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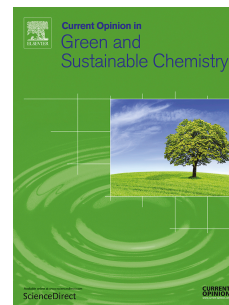
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Technology advancing Polymers and Polymer Composites towards sustainability: A review

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Abstract

It is reported that plastic waste is accumulating in the oceans at an alarming rate. A significant proportion of this waste is plastic packaging materials, even though recycling routes and options are available to retain and reuse this oil-based resource. In addition to thermoplastic materials, fibre reinforced polymer materials are finding increased use and soon will become a concern with respect to disposal. This review presents current research and technology that aims to embrace plastics and polymer composites into a circular economy. The benefits and challenges associated with polymer recycling are highlighted, not least being the strategies required to encourage industry and society to recycle resources rather than dispose and renew.

Keywords: polymer, composites, sustainability, recycling, life-cycle, circular economy.

Introduction

Polymers and polymer composites are low density, durable materials that can be processed into simple and complex shapes. Therefore it is hardly surprising that the demand for such materials increases every year, with an expectation that annual demand will double in the next 20 years to in excess of 600 million tonnes [1]. Plastic is a popular material for packaging, from the notorious disposable plastic carrier bag to the plastic milk bottle. Such single use plastics are a cause for concern as they become a vital lost resource if they end up in landfill. 'The new plastics economy' report [1] aims to encourage industry and society to move towards a 'circular economy' model for plastics, identifying barriers to global materials flows and enablers such as digital technologies.

The European commission has proposed a 'Plastics Strategy' to try and mitigate against such loss of a finite resource [2] – and the vision is that by 2030 all plastic packaging on the EU market will be recyclable or reusable. Therefore technology to

economically recover these polymeric materials and return them into the materials supply chain needs to be realised.

Similarly, the demand for energy, another limited resource, has been instrumental in the increasing popularity of fibre reinforced polymer composites. Composites enable the light-weighting of vehicles (cars, trains, boats and aeroplanes) which results in increased fuel efficiency. In addition, the wind turbine requires lightweight turbine blades and the natural choice is fibre reinforced composites. Although composites are durable, there is waste arising from the manufacturing process, an immediate concern, and as end-of-life approaches there will be future concern for 'disposing' of large composite structures [3,4]. The Composites UK report [3] identifies the options for recycling composite materials and presents the environmental impact of the different recycling processes.

Sustainability and the environment

The 'circular economy' is regarded by some as the solution to sustainability and protection of the environment. Plastic recycling is one stage in moving towards a circular economy. However McDonough [5] proposes that striving towards a 'carbon neutral' state is not sufficient and society should work towards a carbon positive situation (i.e. carbon is restored back into the earth rather than allowed to escape to the atmosphere as CO₂ or to the rivers and seas as (plastic) waste). Utilising CO₂ to produce polymers is a carbon positive action and if these polymers can remain in use, re-used or recycled, then this will be beneficial. Unfortunately simple recycling of thermoplastics can bring about changes in material properties (or quality issues). A short communication [6] focusses on plastic packaging and identifies how designing for 'improved quality' can limit recyclability and prevent these plastics entering the circular economy. It also presents opportunities and challenges provided by initiatives aimed at improving the position of plastics in the circular economy.

These quality implications are that it is easier to produce lower value products with recycled polymers, accept degradation and move towards products which are no longer recyclable (due to economic or material factors) than to design towards perpetual recycling. To combat this move towards limited recycling new strategies are required [1]. One such strategy is a move to chemical recycling. The benefits of

chemical recycling over conventional recycling and the areas for process improvements/plastics' chemistry design to aid recycling on a commercial scale are presented by Rahimi and Garcia [7].

Routes for recycling of thermoplastic waste

Polymer recycling methods are categorised as:

- Primary – mechanical reprocessing plastic to produce the same product as the original plastic (e.g. closed-loop recycling of HDPE milk bottles [8,9])
- Secondary – mechanical reprocessing to lower value products (downgrading) [7,9]
- Tertiary – chemical recycling to recover petrochemical components in plastics [9,10,11]
- Energy recovery (incineration) – of limited value because energy recovered as heat is a fraction of energy conserved by recycling [12]

Several recent reviews have focussed on thermoplastic polymer recycling covering the aforementioned 4 main methods [7,8,9,13] and discussing the opportunities, benefits and disadvantages of each. Specifically, El Mehdi Mekhzoum et al [13] present technological and economic issues preventing increased volumes of thermoplastic waste being recycled in addition to the varied methods and options that can, and are, used to recover plastic resources.

Thermoplastics lend themselves to relatively simple recycling technology because application of heat and pressure can reform thermoplastics into alternative structures. However, prior to 'reforming' to re-use or recycle, the thermoplastic waste needs to be separated from other waste, isolated into a single polymer type (e.g polypropylene, polystyrene, polycarbonate etc.) and free from contamination (food waste, adhesive labels and printing inks etc.). Such processes are termed 'mechanical' because the waste is sorted, shredded, washed, extruded and granulated mainly by mechanical methods.

Primary recycling is the preferred route as it retains value and should maximise energy savings (when compared with use of virgin polymer source). However, current technologies can only apply primary recycling to a very small amount of annual arisings of plastic waste. The reasons for this are the primary route needs;

- clean (uncontaminated) waste of a single polymer type – much waste is contaminated with food waste, multiple plastic wastes, other waste streams (fibre, mineral, metal etc.) [14]
- many packaging materials are a mixed plastic, composite formulation, maximising required properties [6,15], and contain additives which might adversely affect subsequent product manufactured with additive containing plastics or escape to the environment during recycling [16]
- processing route can cause polymer degradation [6,7,9], and therefore use of recycled plastic into products for many applications (closed loop) is generally less than 50% [17,18]

Secondary recycling processes are valuable in that they reduce the total energy required for processing (new material requires energy input to manufacture that is significantly greater than the material preparation by the recycling process) and is preferable to the simple energy recovery route [19].

Tertiary recycling has the benefit of being most flexible regarding how the 'recovered material' is subsequently used – essentially breaking down the waste plastics to their building blocks and then producing new polymers [8,20]. However it is not the most efficient recycling route in terms of energy use as energy is used to break down the polymers and then rebuild. Thus, it could be argued that use of virgin material is preferable. To ignore the tertiary route would be to lose a processing route that furthers the sustainability cause. Therefore, reasoning based on resource management and sustainability recognises this route as a valid option to be considered [5].

An alternative to chemical treatment of plastic waste is biological treatment [20] – use of enzymes [21] and organisms [22] that can digest plastic, trapping the carbon and returning it to the earth as an available resource [5].

And perhaps the Byfusion Process is the most environmentally-friendly method for re-using plastic waste [23]. Peter Lewis devised a process, using only pressure and steam, to convert mixed plastic waste into 'building blocks' for construction.

The numerous and diverse methods that can be applied to (thermoplastic) polymer waste are not sufficient to ensure plastics do not end up in landfill and oceans. The main drivers can be classified as economic, environmental and social [24]. In order to ensure recycling is maximised each of the drivers need to be addressed – providing incentives and education on a global scale [1,2].

Recycling of fibre reinforced polymer composites

The challenges associated with recycling and re-use of thermoplastics have been outlined in the earlier paragraphs. However fibre reinforced plastic (FRP) composites add another dimension [3]. The majority of FRPs comprise a thermoset polymer matrix and glass or carbon fibre reinforcement. Reprocessing of thermosetting resins requires chemical 'digestion' to the monomer and then polymerising to produce a 'new thermoset' product. Currently such a process is not economical. Therefore the main strategies for recycling FRPs are [3,25,26,27]:

- Incineration – recovering some embodied energy in the form of heat, with residues going to land-fill [28,29]
- Thermal and chemical recycling (solvolysis, pyrolysis) and recovery of reinforcing fibres [29,30,31]
- Mechanical recycling – size reduction to produce a fibrous or powder product for re-use [29,32]

Mativenga et al [26] discuss the drivers, sustainers, barriers and volumes of composite waste applicable to recycling of composite waste in the UK and South Africa – concluding that a common driver and sustainer for such recycling strategies are the opportunities to reduce manufacturing costs.

The incineration process is the least favoured route as it has little value over landfill. However a variant, co-incineration of glass fibre composites in cement kilns offers combined material and energy recovery [3,25].

Thermal and chemical recycling appear to be the preferred route, attracting much interest. The variations on thermal recycling/pyrolysis [3] include chain conveyor pyrolysis, fluidised bed pyrolysis [33] and microwave assisted pyrolysis [34]. The pyrolysis process allows recovery of the valuable carbon fibre reinforcement and the polymer matrix is reduced to a gas or liquid fraction that can be collected and re-used [34] (as a fuel or precursor to synthesis of new hydrocarbon materials e.g. synthesis of new polymeric materials). Current chemical recycling methods focus on solvolysis and a comprehensive review [29], discussing the merits of a variety of recycling techniques for fibre reinforced polymer composites, demonstrates why solvolysis could be the future preferred route.

Solvolysis is similar to pyrolysis, the end-product for re-use is the carbon fibre, with the resin being degraded and 'washed' away from the fibres. Researchers have successfully separated carbon fibres from cured carbon fibre reinforced plastic waste and reused the fibres to build a 'new' CFRP structure [35,36].

The ideal solution would recycle all components of polymer composite waste. However this would require significant progress in the chemistry of the thermoset resin; use of a chemical structure that could be reverse polymerised [37,38]. In 2014 poly-hexahydrotriazines were thought to be the way forward to fully recyclable thermosetting plastics [38]. However there is little mention of these 'wonder-materials' since 2014. The current 'recyclable thermoset' [37] is promoted as an epoxy-based thermoset that can be used to produce advanced carbon fibre reinforced composites. At the end-of-life the composite matrix can be uncrosslinked producing a thermoplastic and releasing the valuable carbon fibre. Then the thermoplastic can be re-used for other, less demanding applications. This technology probably still has significant development requirements before being accepted into the market.

The alternative to the aforementioned 'recycling routes' for fibre reinforced polymer composites is simple re-use of composite panels as construction materials. A characteristic of polymer composites is their durability – and with the expectation that increased use of these composites will result in increased waste volumes due to necessary 'disposal' at end-of-life, solutions for such disposal need to be sought. Original large wind turbine blades (>30m length) are nearing end of life and novel

design concepts have been reported to utilise these large composite structures to construct low-cost housing [39,40].

The main driver for recycling 'waste' is economics – based on costs (profitability), resource availability (sustainability), energy considerations (environmental). In contrast to thermoplastic waste which has economic value [24], thermosets and thermoset composites have no immediate economic value [41]. This is a short term view, as if all waste is disposed of rather than recycled then resources are 'lost'. As resources dwindle, recycling thermoset composites could become profitable. However, even if the pendulum does not swing towards profitable recycling, disposal and production of 'fugitive carbon' [5] is not a socially responsible option. Therefore legislation, forcing industry to subsidise recycling or 'producer responsibility for recycling', is being proposed as the driver to encourage recycling and sustainability practices [2,41].

The Future for Plastics Recycling

Currently the technology exists to recycle plastics (thermoplastic waste and thermoset composite waste) to some degree, and recover resources from the waste (materials and/or energy) [1,3,42]. However, recycling rates for these materials remain low [26,43,44] and, in the short term, legislation appears to be the solution to encouraging industries to increase recycling rates [1,41].

Research investigating the benefits from recycling with respect to environmental considerations (energy costs and savings) can identify the full value of recycling [45,46,47]. For composite waste, several studies have been published focussing on mechanical recycling [48,49] and a comparison of mechanical recycling and chemical recycling [50]. Such studies can identify the hidden benefits to recycling, but more importantly should be able to determine inefficiencies in processes and technologies and focus on technological advancements to improve these. Gu et al [51] investigated life cycle environmental issues associated with mechanical recycling of plastic waste in China. The findings were that recycled raw materials generally proved to be environmentally preferable to virgin materials, with manufacturing virgin composite products being cited as having four times the environmental 'impact' compared with recycled composite production.

Conclusions

The global volumes of plastic waste recycled could be significantly increased just by applying the recycling technologies already available. However there are insufficient incentives (financial gain or market advantages) to encourage industry to increase recycling activities. Although legislation could change the recycling landscape another limiting factor impacting recycling is the design and use of multi-component plastic structures. Primary recycling is difficult with multi-layer bottles/packaging, and downgrading is inevitable, preventing multiple recycling of such products. Therefore to maximise re-use of plastics either manufacturers need to confine their products to single component plastics (maximising recyclability), or industry needs to develop methods to separate the components from the multi-layer structures. Current composites recycling technologies could provide the required strategies – as one route for composite recycling requires the plastic component to be ‘solubilised’. Of course the missing element is the separation of different plastic ‘monomers’ and then the re-polymerisation.

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** of outstanding interest

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