

From plastic waste to new materials for energy storage

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The use of plastic waste to develop high added value materials, also known as upcycling, is a useful strategy towards the development of more sustainable materials. More specifically, the use of plastic waste as a feedstock for synthesising new materials for energy storage devices not only provides a route to upgrading plastic waste but also can help in the search for sustainable materials. This perspective describes recent strategies for the use of plastic waste as a sustainable, cheap and abundant feedstock in the production of new materials for electrochemical energy storage devices such as lithium batteries, sodium batteries and supercapacitors. Two main strategies are described, the development of conducting carbons by combustion of plastic waste and the depolymerisation of plastics into new chemicals and materials. In both cases, catalysis has been key to ensuring high efficiency and performance. Future opportunities and challenges are highlighted and hypotheses are made on how the use of plastic waste could enhance the circularity of current energy storage devices.

Introduction

The use of plastic waste to develop high added value materials has emerged as a promising strategy towards improving sustainability. Global plastic consumption has increased to 359 million metric tons in 2018, with almost 50% of these plastics used in short-term applications.¹ The constant increase of plastic consumption and the relatively poor management of plastic waste (only 32.5% of the collected plastic from municipal solid waste (MSW) in Europe (2019) and 8.7% in the United States (2018) is recycled) is causing increasingly obvious environmental problems. With more than 368 millions tons of plastic produced in 2019 and less than 10% recycled worldwide, the plastic industry is sadly well-known for being one of the most contaminating.¹⁻⁴ This idea is reinforced by images of marine animals trapped in plastic bags, African beaches covered by synthetic textiles and the accumulation of plastic waste in Asian rivers frequently shown in the media. Consequently, the treatment of discarded plastics has become a global challenge and the scientific community is exploring all kinds of recycling technologies to better manage this waste. These strategies range from recycling methods as simple as mechanical repurposing or incineration to more complex processes, such as chemical recycling or enzymatic scission.

Indeed, one promising alternative for managing plastic waste resides in the concept of upcycling, which can be defined as “the use of plastic waste (*i.e.* post-industrial or post-consumer usage) as a feedstock for the synthesis of innovative products with added-value for alternative applications”.^{5,6} One potential value-added application is energy storage including next generation of batteries and supercapacitors. As the world has entered the so-called “fourth industrial revolution”, the development of energy storage solutions is at a crossroads with the emergence of new technologies such as the Internet of Things (IoT), robotics, electric unmanned aerial vehicles, “smart grids” and the electrification of the transportation sector. These technological shifts are driven by environmental and sustainability concerns (*i.e.* global warming, peak oil), but also by the constant increase in the world population and energy consumption demands. Nowadays, lithium-ion batteries are the current flagship technology in various applications including mobile devices, electric vehicles, and even stationary energy storage, where high energy density energy storage solutions are required. However, the continuous increase in energy demands, and thus, in energy storage devices of all kinds, has induced pressures on the battery market, with several raw materials used in lithium-ion batteries (LIBs) (*i.e.* lithium, cobalt, nickel, graphite) now recognised as critical raw materials in the recently updated list from the European commission.

Considering this background, the development of low-cost, environmentally friendly materials is a necessity to sustainably answer energy demands. Several recent reviews have been focused on the management of discarded batteries and

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although several challenges must be overcome for proper materials recycling, there is a clear need to limit dependence on finite resources for battery fabrication.^{7–11} Many of the components of batteries are high performance synthetic polymers derived from fossil resources such as poly(vinylidene difluoride), polyolefin porous separators or poly(ethylene oxide). Research is active to replace these petroleum-based synthetic polymer derivatives by biomass-derived organic materials. To date, various biomass derivatives, such as carboxymethyl cellulose, cellulose nanofibres, lignin, melanin, juglone and humic acid, have been explored as binders, polymer electrolytes or active electrode materials, and have shown great promise for application in supercapacitors or rechargeable batteries.

Considering the exponential growth of the battery market, the valorisation of plastic waste that would otherwise be burnt or put into landfill, represents an opportunity. Such approach will provide alternative end-of-life management for plastic, complementing the European/governmental incentives towards a carbon neutral and circular economy for plastic, as highlighted by the circular economy action plan of the European Union.¹² In the framework of the European green deal, the battery sector has been pointed out as one of the most finite resources consuming and wastes producing sector. As such, sustainable criteria are being implemented for transitioning towards circular production and consumption models which includes the use of alternative resources and the promotion of recycling.¹³

This perspective is focussed on the development of novel materials for energy storage applications from plastic waste. Two main trends are observed which includes first the development of conducting carbons by combustion of plastic waste. The second one consists in the chemical recycling of plastic waste into new chemicals and materials as components for battery cathodes, anodes or electrolytes. (Fig. 1) The use of new materials generated from plastic waste will be described in terms of sustainability and considering potential applications in different energy storage technologies such as LIBs and supercapacitors. Moreover, the use of these materials in emerging battery technologies such as sodium batteries, all organic batteries and solid-state batteries will be discussed.

Carbon nanostructures by combustion of plastic waste

The most widely investigated approach to upcycle plastic waste for energy storage applications is through combustion to produce carbonaceous materials.^{14–18} Carbon materials with large specific surface area and high electric conductivity are commonly used in electrochemical energy storage. Indeed, the current LIB technology already uses a carbonaceous material as anode electrode, namely graphite, which is generally produced either through mining or synthetically *via* the carbonisation of unsaturated petroleum derivatives. Both production methods use finite resources as a feedstock, which has led to some concerns regarding future supply issues, particularly given the expected exponential growth in demand for LIBs

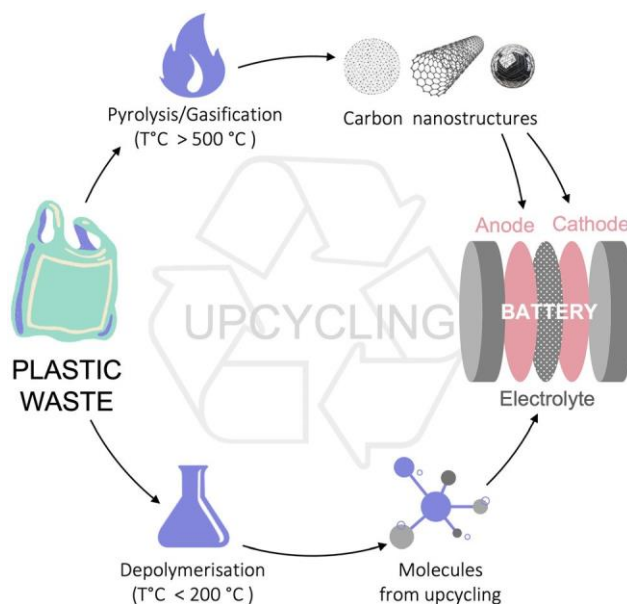


Fig. 1 The two main trends for the transformation of plastic waste into materials for batteries.

over the next 10 years. Consequently in the 2000's, the growing interest for producing such carbonaceous materials led to preliminary studies demonstrating how polymers, and especially polyolefins, could be used as a feedstock for their synthesis.^{14–16} The combustion of polypropylene (PP) for obtaining carbon nanotubes was assisted by nickel-based catalysts^{19–21} or mixtures of cobalt and ferrocene²² for example. However, the objective of these articles was less focused on the treatment of plastic waste and more directed towards low-cost alternatives for the synthesis of active carbonaceous frameworks.

The first studies conducted with the claimed objective of upcycling plastic waste into materials for electrochemical energy storage was carried out by Pol and Thiyagarajan in 2010.²³ In this study a wide range of waste polymeric materials *e.g.*, low density polyethylene (LDPE), high density polyethylene (HDPE), polyethylene terephthalate (PET), polystyrene (PS), and mixtures were thermally treated at 700–800 °C for 1 to 3 h in order to obtain conducting and paramagnetic carbon microstructures. Depending on the nature of the starting materials and the thermal treatment parameters, the morphology of the carbonaceous product varied from spheres to nanotubes. The obtained products, which were characterised by TEM (Transmission Electron Microscopy), Raman spectroscopy and XDR (X-Ray Diffraction), were considered suitable for use as anode materials for LIBs. It was previously demonstrated that to increase the graphitic order and, consequently, the properties of the final material, the addition of a large quantity of cobalt acetate as catalyst was necessary.²⁴ Used as anode material in LIB, these carbon nanotubes have demonstrated a stable capacity of around 240 mA g⁻¹ at 0.2 A g⁻¹ for at least 200 cycles.²⁵

In 2019 Lian *et al.* synthesised porous carbon from PE waste such as plastic bags by ball milling and carbonisation in the presence of magnesium carbonate pentahydrate as flame retardant.²⁶ This compound was proven to be key for the formation of porous carbon structures not only because it serves as template but also because it greatly improves the thermal stability of PE towards carbonisation. In a second step, after ammonia activation, high specific surface area and mesoporosity were achieved. As supercapacitors, the so-obtained material showed a remarkable capacitance and good cycling stability: a high value of energy density of 43 Wh kg⁻¹ was assessed at 4 V which was attributed to the high purity and low O and N contents. In addition, 97.1% capacitance was observed after 10 000 cycles at 2 A g⁻¹. These promising results were a considerable improvement in comparison with previous results reported in literature, demonstrating the potential of PE waste as raw material.

Similarly, Zhang *et al.* employed LDPE as precursor of well-defined porous carbon spheres by autogenic pressure carbonisation and KOH activation.²⁷ Even in catalyst absence, owing to high pressure, 45% carbon yield was obtained when the ulterior KOH activation creates hierarchical porous matrixes which can be tuned to control specific surface area. This material was tested as electrode for being used as supercapacitor. As the electrochemical characterisation revealed high specific capacitances were obtained, up to 355 F g⁻¹ at a current density of 0.2 A g⁻¹ in 6 M KOH electrolyte and energy density of 9.81 Wh kg⁻¹. This article provides a new pathway to transform LDPE into high added value materials for supercapacitors instead of the classical incineration.

In a similar approach, Chen and co-workers converted PET into carbon microspheres by employing supercritical CO₂.²⁸ In this study the reaction temperature was decreased to 500–650 °C, making the process less energetically demanding while keeping similar reaction times (3 h). It was demonstrated that high reaction temperatures and long reaction times promoted graphitisation, yielding higher amounts of carbon microspheres (47.5%) at 650 °C for 9 h. The analysis of different samples by GC-MS showed that short reaction times promoted the formation of aromatic hydrocarbons, while longer reaction times were required for their conversion into carbon microspheres. Their applicability as anode materials for LIBs was evaluated, delivering an initial discharge capacity of 505 mA h g⁻¹ at a current density of 0.1 A g⁻¹ and 247 mA h g⁻¹ at a current capacity of 0.16 A g⁻¹, of which 40% remained after 20 cycles.

PET can also be carbonised in a control manner to produce three dimensional porous carbon nanosheets to create capacitors.²⁹ In a process developed by Mu *et al.*, PET is carbonised in a reaction kettle at 700 °C in the presence of MgO/Co(acac)₃ as template catalyst, which was previously prepared by mixing MgO and Co(acac)₃ in a ball mill in a ratio 2 : 1. The authors report the selectivity of this catalyst to yield 36.4 wt% of highly pure nanosheets. Later the obtained nanosheets were mixed uniformly with MnO₂ nanoflakes in a redox reaction process to provide the PCS-MnO₂-² final composite which demonstrates

superior performances as supercapacitor. Owing to its very high specific surface area and porosity in conjunction with the adequate MnO₂ load a gravimetric capacitance up to 210.5 F g⁻¹ were achieved with good cycle stability, showing the importance of the catalyst choice on the morphological control of the products and their ulterior electrochemical properties.

To further improve the properties of materials for capacitors obtained from recycling, the direct obtention of doped materials must be considered. Kashyout co-workers showed how the thermal treatment of PET wastes with urea can lead to 3D nitrogen doped graphene.³⁰ Experimentally, raw PET is mixed with urea in 1 : 1–2 ratios in an autoclave before being introduced in an oven at temperatures of 600–800 °C to produce N-doped graphene. The results provide insight into the effect of synthetic conditions such as temperature or urea content among others impact on the morphology, specific surface areas or surface functionalisation. In comparison with previous works, capacitance as high as 405 F g⁻¹ at 1 A g⁻¹ were obtained. Maximum power density of 558.5 W kg⁻¹ and energy density of 68.1 Wh kg⁻¹ were reported in a KOH electrolyte with a proper cycle stability even after 5000 cycles at 4 A g⁻¹, demonstrating the high impact of the nitrogen doping.

More recently, Min *et al.* reported the possibility to obtain well-defined 3D porous carbon frameworks from mixtures of plastic waste containing HDPE, LDPE, PS, PP, and Polyvinyl chloride (PVC) by employing a general structural template MgO/Tris(acetylacetonate) iron III (Fe(acac)₃).³¹ The carbonisation process at 700 °C under argon atmosphere led to conversion over 70% (Fig. 2). The XRD analysis revealed that Fe (acac)₃ was converted into Fe₃O₄ nanoparticles during the carbonisation process, which resulted in the formation of hollow carbon sphere architectures, while the MgO template both supports and catalyses the reaction. This unique 3D structure was used as anode material in a LIB, demonstrating excellent performance with specific discharge capacity as high as 802 mA h g⁻¹ after 500 cycles at a current density of 0.5 A g⁻¹. Previous work from the same group has also demonstrated the possibility to transform PS waste using only manganese oxide. The resulting porous material had notable electrochemical properties, *i.e.* 247 F g⁻¹ at a current density of 1 A g⁻¹ with a high cycling stability of over 93% after 10 000 cycles at 10 A g⁻¹.³²

In a recent paper, Fonseca *et al.* described the carbonisation of PS waste into electrode materials for another type of battery: Sodium ion batteries (SIBs).³³ The high temperature (*i.e.* 700 °C) and high pressure applied in the reported process allowed for the conversion of the plastic into carbonaceous material with a low graphitisation content. Contrary to what is required for LIBs, batteries based on sodium ions requires higher interlayer distances than what it usually found with low graphitisation degree, because of the larger size of the sodium ion (0.116 nm) compared to lithium ion (0.076 nm). For this reason, current graphite anode materials developed for LIBs cannot be employed in Na-ion batteries. Thus, the possibility of producing carbonaceous anode materials, which are

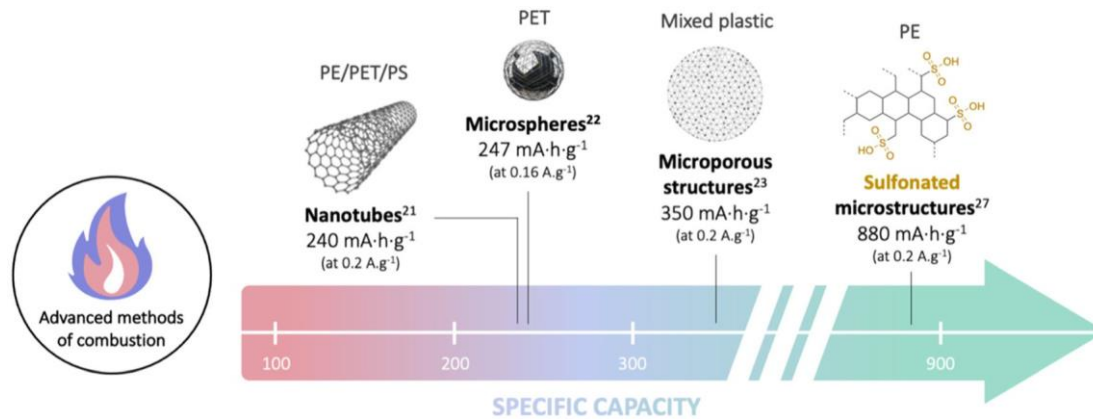


Fig. 2 Advanced combustion methods of plastic waste for obtaining carbon materials for energy storage devices and their performances in lithium batteries.

suitable for Na-ion batteries and produced from low-cost feedstock such as plastic waste, is very appealing from both economic and environmental perspectives. The material was investigated as anode material for SIB and demonstrated a specific capacity of 116 mA h g^{-1} at a current of 20 mA g^{-1} , with good stability for at least 80 cycles.

Similarly, the carbonisation of PVC was performed by Bai *et al.* with the aim of obtaining suitable materials for SIBs.³⁴ Hard carbons obtained from the pyrolysis of PVC nanofibres (previously dissolved and deposited by electrospinning) at 600, 700 or 800 °C demonstrated suitable properties for such applications, especially the material obtained from the pyrolysis at 700 °C which had an initial reversible capacity of 271 mA h g^{-1} at 12 mA g^{-1} , retaining 215 mA h g^{-1} after 120 cycles. In this article, the comparison with the material obtained from the pyrolysis at 700 °C of PVC particles was unequivocal as only a reversible capacity of 206 mA h g^{-1} was attained at 12 mA g^{-1} , with a rapid decrease up to 126 mA h g^{-1} . The authors claimed that the high cycling performance was possible because of the moderate graphitisation, the small particle size and large interlayer distance of the resulting material, allowing the proper insertion of sodium ions.

In a similar way, Sun co-workers converted PVC in carbonaceous porous residues.³⁵ In contrast with previous example in this case PVC was first dehalogenated through KOH in DMSO or DMF solutions at room temperatures prior to be annealed at 600 °C to obtain the final carbonaceous porous material. This procedure was first applied to virgin PVC before being applied to common wastes such as plastic wraps. Interestingly the carbon collected from plastic wraps showed an outstanding performance as aqueous symmetric supercapacitors providing values of 399 F g^{-1} at 1 g^{-1} in 6 M KOH electrolyte demonstrating the potential of upcycling PVC for energy storage supercapacitors devices.

To improve further the cycling performance of the carbonaceous anode materials, the use of additives such as organic or inorganic compounds and structural templates could be necessary. Recently Pol and co-workers reported a strategy to

treat LDPE *via* microwave irradiation to create sulfonated scaffolds that can be applied to lithium-sulphur batteries.³⁶ First, LDPE plastic bags were sulphonated by placing them in sulphuric acid at 100–120 °C under microwave irradiation. The microwave treatment not only increased the rate of the sulphonation reaction but also created pores on the plastic, thus increasing the specific surface area. In a second step, the sulfonated LDPE was washed with water and dried prior to being carbonised at 900 °C under an inert atmosphere for 2 h. The resulting material, containing highly negatively charged sulphonated groups, was investigated as anode material for LIBs, delivering an excellent specific discharge capacity of 979 mA h g^{-1} at 0.5 C (around 0.1 mA g^{-1}) and a capacity retention of 79% after 200 cycles. In a subsequent study, the same procedure was applied to both HDPE and LDPE to yield amorphous carbon chips for LIB anodes.³⁷ The electrochemical study of the material demonstrated a reversible capacity of 230 mA h g^{-1} at 0.2 C for the material derived of LDPE and 350 mA h g^{-1} for this of HDPE.

Later, Lian *et al.* studied the upcycling of PE waste to graphene mesoporous carbon for high voltage supercapacitors.³⁸ In this work plastic bags were blended with $4\text{MgCO}_3\text{-Mg(OH)}_2\text{-5H}_2\text{O}$ until a homogeneous powder was obtained, which was subsequently mixed with graphitic oxide prior to being carbonised. Several graphitic oxide contents and carbonisation temperatures were investigated to find optimal conditions at 2.5 wt% of graphitic oxide and 700 °C. Morphological studies by means of Scanning Electron Microscopy (SEM) revealed the coexistence of graphene and agglomerated mesoporous particles that were irregularly distributed creating a very high specific surface accessible by the electrolyte ions. This strategy was proven to be an efficient way to obtain mesoporous materials suitable for use as electrodes in aqueous electrolytes, ionic liquids, and hybrid Li_2MnO_4 based supercapacitors, offering good performances, up to 114 F g^{-1} specific capacitance at 1 A g^{-1} or 76 F g^{-1} at 5 A g^{-1} , maintaining over 89% of this capacitance for 5000 cycles at this current density.

New chemicals and materials from depolymerisation of plastic waste

The second approach for upcycling plastic waste into high-performance materials for batteries is based on the depolymerisation of waste polymers to specific chemical sequences. This alternative broadens the scope of the possibilities for materials synthesis and catalyst development. Several organic materials are investigated in different components of energy storage devices such as the cathode, anode, binder, and the electrolyte. The transformation of plastic wastes into alternative chemicals to fossil-based resources could have an important impact on the overall sustainability of energy storage devices.

In 2020, Ghosh *et al.* synthesised disodium terephthalate (Na_2TP) from waste PET bottles as the main material for making anodes for SIBs and LIBs.³⁹ The microwave-assisted depolymerisation of PET polymer yielded Na_2TP in very short times (<5 min). The anodes made of this simple organic compound mixed with carbon black (Super P) were electrochemically analysed and revealed promising results. Discharge capacities of 182 and 224 mA h g^{-1} at a current density of 25 mA g^{-1} were found for Li-ion and Na-ion cells, respectively. The use of terephthalate derivatives is an interesting alternative because it is rapid and easy to produce from the upcycling of PET at mild conditions. In a similar work, Na_2TP was obtained from waste PET bottles by treating them with concentrated sulphuric acid at 120 °C for 6 h.⁴⁰ The terephthalic acid obtained was purified by precipitation in water prior to being treated with a solution of Na_2CO_3 in ethanol for 12 h to finally obtain Na_2TP . XRD analysis was performed to confirm the purity of the product and investigate the structure, while the electrochemical experiments on the electrode made of Na_2TP revealed promising results. More precisely, the electrode made of 55% of Na_2TP was able to provide a discharge capacity of 190 mA h g^{-1} , while the composite synthesised with single walled carbon nanotubes achieved a specific discharge capacity of 241 mA h g^{-1} at 0.1 C and showed good stability up to 50 cycles.

Recently, some of us proposed a novel chemical upcycling approach to produce redox-active nanoparticles from PET waste for energy storage applications.⁴³ First, the depolymerisation of PET was performed using a recyclable organocatalyst, resulting in bis(2-hydroxyethyl) terephthalate (BHET) in high yields and purity. (Fig. 3a) Then, the BHET was converted into a methacrylate-based terephthalate monomer using methacrylic anhydride. The resulting monomer was then used to synthesise well-defined terephthalate-based nanoparticles *via* an emulsion polymerisation method. Cyclic voltammetry results using a 0.1 M TBAPF₆ (Tetrabutylammonium hexafluorophosphate) electrolyte solution in acetonitrile showed reversible oxidation and reduction of the terephthalate-based nanoparticles at a potential of -1.62 V and -2.26 V vs. Fc/Fc⁺, confirming their potential as an ultra-low potential anode material for application in all-organic batteries. Unfortunately, the authors also reported that significant irreversibility between the reduction and the oxidation processes was observed when employing a lithium-based electrolyte, limiting their potential use in LIBs. The galvanostatic cycling of the terephthalate anode electrode in 0.1 M TBAPF₆ electrolyte revealed good cycling stability and performance at an elevated C-rate (*i.e.* $\geq 5\text{C}$), reaching a stable specific discharge capacity of 32.8 mA h.g^{-1} at a C-rate of 30 C, of which 94% remained after 100 cycles.

In contrast to previous works where the materials were used as anodes, Saito *et al.* have reported a novel methodology mediated by organocatalyst to transform Bisphenol A-based polycarbonate (BPA-PC) into aliphatic polycarbonates for their application as electrolytes in solid state batteries.⁴¹ Solid state batteries contain both solid electrodes and a polymeric solid electrolyte avoiding the safety disadvantages of flammable toxic liquid electrolytes. (Fig. 3b) The depolymerisation reaction of BPA-PC was conducted for 2 h at 160 °C under a nitrogen atmosphere and catalysed by an acid/base mixture based on triazabicyclodecene (TBD) and methane sulfonic acid (MSA). The resulting diol-terminated carbonates were employed as monomers for the polycondensation of aliphatic polycarbonates. The ionic conductivity of the copolymers with

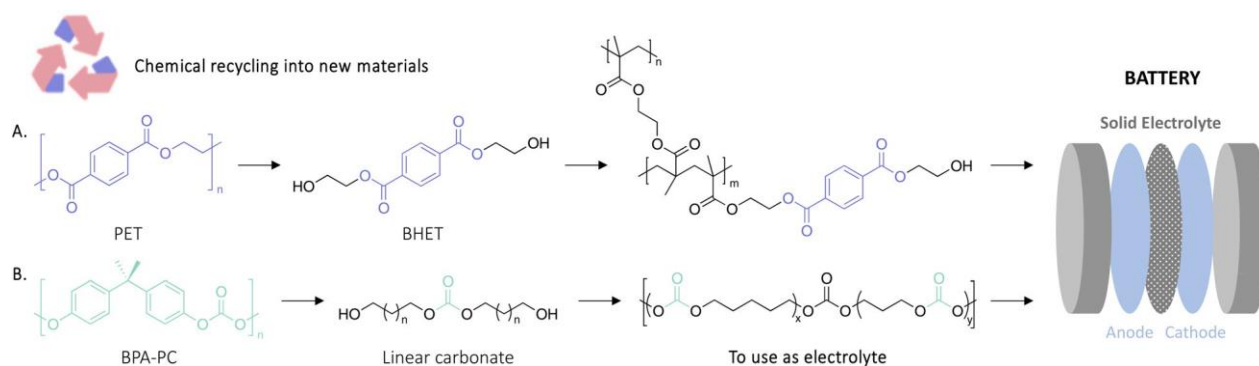


Fig. 3 Depolymerisation of (A.) PET into BHET for nanoparticles synthesis; (B.) BPA-PC into carbonates and their subsequent polycondensation into electrolytes.

different aliphatic chain lengths in a random fashion offered superior properties compared to their homopolymer counterparts, while the lithium transference number was 0.45. Although these electrochemical results are not better than the current state-of-the-art polymer electrolytes, it demonstrates the possibility to retain interesting chemistries precisely and selectively from plastic waste through chemical recycling methodologies.

Another reported example by Chen *et al.* consists on a novel procedure for the degradation of aramid fibres into nanofibres.⁴² These products possess adequate properties for application on the battery field as separators, electrodes or membranes. Aramid-based polymers have high mechanical, thermal and chemical resistance which render them materials of choice for the production of bulletproof artefacts. This work describes a simple procedure in which after reacting the disposal material with 2 wt% of aqueous KOH/DMSO mixture aramid nanofibres can be obtained at room temperature in less than half an hour. The changes observed on the aramid morphology were characterised by *in situ* monitoring techniques and the so-obtained products were employed on the

formation of aerogels which have shown high thermal decomposition temperatures (<500 °C), good mechanical properties and low thermal conductivities.

Importance of catalysis in the preparation of chemicals and materials from plastic waste

The use of catalyst enables a chemical reaction to proceed at a usually faster rate or under milder conditions than the uncatalysed process.⁴⁴ In several of the chemical recycling works the catalyst plays a key role not only on reducing the time and temperature of the reaction but also on the obtained material properties (Table 1). The use of very high temperatures for the recycling of carbon-rich polymers means that a high amount of energy is required for obtaining valuable materials. The ratio energy input over quality of the resulting material needs to be considered for the different technologies with the objective of turning plastic wastes into carbonaceous materials for energy storage applications. For example, Gong *et al.* demonstrated that the upcycling reaction of PP waste carried out without catalyst provides an amorphous carbon structure while using 5 wt% of OMMT to mediate the reaction leads to

Table 1 Summary of the upcycling technologies leading to materials for batteries encountered in the open literature

Ref	Feedstock	Technique	Temp. (°C)	Catalyst	Type of material/molecule	Yield (wt%)
14	PP	Pyrolysis	700	OMMT/Ni ₂ O ₃	Carbon nanotubes	60.1
16	PP	Pyrolysis	800	Ni/Ca-Al Ni/Zn-Al	Carbon nanotubes	10.6 2.4
19	PP PPMA	Pyrolysis	700	OMMT/Ni ₂ O ₃ NH ₄ -MMT/Ni ₂ O ₃ H-ZSM-5/Ni ₂ O ₃	Carbon nanotubes	44.8 56 42
20	PP PPMA	Pyrolysis	600	OMC/Ni-cat supported on silica-alumina	Carbon nanotubes	41.1
21	PP	Pyrolysis	900	OMMT/nickel formate	Carbon nanotubes	50
22	PP	Pyrolysis	900	OMMT/cobalt acetate/ Ferrocene	Amorphous carbon	18
23	PE, PS or PET	Pyrolysis	700	—	Carbon microspheres	40
25	PE	Pyrolysis	700	—	Carbon microspheres	50
26	PE	Pyrolysis	700	Cobalt acetate Magnesium carbonate pentahydrate	Carbon nanotubes Porous carbon	50 —
27	LDPE	Pyrolysis	—	—	Porous carbon spheres	45
29	PET	Pyrolysis	700	MgO/Co(acac) ₃	Porous carbon nanosheets	36.4
30	PET	Pyrolysis	600–800	—	3D nitrogen doped graphene	—
31	PE, PS, PE or PVC	Pyrolysis	700	Magnesium oxide/iron acetylacetonate	3D structured hollow carbon sphere/ porous carbon flake (HCS/PCF)	≥70
32	PS	Pyrolysis	700	Magnesium oxide	Porous carbon flake (PCF)	25
33	PS	Pyrolysis	600, 700	—	Carbon microspheres and carbon film	—
34	PVC	Pyrolysis	600, 700, 800	—	Hard carbon	—
35	PVC	Pyrolysis	600	—	Carbonaceous porous residue	—
36	Sulphonated Polyethylene (S-PE)	Pyrolysis	900	—	Porous sulphonated carbon	—
37	Sulphonated Polyethylene (S-PE)	Pyrolysis	900	—	Amorphous carbon	63
38	PE	Pyrolysis	700	Magnesium carbonate	Graphene mesoporous carbon	—
39	PET	Depolymerisation	25 Microwave	—	Disodium terephthalate	—
40	PET	Depolymerisation	120	Sulphuric acid	Terephthalic acid	72
41	BPA-PC	Depolymerisation	160	TBD:MSA	Linear carbonates	67 to 92
42	Aramid fibres	Depolymerisation	25	—	Aramid nanofibres	—
43	PET	Depolymerisation	180	TBD:MSA	BHET	92

filamentous carbon as well as optimal reaction rates.¹⁴ Similarly, Wu *et al.* reported the influence of the nature of the catalyst on the production of carbon nanotubes and syngas from different plastic waste feedstocks. Employing an optimised amount of Ni/Ca–Al complex as catalyst allowed the formation of carbon nanotubes as filaments on the surface of the catalyst while mediating the reaction with Ni/Zn–Al shifted the reaction towards the highest production of hydrogen, 20% more than with Ni/Ca–Al.¹⁶

Despite of the predominance of metal based catalysts there is a trend to substitute them by more benign organocatalysts. Although most organocatalysts have relatively low thermal stability, some acid/base mixtures seem to be capable of withstanding the high temperatures required for plastic degradation reactions. TBD:MSA in particular has been reported in many articles due to its excellent chemical activity and selectivity. Saito *et al.* employed this catalyst mixture to decompose polycarbonates into linear diol carbonates in high yield. These diols were subsequently polycondensated to produce polymer electrolytes for batteries.⁴¹

In another example of the use of organocatalysis, Goujon *et al.* depolymerised PET into BHET using TBD:MSA due to its high selectivity avoiding the formation of dimers and trimers and excellent reaction rates. The recovered BHET was then transformed into an intermediate for the synthesis of redox nanoparticles that show promising performance as anode materials for battery applications.⁴³

Conclusions and Outlook

The examples described in this perspective show how plastic waste could potentially provide sustainable feedstock materials for battery technologies. The proof-of-concept studies reported to date highlight the potential of producing battery components in a sustainable manner, using plastic waste as feedstock, without sacrificing performances. However, the upcycling of plastic waste towards the production of various battery components (*i.e.* so far only electrode materials or electrolytes) is still at a very early stage and requires further research and optimisation of the processes to become a reality.

On the one hand research should be directed towards the development of advanced catalysts for the depolymerisation of plastic waste to enable the recovery of specific synthons or materials with various complexity in terms of chemistry and structures to target end-application requirements in a cost-effective manner.

On the other hand, as reported by Fuentes *et al.* it is also important to find recycling solutions for the inorganic components used in energy storage devices.⁴⁵ In this regard, they investigated the use of zinc and cobalt oxides, obtained from the recycling of spent alkaline and LIBs batteries, as catalysts for the chemical recycling of PET waste. This highlights again how important cross field research is to reach sustainability targets, as an issue faced by one industry sector could be the solution of a problem faced by another.

Finally, analysis of the environmental impact (*e.g.* life cycle assessment, techno-economic analysis, *etc.*...) of using these new feedstocks (*i.e.* plastic waste or other renewable resources) is required to properly assess their relevance as a replacement of mainstream materials in terms of sustainability. It is clear that the environmental impact of any of the discussed upcycling methods should be minimised and evaluated by means of LCA or similar method. We have recently calculated the carbon footprint for different upcycling approaches and the results demonstrated that no matter the upcycling method employed it can be environmentally beneficial in all cases. Besides, other considerations must be taken into account such as the possible pollution of the process or the quality of the final products in early stages and should be critically evaluated for a proper valorisation of plastic waste.⁶

The energy sector faces numerous challenges to reach a circular economy for energy storage devices. At the same time, these challenges bring new opportunities, and exciting discoveries would certainly raise on the transformation of plastic wastes into new materials for energy devices which will be facilitated by performant and unique catalysis.

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