. The galvanostatic charge/discharge curves at the current density of 75 mA/g demonstrated an initial discharge capacity around 541 mAh/g in ionic liquid electrolyte. After 75 cycles, the coulombic efficiency was 94% with a reversible capacity of 509 mAh/g. At the current densities of 400 and 4000 mAh/g, the reversible capacities were 390 mAh/g and 179 mAh/g, respectively. In addition to the aforementioned properties, the electrode was stable before and after cycling [22].

Very recently, it was demonstrated that collagen extracted by fish waste could be used to obtain porous materials for LIB electrodes, as schematized in **Figure 2**. Odoom-Wubah et al. [60] extracted collagen from Tilapia fish with an alkaline treatment followed by an acid treatment. The marine collagen was impregnated by Palladium nitrate followed by a heat treatment in nitrogen and then used in combination with polyvinylidene fluoride (PVDF), N-methyl-2-pyrrolidone and carbon black as an anode material for Li, Na and Mg half-cells. Results of electrochemical measurements revealed that the reversible capacities for Li, Na and Mg-based cells were 270 (**Table 1**), 120 (**Table 2**) and 100 mAh/g, respectively [60]. The proof of concept of extracted porous carbon from marine collagen was demonstrated, but further studies are still required to optimize the preparation and performance.

In **Table 1** the performance of porous carbon-based materials obtained from fish industry waste for LIBs is compared with those of commercial graphite-based anodes for LIB in terms of specific capacity and cycle life. This latter indicates the number of charge/discharge cycles of the battery until the end of its lifetime. For LIBs, the cycle life is significantly dependent on the depth of discharge, which is an indication of the amount of storage capacity of the battery. It is typically in a range between 300 and 500 cycles for commercial LIBs, even if some manufactures have claimed 1000 cycles [88].

Table 1. Performance of porous carbon-based materials obtained from fish industry waste for Li-ion batteries.

Fish Waste Source	Application	Current Density (mA/g) or C-Rate	Initial Discharge Capacity (mAh/g)	Reversible Specific Capacity (mAh/g)	Capacity Retention	References
Crab Shell	Si-encapsulated nanostructured anode	C/10-1C		3060 @C/10 1580 @1C	95% @200 cycles	[72]
Crawfish shell	nanoCo_3O_4 doped anode	100	1223	1060	98% @100 cycles	[87]
Prawn shells	anode	50–1000	1735	950 @50 mA/g	84% @90 cycles	[85]

Fish Waste Source	Application	Current Density (mA/g) or C-Rate	Initial Discharge Capacity (mAh/g)	Reversible Specific Capacity (mAh/g)	Capacity Retention	References
				300 @1000 mA/g		
Prawn meat	anode	50–1000	1132	420 @50 mA/g 100 @1000 mA/g	40% @90 cycles	[85]
Prawn Shells	anode	0.1	740	732	99% @150 cycles	[89]
Fish scales	N-doped nanoporous anode	75 400 4000	541 418 214	509 390 179	94% @75 cycles 93% @75 cycles 84% @75 cycles	[22]
Collagen from Tilapia waste	nanoPd doped anode	1C	600	270 @1C	100% @20 cycles	[60]
Crab Shell	anode	50	1758	703 @50 mA/g	83% @200 cycles	[90]
	Commercial graphite-based anodes			372 theoretical	300–500 cycles	[91][92]

3. Applications in Sodium-Ion Batteries (NIBs)

Li-ion batteries cannot meet the growing needs of the energy storage market because Li is an expensive, limited and unequally distributed resource [73][93]. Sodium-ion rechargeable batteries are attracting great attention due to their similarity to LIBs and the use of sodium ions (Na^+) as the charge carriers [94]. Though sodium cannot compete with lithium’s energy density, that shortcoming is compensated due to its availability and price [95]. Therefore, compared to more widespread LIB, Na-ion batteries (NIB) have lower cost and do not use scarce resources [96]. However, sodium has two disadvantages. First, its weight is three times higher than lithium; even if only 5% of the overall battery weight is related to lithium, NIBs are heavier. In addition, the Na^+ ion has a larger ionic radius than the Li^+ ion, leading to more sluggish diffusion kinetics and more significant volumetric changes during repeated charging/discharging cycles. Therefore, Na-ion batteries are less powerful, primarily due to the low ability of the graphite anodes to absorb sodium. A possible solution for achieving higher storage capacities could be the

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replacement of the graphite anodes commonly used today with electrodes of graphene or hard carbon [97]. The latter is a disordered mainly sp^2, non-graphitic carbonaceous material consisting of single layers of carbon atoms that are arranged in a planar hexagonal network, but irregular and disordered along the c-axis [98][99]. These carbon-based materials can be obtained from biomass [47][100][101].

Table 2. Performance of porous carbon-based materials obtained from fish industry waste for Na-ion batteries.

Fish Waste Source	Application	Current Density (mA/g) or C-Rate	Initial Discharge Capacity (mAh/g)	Reversible Specific Capacity (mAh/g)	Capacity Retention	Ref.
Prawn Shells	Na-ion batteries anode	100	370	325 @1C	100% @200 cycles	[89]
Fish collagen (Tilapia)	nanoPd doped anode for Na-battery	1C	60	NIB: 120 @1C	40% @20 cycles	[60]
Crab Shell	anode	50 mA/g	1211	283	62% @300 cycles	[90]
	Commercial graphite-based anodes	25	250	184 @C/10	100 cycles	[102] [103]

Recent studies have reported the suitability of porous carbon-based materials obtained from fish industry waste as electrodes for NIB batteries. Previous investigations demonstrated that by a low cost, simple and environmentally friendly approach, nitrogen-doped hierarchically porous carbon material obtained from prawn shells could be obtained. The mechanisms underlying their great suitability for replacing common carbon electrodes is due to their porous structure, high inherent nitrogen content and the presence of macro, meso- and micropores that facilitate the storage and transport channels for Li and Na ions. Detailed investigation on this subject was conducted by Elizabeth et al. [89], who obtained a porous N-doped structure from prawn shells using a protocol similar to that reported in Figure 2. Experimental results revealed that, due to the high Nitrogen content in porous carbon material, electrical conductivity and active sites for Li/Na storage were increased, which led to an improved electrochemical performance. Galvanostatic charge/discharge tests showed that the initial capacity at the current density of 0.1 A/g was 1013 mAh/g, which was three times more than the capacity of conventional graphite carbon material. The formation of solid-electrolyte interphase and the irreversible trapping of Li in the pores were responsible for the heavy capacity fade at the beginning. Cyclic charge/discharge tests were conducted on the porous carbon as an anode material for sodium-ion batteries. In the first cycle, the charge specific capacity was around 660 mAh/g and the discharge capacity was 370 mAh/g with coulombic efficiency of 56%. This low coulombic efficiency was due to the irreversible capacity loss because of the solid-electrolyte interphase formation and Na trapping in the porous structure. At the current density of 0.4 A/g, the electrode reversible capacity was 234 mAh/g after 150 cycles, which was superior in comparison to other biomass-derived carbon materials used in the literature. This was ascribed to the hierarchical porous structure and N-doping of carbon. Moreover, TEM results

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also confirmed that, after 200 cycles, the porous structure showed little damage, which was an indication of the high structural stability of this electrode material [89].

In addition to the porous carbon derived from crustacean shells, suitable electrode materials can be obtained from fish waste, as demonstrated by Odoom-Wubah et al. [60], who extracted collagen from Tilapia waste (according to a procedure described in the previous paragraph) to obtain an anode material for NIB, achieving a specific capacity of 120 mAh/g. Other investigations have been performed to apply sustainable anode materials obtained from fish collagen to Magnesium-ion batteries (MIBs) [60], or from seafood-derived chitin for Potassium-ion batteries (KIBs) [104], respectively. Reversible capacities of 105 mAh/g [60] and 154 mAh/g [104] were obtained for MIBs and KIBs, respectively.

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