



Recycling of Polymers

by John Hind

Introduction

Plastics have become an indispensable part of modern life. Cars, computers, baby-bottles, telephones, clothing, packaging; no aspect of our society is untouched by plastics. Their versatility in terms of physical properties and the unique formability, which makes the term "plastic" so appropriate, have made them ubiquitous.

These intrinsic physical properties also make them eminently suited to recycling. Plastics have indeed been recycled in various ways ever since their introduction over a hundred years ago. Natural horn is a thermoplastic material and offcuts were fused together by heat and compression [1]. The driving force for recycling was purely economic: plastics were too rare and valuable to be wasted.

Even when synthetic plastics became widely available in the 1950s and 1960s they were carefully husbanded. In 1954 the Plastic Bag Co. gave instructions for the washing and drying of polyethylene bags, which cost about a shilling each [2].

With increasing application and decreasing prices, plastics became the symbol of disposability in consumerism and hence an inevitable object of concern in the relatively recent heightened public awareness of environmental issues.

What is recycling?

Recycling may be defined as any activity involving reclamation, recovery or reuse of materials or articles. In other words, any method of extracting value in the form of energy or material from waste generated at any time in the cradle-to-grave life cycle of a product may be included in the definition. Recycling may be mechanical, chemical or thermal. The French word valorisation is now being used more widely and embraces energy from waste (often abbreviated to EfW) as well as other forms of recycling.

The "cycle" part of the word is important and to complete a cycle four phases must be gone through: collection, sorting, reclamation and marketing.

In the course of its life, the value of an item changes as it undergoes depreciation with time until, quite suddenly, it becomes a liability; it has negative value. Whatever is now done with the item will entail a cost. Recycling is only sensible if the cost of processing the scrap at the end of its life is less than the value of the recyclate produced. Having been promoted to a positive value, the life cycle may be repeated in principle any number of times. [3]

Economically, the best option is often landfill and indeed it may be argued that landfill disposal of stable polymers can be part of responsible polymer waste management and that a certain polymer content in a landfill site contributes to its stability. [4]

How can recycling of polymeric materials be achieved?

The methods by which value may be regained from waste or used plastics may be divided into four categories according to two imperfectly coincident hierarchies:

- reuse or: primary
- mechanical recycling secondary
- chemical recycling tertiary
- thermal recovery quaternary

At each level the original structure, first of the article then of the polymer molecule itself, is further dismantled.

It may be argued that reuse is not really a form of recycling as many containers for instance are intended for repeated use. However, it would seem sensible to include beer bottles but not, for example, suitcases in this category.

Mechanical recycling may be primary or secondary, while the third and fourth categories have an exact correspondence.

Primary recycling involves process scrap, or a single clean waste type. An important subclass of primary recycling is known as "Closed loop recycling" in which polymer from a single product or product type is collected and recycled in-to the same product. Polypropylene from battery cases is a well-established example. [5]

Secondary recycling uses reclaimed post-consumer materials as a source of material for new products. Tertiary recycling involves chemical breakdown of materials which are then re-utilised further back in the feedstock chain. Quaternary recycling is often termed energy from waste.

There are many initiatives to be found, but for all types of recycling the question of whether it is economically worthwhile must be addressed. Either the arisings must be very abundant or they must be very valuable. The high-volume plastics are polypropylene (PP), polyethylene (PE), polystyrene (PS), polyvinyl chloride (PVC), polyethylene terephthalate (PET) and acrylonitrile butadiene styrene (ABS) (see Appendix and Section 6) and naturally most work has been done on these polymers although high value polymers such as polyether ether ketone (PEEK) have also received attention. [6]

3.1. Reuse

The simplest form of recycling is reuse: in Denmark, for instance plastic drinks bottles are reused (as are glass bottles in the United Kingdom). [7] While this works well for simple, well defined, easily recognised items like branded bottles, the costs of collection and cleaning must be taken into account: clearly, if more energy is used in recycling a bottle than in making a new one then nothing is gained.

3.2. Mechanical recycling

On the next rank in the hierarchy is mechanical recycling whereby either process scrap (start-up, shut-down, rejects, trimmings, etc.) or post-consumer material is regranulated so that it can be melted and formed again. The post-consumer waste may be primary or secondary.

One sub-class of primary mechanical recycling is widespread and largely unrecorded. It is very common for sprues and runners and defective mouldings to be regranulated next to an injection moulding machine and put back into the feed material. Likewise, the edge trimmings from extruders and blow moulders are reprocessed virtually at source. This practice has a history as long as moulding itself and will be discussed in the section dealing with the survey (see Section 6).

New articles may be produced directly from the waste. The survey shows that many factories re-use their process scrap and a number have it reprocessed outside for their own consumption or sell it to be used ultimately by others. A few even buy regranulated material from outside suppliers.

It is post-consumer recycling that has received most attention in the literature, especially in the more general press. In stark contrast to primary recycling, where the material is of known provenance, in secondary recycling of plastics, mixed and dirty polymer arisings which are part of the municipal solid waste (MSW) stream must be dealt with.

3.2.1. Examples of mechanical recycling

A fundamental consideration for any recycling process is the investment of effort and energy. Therefore, much of the work in the field has been aimed at handling mixtures and trying to overcome the problems of incompatibility. At the same time processes with low energy input are favoured.

A process developed by Smile Plastics of Shrewsbury takes clean, granulated post-consumer high-density polyethylene (PE-HD) bottle scrap and compresses it into multicoloured board.

That the forming is done without remelting the polymer is not only energetically favourable, it is essential to the process since heating and mixing would destroy the attractive and unique patterns which are produced in each board as the different coloured granules are fused into sheet.

Physical properties are much enhanced by additives which prevent delamination and the finished boards can be sold at a premium over virgin, plain PE-HD sheet. [8]

Solid state shear extrusion invests much more energy in the polymer and attempts to homogenise and compatibilise mixtures of materials including not only thermoplastics but also thermosets, rubbers and some natural products like wood.[9] The process is limited in terms of the colours which can be produced.

ABS is widely used in the automotive and electrical industries for switch housings etc. The mechanical properties of recycled ABS contaminated with ink and other polymers added at various levels to virgin polymer have been studied. [10,11]

Nylon 6,6 (PA6,6) from automotive radiator end caps can be recycled after the glycol antifreeze has been extracted. This represents a low tonnage, high value, easily separated

source. Work has been carried out on extraction, cleaning, reuse and mechanical properties.[12]

Increasing use of polycarbonate (PC) for optical discs leads naturally to identifiable arisings with the potential for recycling. The difficulty lies in the separation of the polycarbonate from the coatings used to record and preserve the stored information. While chemical separation and melt filtration are possible, mechanical recycling has energetic advantages.[13] Work on the rheology of recycled grades of polycarbonate.[14] is also of significance to the moulder as processing is very dependent on flow properties.

Polyether ether ketone is an engineering plastic used in such low tonnages that it does not appear in the top 20 polymers used in the UK according to BPF figures.[15] It is also very expensive, being twenty times the price of general-purpose ABS,[16] (grades containing carbon fibre are even more expensive) making recycling economically attractive even in small quantities. "Prepregs" are resin impregnated glass or carbon fibre reinforced sheets in a semi-finished condition. As they are shaped, offcuts arise which, rather than being discarded, can be re-extruded to produce a grade of carbon fibre reinforced PEEK suitable for injection moulding. Good results are reported in terms of processing and mechanical properties. In one of the few pieces of work to include a cost analysis it is concluded that "in principle this method could be commercially viable" with the implication that it is not so in practice.[17]

Plastic bottles are made almost exclusively from one of only three polymers: PE-HD, PET or PVC (with smaller volumes of PP); they are made in huge numbers - in the order of 2 thousand million per annum; they are very visible and present a litter problem in many countries. For all these reasons much attention has been directed toward schemes to recycle drinks bottles. Such initiatives must all address the problems of identification, sorting and cleaning before the material can be reclaimed and marketed.

Much early work was done on compatibilisation of PET/PE blends because the clear PET bottles were supported on pigmented PE base caps as the process could not at the time produce flat bottomed bottles. [18-21] The advent of "petaloid" based bottles obviated the need for a second moulding in a different polymer, and thus raised the PET content to nearly 100%. However, closures, labels and contamination with PE, PP, PS and PVC may still need to be allowed for.

A major problem with polyethylene terephthalate, as with other polyesters, polycarbonate and polyamides, is that of hydrolytic chain scission. In other words, when the reclaimed polymer is heated, melted and re-extruded any moisture in the system will break the polymer chains which reduces molecular weight and viscosity while increasing the degree of crystallinity and lowering impact strength. Scission is also promoted by the thermal degradation products of PVC, which is inevitably present in post-consumer PET.[22] These effects can be countered to an extent by thorough drying before reprocessing [23] or else chain extenders can be introduced during extrusion to increase molecular weight.[24]

Two techniques which have been available for decades but which have more recently been utilised for recycling are co-extrusion and co-injection. The theory is simple: virgin material is moulded on the outside of the article and recycled material is hidden on the inside. Thus discoloration, a certain amount of contamination by inks, labels and even metal particles can be tolerated in the core provided the final mechanical properties of the composite are acceptable. Polypropylene in mixed colours, PP carpet scrap, and PS/aluminium laminated foil (not separated) and plastics derived from MSW have all been

successfully co-injected [25] Window frame profiles have also been extruded on the same principle. [26]

3.2.2. Solvent extraction

An interesting physical, although not mechanical, reclamation method is solvent extraction which has been demonstrated for polyethylenes and polyacrylates. [27] The polymer arisings are dissolved in a solvent, filtered and reprecipitated by a non-solvent. The advantage is the much gentler treatment of the polymer.

3.2.3. Barriers to mechanical recycling

There are several technical barriers to be overcome in mechanical recycling. It is in the nature of waste products that they are dirty and mixed. It takes time, energy and money to undo this degradation.

Once plastics have entered the municipal solid waste stream their almost inevitable fate is landfill although there are some incinerators which can recover energy from plastics in waste. [28]

3.2.4. Identification and separation

Preventing the mixing of plastics in the waste stream makes identification and separation much easier, but at the expense of a more complicated collection system.

An alternative is the so-called "bring" scheme which operates in the same way as the familiar bottle banks. Cross contamination by other types of waste and by misuse or abuse of the collection points must also be coped with. A successful variation of this is "reverse vending": the "Save a Cup" company provides bins which accept and stack polystyrene drinks cups as well as providing a container for the dregs. In this way a compact, single polymer is collected which is only lightly contaminated by drinks residues. The reclaimed polystyrene is not used for food applications.[29] It is perhaps disappointing that the collection of the used cups is not done at the same time as servicing the vending machine using the same vehicle and that the reclaimed polymer is not used again for cups, thus closing the loop completely.

Polymer arisings, however collected, must be identified. In the case of bottles for instance, PET and PVC bottles are clear while PE and PP are opaque. More exact sorting than this requires either great skill on the part of a human worker or else some kind of technology for automatic separation. Since manual sorting is slow and rather unreliable a number of techniques for identification and separation of mixed polymer arisings have been developed and are now reaching maturity.

Tecoplast Govni of Italy filed a patent for the first system for separating and reprocessing PET and PVC in 1988. The problem of collection was addressed by using vending machines which accept empty bottles and warm and flatten them to reduce volume for transportation. The two polymers were differentiated by X-ray. [30]

The automotive industry has expressed interest in two devices one based on a laboratory type spectrophotometer and the other a very simple hand-held device called the "Tribo pen" which works on the triboelectric principle. [31] (Triboelectricity is generated when surfaces are rubbed together.)

Ultrasonics [32], density differences [33], differential solution [34], infrared, near infrared and Raman spectroscopy have all been reported. [35-38] An interesting refinement to infrared spectrometry using a Fourier transform technique (a type of mathematical processing) widens the range of plastics which can be identified and copes even with black pigmented polymers, which usually present problems. [39]

A different approach was suggested by Soler [40] who suggested compounds with strong spectroscopic signatures be added as tracers to the polymer during manufacture.

3.2.5. Handling mixtures

Rather than invest effort and energy in identification and separation of polymers, some workers have directed their efforts to finding ways of using mixtures as they arise. Two barriers have to be overcome: the natural incompatibility of various thermoplastics for each other and their wide range of melting points. PVC, for instance, degrades with increasing rapidity above 200°C and would be dangerously unstable at the PET processing temperature of around 280 °C.

Assuming that PVC can be avoided, the remaining polymers are incompatible to the extent that components made from the resulting mixture have such poor physical properties that only the least demanding of products can be made. This of course means the cheapest which makes it difficult to meet the cost of recycling. Work was done by La Mantia [41] in which the reclaimed, partially degraded mixture was blended with low density polyethylene to give secondary materials of acceptable mechanical properties apart from elongation at break. In subsequent work elastomers were added to improve this property too. [42] Thermoplastic elastomers were found by others to restore mechanical properties in recycled polyethylenes contaminated by other polymers by improving compatibility between them and the principal polymer. [43, 44]

The problem of incompatibility has been addressed by several groups using such techniques as dialkyl peroxide [45] and maleic anhydride [46] modification during extrusion.

3.2.6. Cleaning

Having by some means obtained an identified, reclaimed polymer it must be reprocessed into a new feedstock. This is usually done by granulation followed by washing and drying. Water content is critical for any polymer susceptible to hydrolysis. From this point on processing techniques are the same as for new materials: melting, extrusion, melt filtration and pelletising.

It is avoidance of the identification and mixture problems which makes primary recycling more attractive than secondary and which makes closed loop recycling more effective.

It is these same problems which make chemical rather than mechanical recycling attractive for post-consumer waste.

3.3. Chemical recycling

On the next level in the hierarchy is tertiary recycling, which involves the use of polymer waste arisings as a feedstock for any of the pyrolytic, chemolytic or hydrocracking processes used to break down polymeric waste into simpler substances [47] subsequently re-polymerised to produce virgin materials. [48-51] Most of this work has been carried out in Germany.

By using plastic waste arisings as a feedstock for some process which breaks down the macromolecule into smaller parts, all problems of compatibility between different polymers, reduction in molecular weight by chain scission, identification of polymer types or grades within a type are circumvented. While it may be argued that destruction of the polymer structure to such a degree is energetically unfavourable compared with primary recycling, it must be remembered that a great deal of energy can be invested in identification and sorting and that for any recycling scheme solutions must be evaluated on the whole life cycle. [52] The other advantage is that if thermal cracking or pyrolysis is used, plant already exists in the petrochemical industry to handle the oily and gaseous breakdown products [53] without changing process parameters [54] Polystyrene can be fed into heavy oil processors, [55] while thermosetting resins like epoxies and polyurethanes, which are usually difficult to handle, can also be recycled in the same way [56] precisely because the chain scission reactions mentioned above are in this instance exploited for polymers with appropriate chemistry. (PET, PC, polyurethane (PUR), PA, etc.)

While most plastics yield fairly non-specific pyrolysis products including methane, ethane, ethene, propene, benzene, toluene, xylene and naphthalene, it is reported that up to 97% of the monomer, i.e., methyl methacrylate and styrene respectively, can be recovered from polymethyl methacrylate (PMMA) and polystyrene. [57] Polyethylene can also be depolymerised by heat alone to yield ethene gas. [58]

In an attempt at milder treatment, glycolysis and hydrolysis of PET have been used to produce monomeric or oligomeric ethylene terephthalate, but once again the problems associated with sorting and contamination are encountered. [59, 60] These low molecular weight products can then be synthesised into PET or other polymers [61] which can be used conventionally or added to concrete. [62]

Polyurethanes may be similarly dismantled by glycolysis to yield liquid products which can be reused as components in a new polyurethane resin. [63]

Processing of heterogeneous plastics waste by pyrolysis in Germany and coliquefaction with coal in the United States of America (USA) are proposed as alternatives to combustion. [64, 65] Eastman, Hoechst and Goodyear in the USA all have glycolysis processes using post-consumer beverage bottles and an existing plant at Bottrop in Germany operated by Veba Öl can convert unsorted plastic waste into a diesel type oil. A number of other plants in the USA and Japan are also reported. [66]

The Bottrop plant has been capable since October 1993 of using 40 000 tonnes per year of unsorted plastic waste as a partial feed to a hydrocracking plant. Further development depends very much upon economics. [67]

Work has also been carried out on production of compost from biodegradation of plastics, although this is not seen as having large scale potential. [68]

A number of problems associated with high temperature treatment of polymers have been reported.

3.4. Thermal recovery

At the fourth level in the hierarchy, a method of valorising polymers in solid waste (also termed "quaternary recycling") exploits the calorific content. All polymers have a high heat content so that incineration with energy reclamation offers a way of winning back some value from polymer wastes where the level of contamination or degree of mixing makes any other process economically unattractive.

It has been estimated that in Australia each person generates a kilogram of garbage per day and thus the waste from a city of 4 million people could be used to generate 2.4 GWh of electricity at an output of 100 MW. [69] While not all of this is derived from plastics, it illustrates the potential of energy from waste.

Pyrolysis of polymers in fluidised beds yields heating oils with calorific values of around 45 MJ kg⁻¹ [70] This may be compared with values of 42 MJ kg⁻¹ for oil and around 30 MJ kg⁻¹ for coal; calculation from first principles gives values of around 50 MJ kg⁻¹ for hydrocarbons and 40 MJ kg⁻¹ for carbon itself. (The figure for PVC, with a 57% chlorine content by weight would be significantly lower). [71, 72] Mixed plastics have a calorific value of around 37MJ kg⁻¹ [73]

3.4.1. Problems with pyrolysis

The main problem associated with high temperature treatment of polymers arises from the so-called hetero-atoms (atoms other than hydrogen and carbon). Total combustion of polyolefins (or any other hydrocarbon) yields carbon dioxide and water and nothing else. Hetero-atoms have varying effects. Oxygen is no problem in an incinerator or from the environmental point of view, although it may be unwanted in a hydrocracker.

Polyurethanes and nylons contain nitrogen which produces oxides of nitrogen under combustion conditions, which must be removed from flue gases.

As it accounts for over half the mass of PVC and is also present in flame retardants in other commercially available plastics, most attention has been given to chlorine.

In a pyrolysis process it manifests itself as hydrogen chloride gas - corrosive and acidic but fairly easily removed in a scrubber. Indeed, one plant, once again in Germany, produces hydrochloric acid from this source on a commercial basis. [74] Many papers are concerned with the fate of chlorine under oxidative conditions particularly with reactions which produce dioxins, polychlorinated biphenyls (PCBs) and related compounds. The technology to deal with these compounds already exists in terms of pre-combustion treatment, incineration and flue gas treatment; the barriers are in management and economics. The levels of chlorinated dioxins in flue gases are reported to be unaffected by the presence of chlorinated plastics in the incinerated waste. [75-80]

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