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SPECIAL FEATURE: REVIEW Chemical Feedstock Recycling 10

Recycling plastics from automotive shredder residues: a review

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Abstract Automotive shredder residue (ASR) is an inevitable by-product of car recycling, i.e. removal of all liquids and hazardous or valuable components from the car and shredding of the hulk, followed by the recovery of steel, iron, and non-ferrous scrap. The European Union (EU) ELV Directive requires attaining higher recovery and recycling rates, resulting in a reduction of the amount of ASR going to landfill. The most plausible methods to achieve a considerable reduction of ASR are as follows: either recycling of separated materials and dismantled bulky parts, such as bumpers, dashboards, cushions, and front and rear windows, or else systematic sorting of the commingled and size-reduced materials, resulting from shredding. After a brief comparison of the actual situation in the EU, the USA, and Japan, the characteristics of actual ASR are reviewed, as well as some of the most prominent efforts made to separate and recycle specific fractions, such as polyolefins, ABS, or polyurethane. Attention is paid to some major players in the EU and to some of the pitfalls that besiege these ventures.

Keywords Automotive shredder residue · Car recycling · Treatment methods · Mechanical recycling · Emission control

Structure of this review

Since more than 50 years end-of-life vehicles (ELV) have first been scrutinised as source of spare parts and then shredded to recover their metal content. Today shredding is an energy-intensive, environmentally sensitive, mature industry, as explained under the heading "Scope and definitions".

About 75 wt% of ELVs is recovered as metal, leaving the automobile shredder residue (ASR) as a balance that is difficult to separate and process to useful materials or to valorise. With the growing number of cars in use such ASR has evolved into a major waste stream that attracted the concern of environmentalists and legislators. Notwithstanding obvious technical and economic complications, the EU ELV-Directive made mandatory materials and energy recycling a priority, as discussed for the EU, Japan, Korea, and the USA in "Legal status".

As a next step, the present techniques used in shredder operations are briefly presented, with emphasis on prior dismantling and ulterior recovery of metals. Thereby ASR arises as a fairly variable flow, for shredders typically process more other metal-containing goods than ELVs proper. Still, based on numerous sources, the generation and typical characteristics of ASR are sketched, respectively, in the parts on "ELV processing" and "Automobile shredder residue".

Automotive shredder residue contains almost all thermoplastics, rubbers, and thermosets that enter increasingly in the manufacturing of modern cars. Yet, it also contains not only unpredictable and useless extraneous matter, but also hazardous substances, such as mercury, lead, cadmium… or polychlorinated biphenyls (PCBs) and legacy brominated flame retardants (BFRs). Hence, ASR is classified as hazardous, yet the bulk of ASR has always been landfilled. Recovering marketable plastics from this stream is seriously challenging for a number of reasons, exposed in "ASR management". Carefully sorted resins are eligible for material recycling; the balance may be subjected to

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thermal techniques such as incineration, gasification, or pyrolysis.

Finally, future tendencies are analysed and the available and future options are compared in the "Future evolution" and "Conclusions".

In a second paper, presented at the International Symposium on Feedstock Recycling (ISFR) of polymeric materials, the fate of waste electrical and electronic equipment (WEEE) is discussed. Large WEEE (white goods) is often processed together with ELVs, yet despite some parallel features some important differences occur in treating ASR and WEEE plastics. The latter show an even wider range of resins used, with much more styrenics and thermosets. Brominated flame-retardants pose more of a problem, since some of these have been prohibited. Hence, managing hazardous materials is more of a problem for WEEE-plastics than for ASR-plastics.

This review is based on an extensive evaluation of contemporary literature, enhanced by the teachings of earlier reviews [1–5] as well as by constant contacts with professional recycling enterprise, starting in the early eighties.

Scope and definitions

Automobile shredder residue comprehends whatever is left over after recovery of most metal from shredded cars (and other waste streams). Typically, ASR consists of plastics and rubber, foam, glass, unrecovered metal (e.g. copper wire, coated metal), stones, mud, felt and fibre, wood, water, and general road dirt and represents ca. 20–25 % of the original car weight. The fraction of each material within ASR is variable and directly related to the age, composition, and state of prior dismantling of vehicles at the end of their life. Moreover, it is very difficult to characterise ASR; the results obtained depend on numerous factors that are not readily controllable, such as the effects of co-processing with other metal scrap.

In affluent countries end-of-life vehicles (ELV) are often sold second-hand, exported to less wealthy customers in faraway Africa, Asia, or Eastern Europe, or else they end up in a scrap yard, where reusable parts of equipment or hulk are cannibalised, either systematically, or removed upon external demand. The resulting car wrecks are first depolluted, i.e. they have all car liquids eliminated, together with those parts that are either objectionable, or really valuable. Systematic dismantling of bumpers, cushions … upfront shredding is strongly advocated, yet rather rarely practised, mainly for reasons of labour cost and low demand for those values retrieved. Hazardous liquids such as fuel, lubricating oil, coolant, brake fluid… and batteries are removed mandatorily prior to shredding, in an effort to reduce the chances of facing shredder fires or explosions and to avoid polluting the residual ASR. After age or an accident ended their useful life, old cars (by average after 13.7 years, cf. Febelauto data in: OVAM 2008 [6]) are still a source of spare parts, or, after shredding, of secondary raw materials, in the first place ferrous & non-ferrous metals.

The first shredders (U.K.: fragmentisers) appeared in the USA in the 1950s, after rising numbers of abandoned cars started disfiguring highways, roads, and alleys. The number of shredders rose rapidly and in the Western world ASR soon evolved into a bulky waste stream. Waste Management Authorities became interested in curtailing its substance and controlling the hazardous or polluting materials contained, e.g. heavy metals such as mercury, lead, cadmium, chromium^{VI} … and persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and heritage brominated flame-retardants (BFRs). Shredder operations have considerably matured over the years; now, not only they address an evolving mix of cars of various makes, but also of other metal-bearing waste streams, such as white and brown goods, miscellaneous equipment and structures, or whatever sources of metals increasing in value by size reduction and subsequent sorting. Recently, an attempt to define best available technology was accomplished in England [7].

Car shredders are manufactured in different sizes, typically involving one heavy fast-turning rotor, revolving in a vertical or a horizontal plane and equipped with (mostly) swinging hammers. These tear and shred the car hulk until its parts are reduced to typically less than a fist, and then the fragments pass through grids and leave the rotor housing. The rotary movement creates a fanning action that blows out the fluff, light fraction, shredder dust (U.S.), or fragdirt (U.K.); dense shredder dust leaves together with the metal mainstream, i.e. through the grid.

Before the advent of shredders, shears treated these streams of bulky metal. The successive steps in shredder development, design, and the various operating principles applied are told in [8]. Pioneering names are Newell, Proler, Luria, and Hammermills in the USA, Lindemann in Germany, Svedala in Sweden, Fuji-Car, Kawasaki Heavy Industries and Morita (anciently Tezuka) in Japan. Upstream equipment tries to provide even feed; it may consist of off-site balers, or on-site feeders, and preshredders. Downstream equipment is required to recover and separate both metals and fluff. Dust abatement proceeds mostly wet (the USA), damp or dry (the EU). Nonferrous scrap should be further separated, typically by successive sink/float classifications involving media of different density, and yielding light, medium, and dense metal fractions. The light fraction consists of magnesium and aluminium alloys and it incorporates dense plastics and rubber [8].

Shredding is energy-intensive, typically with capacity of 1000–3000 hp (1 hp = 746 W) in the EU and even 2000–4000 hp in the USA. In the UK BAT-study [7] they were classified as small (<1000 hp), medium (1000–3000 hp), large (3000–5000 hp), and mega (>5000 hp). Shredder enterprise is relatively capital-intensive, with heavy shredder equipment and sorting lines typically costing 5–10 M€. Building and Construction (B&C) waste is another EU priority waste flow, designated for reduction and recycling; given the low amount of metal it is addressed, however, in entirely different demolition yards.

In the early 1970s cars were baled prior to their cryogenic cooling and shredding in a Belgian pilot plant (Ets. Georges, Ile Monsin, near Liège): steel plate becomes brittle below ca. −50 °C. Conventional shredding is confronted with the ductility of steel, which is wrinkled and thus incorporating extraneous matter, such as electric wire. This leads to loss of copper, as well as lower quality of the now copper-bearing steel scrap. Cryogenic shredding shatters steel plate to pieces the size of an inch; these are flat and do not harbour any inclusions: the resulting steel scrap is lean in copper. Stainless steel, aluminium, brass, etc. remain in massive pieces. Shredding was conducted in units of only 500 hp. A typical car consumed ca. 600 l of liquid nitrogen, however, rendering cryogenic operation too expensive [9].

The EU ELV Directive (2000/53/EC), as well as Japanese Legislation calls for high recycling rates, resulting in a reduction of the amount of ASR to be landfilled [10]. Increased recycling of plastics from ASR is the key to achieve the European reuse and recycling target of 85 % by 2015 [10, 11]; it is expected that an additional 6–10 % of the total ELVs mass can thus be recycled. Conversely, the share of plastics in new cars will continue increasing with time, because of the trend towards lower vehicle mass [12]. Car hulks from glass-fibre reinforced resins as well as a rising share of plastics and light metals in a car could considerably exacerbate the mandatory recycling problem: car hulks from glass fibre reinforced plastics are a real nightmare for shredder operators [13]. Also hybrid cars pose problems.

Legal status

Concerns about old cars started in the USA, with the Highway Beautification Act inspired by Ladybird Johnson's campaign to make the U.S. local landscape wreck-free [8]. Sweden, Switzerland, and Japan also showed early concerns about car wrecks and how to manage them. Sweden pioneered policies for vehicle recycling, enacting a deposit-refund programme in 1975. In 1997 it opted for an Ordinance on Producer Responsibility, requiring manufacturers to accept ELVs free of charge. Swedish automobile industry soon started environmental car recycling using advanced methods for dismantling and sorting vehicle components [14]. Switzerland and Japan acted as forerunners in the thermal conversion of ASR, both driven by scarce landfill capacity. Swiss automobile importers set up The Foundation for the Environment-Friendly Disposal of Motor Vehicles (1992); Switzerland became the first country to dispose of all shredder residues by thermal processing [15]. Dutch automobile industry established auto recycling Nederland (ARN) to collect scrap cars and supervise their dismantling and recycling, without cost to the last owner. Waste disposal fees are financed during car registration [14]. ARN also proposed the Eco test as a decision support tool for managers who need to base sustainability decisions on facts about the production chain [14].

The EU addressed the ELV-problem by means of several important Directives: ELV Directive of the European Parliament and of the Council of 18 September 2000 on end-of-life vehicles 2000/53/EC [10, 11, 16]; Directive 2000/76/EC (December 4, 2000) on the incineration of waste [17]; Directive 1999/31/EC (April 26, 2000) on the disposal of waste in landfills [18]. Also the EU Regulation on Registration, Evaluation, Authorisation, and Restriction of Chemical substances (REACH, 2007) [19, 20], intended to protect people and the environment from chemical hazards, impacts the potential recycling of automotive materials.

The European Union (EU)

The ELV Directive seeks to prevent pollution and render vehicle dismantling and recycling more environmentally friendly. It emphasises the importance of increasing the use of recycled materials in vehicle manufacturing. Carmakers must reduce their use of hazardous substances (mercury, hexavalent chromium, cadmium, or lead) when designing and producing vehicles. Producers must use International Organization for Standards (ISO) guidelines for labelling and identification of vehicle components suitable for reuse and recovery and meet the costs for collection and recovery. EU Member States are required to establish collection systems for ELVs and ensure that all vehicles are transferred to authorised treatment facilities, through a system of vehicle deregistration based on a certificate of destruction. The last holder of an ELV may dispose of it free of charge (free take-back). Vehicle dismantlers must obtain permits to handle ELVs. Storage and treatment of ELVs are strictly controlled through de-pollution procedures and designated parts removal requirements (cf. 2006/12/EC [21]). Vehicle manufacturers compile data and report regularly to the authorities designated. Every 3 years Member States are required to report to the European Commission on implementation. The ELV Directive (2000/53/EC) is an early application of the extended producer responsibility (EPR) principle.

In the EU ASR is classified as hazardous waste. Still, most ASR is landfilled, even though to a rising extent energy is recovered, mainly by co-combustion with municipal solid waste [22] or in cement kilns. The ELV Directive states that by 2005 only 15 % of the vehicle's weight can be disposed of (i.e. landfill or incineration without heat recovery), and by 2015, only 5 %. A further 10 % can be incinerated with energy recovery; yet many countries do not have the incinerator capacity required, due to strict European Pollution Regulations and requirements regarding thermal efficiency. The application of the ELV-Directive has been monitored in the different Member States [11]. Verifying recycling efficiencies is stymied by the variable composition and state of dismantling of old cars and by the circumstance that these are a fraction only of shredder input. Several reports deal with these issues [23, 24].

Japan: South Korea

In Japan a law on automotive recycling was implemented in 2004 [25]. It requires to retrieve chlorofluorocarbons (CFCs), airbags, and automobile shredder residue (ASR) from ELVs and to recycle remaining materials properly. Therefore, most car producers invested in recycling business and developed easy-to-recycle cars by adapting their design. Targets for recycling were set at 50 % of ASR by 2010 and at 70 % by 2015 (including thermal recovery), values comparable to those in the EU [26]. Under the 2002 ELV Recycling Law, based on shared responsibility, consumers in Japan pay a fee when they purchase a new car (or at the time of the first mandated regular inspection), managed by the Japan Automobile Recycling Promotion Center (JARC) [27]. Recycling rates are high: parts recycling 20–30; material recycling 50–55 %; ASR recycling 12 % of ELV; only 5 % of ELV was landfilled [28].

South Korea's 2007 Act for Resource Recycling of Electrical and Electronic Equipment and Vehicles [29, 30] creates a framework to hold producers and importers responsible for their use of resources. The law also addresses the use of hazardous substances, the recyclability of materials, collection of ELVs, recycling rates, and information exchange through an on-line database.

The USA

The USA has no federal law governing EPR, ASR (or WEEE): they prefer the term product stewardship [31, 32] calling upon all parties involved (i.e. producers, manufacturers, retailers, users, and disposers) to share responsibility for reducing their product's impacts on the environment. The focus has been on voluntary measures to address contaminants (e.g. mercury switches) or to highlight specific recycling goals. The National Vehicle Mercury Switch Recovery Program (NVMSRP) [33] has launched a voluntary effort to promote a safe removal of mercury switches from ELVs before shredding. End of life vehicle solutions (ELVS) [33] is a national not-for-profit corporation formed by automobile manufacturers; it provides educational materials and collects and recycles automotive switches at no cost to dismantlers and recyclers. The partnership for mercury-free vehicles developed model legislation to address this removal [34]. Policy makers have paid particular attention to vehicle tyres, supporting their recycling and reuse. A number of individual States have taken specific actions to prevent pollution associated with mercury, scrap tires, and lead-acid batteries (EPA).

Ford, Chrysler, and General Motors founded The Vehicle Recycling Partnership (1992) [35]. The International Materials Data System [36] facilitates reuse and recycling, through sharing information on materials used in vehicles. Life cycle analysis (LCA) is used to evaluate the modes manufacturers develop vehicles [14].

ELV processing

Car dismantling, crushing, and flattening

Car dismantling refers to the selective removal of parts, either for use as spare parts, or to retrieve valuable materials that otherwise could be lost to ASR, or also to take out potential pollutants, such as mercury-containing switches, car batteries, and also all car fluids (motor fuel, hydraulic oil, coolant, CFCs, etc.). An excellent Guidebook [37] describes appropriate procedures. Recoverable parts may include wheels and tyres, steering columns, fenders, radios, engines, starters, transmissions, alternators, selected plastic parts and components, air bags, glass, foams, catalytic convertors, and other components, based on aftermarket demand. Car Recycling Systems developed a proprietary dismantling line technology cf. http://www.youtube.com/watch?v=98JWyfGGsko.

Car dismantling may also address some large items, such as bumpers, cushions, or dashboards [38]. Bumpers are often from polypropylene, yet they may be glass fibre filled and attached with nylon clips. Filled resins cause much wear when being reprocessed. Car cushions could provide polyurethane (PUR) foam unpolluted by the dust, heavy metals, and organics of fine shredder dust. There is some market for compressed PUR, yet there is already ample supply from working PUR in new foam blocks. In the Netherlands ARN has extensive car dismantling procedures; yet, this tendency meets with some disapproval, because of associated labour cost and poor outlets for most materials retrieved. Matching supply and demand is problematic; the process of identification, storage, and retrieval of parts is costly.

Car crushing is suitable for countries lacking a dense shredder plant network [39]. The car is flattened, reducing its volume. Transporting flattened cars is more effective. Decontamination must be complete before baling or flattening with a mobile press or a crane grapple. Mobile equipment tours car cemeteries in sparsely inhabited countries.

Shredding and separation

The shredder reduces the metal hulk to wrinkled pieces of steel leaving the rotor housing through one or more grates. Dense fluff follows the metal; light fluff is blown out and gathered by large cyclones. Both fractions report to conveyor belts that move the materials through some steps of sizing and separation: redundancy improves both yield and purity. Magnetic separators sort out the ferrous fraction from composite flows; large electromagnetic separators have been partly replaced by strong fixed magnets, incorporating specific rare earth elements. The magnetic fraction is cleaned by magnetic agitation, i.e. exposing magnetic items on a conveyor or in free fall to a field of variable polarity so that they turn over and liberate adhering plastics or textiles. Similar results can be pursued using ballistic separators, powerful air knives, etc. Magnetic systems may select between small and large and between strong and weakly magnetic materials. After removing all magnetic metal thoroughly the non-ferrous metals are rejected from ASR by eddy current separators. Heavy media sink/float units separate the non-ferrous fraction on the basis of density, generally in light metal (magnesium and aluminium alloys), intermediate- and high-density metal (mainly copper, zinc, and stainless steel fractions). An alternative is exportation to low-income countries, for manual sorting. Surface-coated or plated metal may deceive visual or instrumental identification. Sorting also supplies some composite fractions, such as plastic-coated wire, rubber-coated metal parts, etc. Typically, EU shredder plant practises dry classification. Wet treatment, by washing, elutriation, settling, and hydro-cyclones may further improve purity [8].

Automobile shredder residue

Composition and characteristics

ASR is a heterogeneous mix of residual ferrous and non-ferrous metals (5–23 %), plastics (20–49 %), rubber (3–38 %), textile and fibre material (4–45 %), wood (2–5 %), and glass (2–18 %) [22, 40–42]. Its composition depends on the input materials, the nature of the shredding operations, and the efficiency of post-shredding processes [22, 43]. The shredder input is rarely restricted to cars; typically, these represent only 20–30 % of the input and other feed materials are composed differently [13]. Data comparison is impossible, due to differences in feed, processing methods and sorting efficiency [25]. Moreover, different authors use distinct classification procedures, e.g. foam (PUR) is sometimes separated, yet typically included in the plastic fraction; ‘wire’ usually is included in the residual non-ferrous fraction, yet reports partly to non-metal residues. The heterogeneity of the feed material, its variable moisture and ash content, and calorific value, as well as its somewhat unpredictable level of contamination (heavy metals, PCBs, PCDD/Fs), also constitute considerable challenges when selecting or designing an appropriate ASR treatment process [43, 44]; de facto ASR is classified into [44]:

- Light fluff, generated during shredding, following spontaneous air classification (75 % of ASR; 10–24 % of the ELV)
- Heavy fluff, remaining after metal separation from the heavy shredded fraction (25 % of ASR; 2–8 % of the ELV)
- Fine soil and sand are sometimes reported separately, but usually part of the heavy ASR (<2.5 % of the ELV)

Most shredders separate dense metal, dropping out of the rotor chamber, from fluff entrained by the air currents provoked by the rotor. Both flows are treated by magnetic separators (cross-belt and in-line overhand and pulley units) retrieving iron and steel, eddy current extractors ejecting non-ferrous metal sized by trommel screens, sieves, ballistic, and aerodynamic separators, etc. The purpose is recovering a maximum of magnetic metal and of even more valuable non-ferrous metal (high yield), with a minimum of adhering plastics, dirt or other contraries (high purity). Each separator performs best on narrow size fractions; repeated separation, magnetic agitation, and aerodynamic cleaning enhance purity, minimising foreign inclusions and adhering material. Such processes generate several fluff fractions that together constitute the total ASR stream. The precise subdivision of ASR into fine and coarse sieving fractions or into light and dense air classification fractions varies from one plant to another, depending on the input materials and the configuration and operating conditions of fluff treatment steps. Shredders differ in grids, separating dense materials from fluff [8]. Particle size ranges widely, from <125 μm up to some 10 cm. Only a small fraction of ASR (2 %) is larger than 10 cm and consists of large pieces of foam, rubber or

Table 1 Composition of ASR, light and dense fraction [25]

	[45]	[6]	[40]	[46]	[47]	[48]	[49]
(a) Composition, as wt%, of ASR, according to the origin: light fluff							
Metal	1–1.7	21	8.8	2.5	3.7	0.3	
Wire	2.9–3		4.7	1	2.2		0.5
Rubber	3.8–4	3.1	2.6	3	8.8	10.3	4.1
Textile	37.5–39.6		36.1	32.5	26.2	8.3	7.9
PUR foam	6.6-20.6		35.3	8			3.8
Plastic	16.1–24.1	31.8	11.7	9	46.1	11.0	8.7
Wood	0.03–0.4			1	2.7	0.6	
Paper	0.8–1.0				0.8		
Soil/sand	6.4–21.6				4.3		
Glass	0	2.3		43 (minerals)			
Others	2.7–6.2		0.8		5.2	69.5 (<10 mm)	75 (fines)
(b) Composition, as wt%, of ASR, according to the origin: heavy fluff							
Metal	0.2–1.4	1.6		5		0.7	
Wire	7.0–12.7			3			0.7
Rubber	14.1–17.3	9.3		55		43.7	44.8
Textile	7.7–11.6			3		10.5	10.5
PUR foam	0.9–2.8						3.3
Plastic	23.8–30.9	8		19		32.6	29
Wood	0.06–0.7			7		4.7	5.6
Paper	1–2.5						
Soil/sand	7.6–12.3			8 (minerals)			
Glass	8.3–11.0	9.4					
Others	4.6–4.0					7.8	6.1 (fines)

plastics. The moisture content ranges from 0.7 to 25 % (depending on off-gas treatment and local climatic conditions) and the ash content varies from 18 to 68 %. The calorific value ranges from 14 to 30 MJ/kg.

Light and heavy fluffs both contain combustibles (typical higher heating value, HHV), such as plastics (ca. 38 MJ/kg), rubber (23 MJ/kg) and textiles (17 MJ/kg) (Table 1a, b). Heavy fluff contains more rubber and non-combustibles; light fluff more textiles, and low-density plastics. Reported HHV values vary significantly [25]. Light fluff contains up to 70 % of fines (Table 1a). The coarse fraction can be defined as exceeding a given mesh size, up to ca. 2 cm; roughly half of ASR reports to the coarse fraction, mainly consisting of PUR (foam), plastics, rubbers, and textiles. In principle, it might sustain mechanical separation into individual possibly recyclable fractions. With lower ash content and high calorific value (15–30 kJ/kg) it can also be used as a fuel. The fine fraction comprises smaller pieces of glass, plastics and metals, along with rust, dust, and dirt. It has a higher ash content and lower calorific value (11–21 MJ/kg) and is less suited for combustion or recycling. Fines are particularly polluted with heavy metals and mineral oils; they are not considered fit for recycling as materials, due to their complex composition and low market acceptance. Only a use as filler, binder, aggregate, or as landfill cover may divert this fraction from landfill [50].

ASR plastics

Plastics in general (mainly from packaging) are recovered very differently in various European countries (Fig. 1). The introduction of the EU-directive on ELVs created strong incentives to reduce the number of different plastics used and to label plastic parts, to facilitate identification during dismantling. Several studies pointed out that these plastics could be recovered from ASR with sufficient purity. The quality of these recycled plastics remains an issue. Shredding companies using post-shredder techniques for separating plastics still suggest that the reuse and recycling target of 85 % (mandatory from January 1, 2015) could be reached.

Up to 27 different types of plastic resins are routinely encountered in ASR. Plastic car components consist of polypropylene (PP), polyurethane (PUR), polyvinylchloride (PVC), acrylonitrile butadiene styrene (ABS),

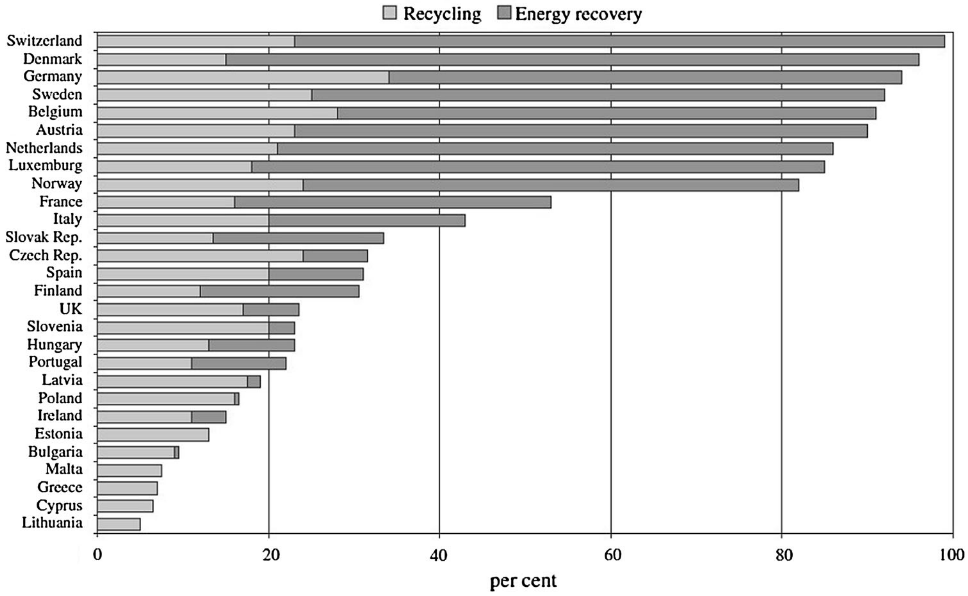

Fig. 1 Recycling of general plastics in various European countries [51]. Recycling rate, mechanical recycling; energy recovery rate, thermal recycling

polymethylmethacrylate (PMMA), polyethylene terephthalate (PET), etc. Each resin still varies in its specific formulation with additives. Their recovery has remained marginal; yet it should develop rapidly now, given the EU requirements regarding material recycling. Manual dismantling allows cleaner recovery, yet it is severely limited by productivity and by the composite, at times embellished structure (e.g. bumpers) of some car components. Conversely, it seems illusory to recover really clean materials, such as PUR, after passing them through a shredder, even though washing procedures have been tested [12]. Plastics' recycling comprises:

- mechanical sorting into different fractions of acceptable purity, to be re-granulated and sold, and
- thermal treatment to recover either chemical building blocks (feedstock or chemical recycling) or heat or fuel (thermal recycling, considered as recycling in Japan, contrary to the EU).

Argonne has pioneered ASR treatment, followed by its only licensee Salyp (Belgium), defunct at present [52, 53]. Their approach focused on three fractions: ABS, PUR-foam, and fines for cement making [12]. An EU survey (2006) mentions several processes [54] running at an industrial scale (Galloo, Sult, R-Plus, Twin-Rec). Galloo, Sult, Citron, and R-Plus operated their proprietary technology; other systems (VW-Sicon, TwinRec, Reshment) are to be licensed to operators. Reshment was never actually tested. Both Citron (Le Havre) and SVZ (Schwarze Pumpe) ceased their activities.

Some enterprises test and develop separators and commercialise sorting processes or systems. Hamos GmbH Recycling- und Separations technik [55] has developed a system for recovering plastics from mixes, yielding ABS, PS, and PP fractions. After liberating large pieces of metal, the mixed plastics are classified and a light fraction is sorted out. Subsequent wet separation allows sorting it into different subfractions, graded according to their density. Then the ABS-PS (styrenics sinks in water) and PE-PP concentrates (polyolefins float) are mechanically and thermally dried. Remaining metal and other conductive contaminations are subsequently separated, using hamos electrostatic separators. Downstream electrostatic separators convert the ABS-PS or PE-PP mixtures into separate fractions. If required, light coloured plastics fractions can be produced using optical-electronic sorting. WEEE-plastics can be treated similarly: Axion selected six sorting systems with financial support from WRAP after comparative testing of a wider equipment selection [56].

Whatever the merits of these separation and cleaning processes, part of the ASR will need to be burned. Testing and analyses conducted in different parts of the world all agree that this combustion is technically and environmentally problematic, yet co-incineration with limited addition of few per cent ASR to another mainstream is feasible without violating emission standards. Indeed, ASR typically contains significant amounts of chlorine (Cl), sulphur (S), nitrogen (N), and a wide range of heavy metals.

Hazardous components and compounds

ASR holds fuel hydrocarbons and other organics. It tends to spontaneous heating and ignition and consequently it

Table 2 ASR contaminants [58]

Compounds	PCDD/Fs	Dioxin-like PCBs	PCB	PAHs
Concentrations	242–329 ng I-TEQ/kg	481–631 ng I-TEQ/kg	13–15 ppm	37–140 ppm

needs careful processing on an occupational health, as well as on a plant safety basis.

ASR routinely contains a range of heavy metals of miscellaneous origins. Various paint pigments, plastics additives, and corrosion products bring in a number of heavy metal compounds; brominated flame retardants (BFRs), e.g. from printed circuit boards or car cushions are routinely accompanied by antimony oxides. Mercury is still used in some switches, relays, and gas discharge lamps, as discussed earlier, regarding US-initiatives. Shredding a single lead battery would impregnate a lot of ASR with lead and its oxides.

A comparison of recent ASR data with earlier analyses shows that the elemental composition of ASR has hardly changed with time. Heavy metals in ASR are cited as zinc (Zn, 2.10 %), copper (Cu, 1.85 %), lead (Pb, 0.26 %), chromium (Cr, 0.16 %), nickel (Ni, 0.12 %), antimony (Sb, 230 ppm), cadmium (Cd, 77 ppm), and mercury (Hg, 3 ppm). When co-incineration with 5 % ASR takes place without extra pollutant reduction, the concentrations for each metal in the residues increase by a factor of 1.1–1.8 when compared with incinerator residues from without ASR [57]. The calorific value (12.2 MJ/kg) and the PCB and Cl concentrations are comparable to those of municipal solid waste [57].

ASR may also contain significant levels of organochlorinated POPs (cf. Table 2), in particular polychlorinated biphenyls (PCBs), dibenzo-p-dioxins (PCDDs), and dibenzofurans (PCDFs) derived from diffuse and undefined sources, such as capacitors from another age. Fortunately, the time trend is clearly descending. ASR fuel value is impaired by the excessive presence of chlorine (most fuel users specify <0.4 wt% Cl) and heavy metals [46]. The chlorine content of ASR ranges from 0.5 wt% (after careful elimination of dense plastics) to >4 wt% due to the presence of chlorinated polymers, mainly PVC and halobutylrubber. Also brominated flame retardants (BFRs) are present, e.g. in cables, cushions and upholstery, and in electronic components. The amount of chlorine is somewhat too high for using ASR directly in cement- and limekilns or for regular co-firing in solid fuel-fired power plant. Care should be taken to avoid combining copper wire with halogen compounds; together they lead to the formation of toxic compounds such as PCDDs (dibenzo-p-dioxins) and PCDFs (dibenzofurans) necessitating proper flue gas treatment to avoid emissions to the environment. Such treatment is unavailable at cement or lime works or in power plant. Van Caneghem et al. [58] report the following concentrations (Table 2).

Other sources report somewhat lower PCB concentrations, ranging from 0.5 to 7 ppm; these PCBs report more to the coarse fraction than to the fines [22, 59]. ASR fingerprints of PCDD/Fs are dominated by higher chlorinated PCDD congeners [60]. Hedman et al. [61] reported fingerprints of the textile and leather fraction from MSW, similar to those of ASR. These data suggest distinct sources of PCBs on the one hand and PCDD/Fs on the other.

ASR management

Survey

In the absence of better as well as affordable solutions landfill has been the traditional disposal route for ASR. Sanitary landfills are engineered to contain various wastes and ASR seems manageable in this context. Often, leaching procedures are used to assess acceptability in landfills. Washing may be helpful for attaining final storage quality [62] and avoid refusal at a landfill site, e.g. because of an excessive PCB-content. ASR-landfill fires are to be avoided at all costs, taking into account the contaminants contained. Such ASR fires are quite polluting [63] (as are car fires). Yet, ASR is light and bulky and tends towards spontaneous self-heating and possibly ignition. Legal, as well as land constraints created the need to avoid landfill, stimulate materials and energy recycling and develop additional post-shredder recovery pathways, yet these new technologies, still today, are rarely applied at industrial scale. They concentrate on extracting plastics from ASR and occasionally on fibres, fillers, etc. The main hurdles are cost and outlets for recyclate. Glass represents some 3 wt% of a car, yet it is never earmarked for recovery [64–68].

Chemical (or feedstock) recycling and (until recently) mechanical recycling are rather exceptional as a daily practice, yet highly publicised as R&D objects. ASR has been pyrolysed during Thermoselect test runs [69]. ASR gasification is proven technology, since the TwinRec plant (Ebara Co.) has operated for almost 12 years at Aomori (Japan) on ASR and various commercial and industrial wastes [70]. In Germany, Austria, and Japan several methods were developed for applying ASR in metallurgical processes, including the blast furnace (Bremen Stahl, Nippon Steel) [71–75].

Co-incineration is practised much more than any other route: vigorous recycling of Municipal Solid Waste fractions has created quite some spare capacity in numerous incineration plants.

ASR incineration

Modern incinerator plant is capable of continuous operation with only one or two annually scheduled shutdowns and plant availability of up to ca. 92 %. Heat recovery by raising steam at typically 4 MPa allows generating electric power, mostly with a heat-to-power conversion efficiency ranging from 15 to 24 %. MSW incineration typically costs 100 €/tonne in the EU; yet possibly ca. 250 €/tonne in Japan. In principle, cost is proportional with heating value: thus ASR pays a premium over MSW.

At first sight, it could appeal burning ASR directly in such plants, recovering some supplemental metal from incinerator residue and possibly even from fly ash. Direct ASR incineration in mechanical grate furnaces stumbles over both technical and environmental factors. Already in the early 1980s this concept was tested together with Belgian shredder enterprise (IWONL/IRSIA R&D-project), but due to its high calorific value and melting characteristics the ASR burned too fiercely, leading to uncontrollable combustion, local lack of oxygen, agglomeration of burning ASR, and carry-over of unburned fines [76].

Fluidised bed and vortex combustion probably provide better means, as long as feeding is steady and ash extraction fully ensured. Still, environmental factors should be evaluated with caution. ASR combustion releases both hydrochloric and hydrobromic acid (arising mainly from PVC and chlorinated rubbers, and from brominated flame retardants, respectively). These acid gases lead to low temperature corrosion and damage metal equipment. Heavy metals, particularly the more volatile ones, sublimate as chloride and bromide and condense onto the finest particles, requiring efficient and hence expensive gas cleaning systems. The presence of highly volatile elements, such as antimony or mercury, could cause formation of difficult-to-remove aerosols and demand even more robust measures at the side of flue gas cleaning plant.

Mixing ASR with lower calorific waste improves incineration ease and efficiency in waste-to-energy plants, while supporting considerable mass and volume reduction and energy recovery. Co-incineration can be conducted in a wider range of incinerator categories, such as mechanical grate furnaces, fluidised bed and vortex combustors, rotary kilns, and cement kilns. Such solutions are called Solution by Dilution by operators of duly licensed hazardous waste incinerators.

If ASR is classified as a hazardous waste, as in the EU, special licensing generally will be required.

Gasification of ASR

Gasification was widely developed, starting from the nineteenth century, at that time to extend the production of town gas manufactured extensively by the coking of coal. After World War I new needs for synthesis gas were created by the synthesis of ammonia, methanol, OXO-aldehydes and -alcohols and—culminating during World War II—the Fischer–Tropsch synthesis of liquid synthetic fuels from solid coal and lignite, both in Germany and Japan. Several major processes have evolved:

1. Fluidised bed gasification (Winkler, Leuna Werke, Germany) at unusually low temperature (750–900 °C). An air factor was used of 25–40 % of the stoichiometric combustion air requirements: partial combustion of char supplies the heat required for autothermal operation. Gasification of carbon with steam or carbon dioxide is still slow below 850 °C. Yet, operating temperatures are restricted by weakening of the ash, leading rapidly to loss of fluidisation. Air as gasification agent introduces large amounts of nitrogen in the producer gas, thereby reducing the calorific value of generator gas.
2. Vertical shaft gasifiers (Lurgi, Frankfurt) operate in counter-current, with rising gasification products and the charge gradually descending towards the hearth at temperatures up to 1200 °C. Air is replaced as a gasification agent by a balanced mix of oxygen and steam. Ash is tapped periodically as molten slag, unless gasification temperature is severely limited.
3. Also a rotary kiln can be used, operating at a higher ratio of (volume reacting gases)/(volume of the charge) and adding tumbling action to the charge. The Landgard partial oxidation plant of Monsanto (Baltimore, MI, USA) was scaled-up too rapidly from pilot to full-scale and fell victim to sequentially occurring operating problems, starting with shredder explosions, solidification of stored shredded fuel, clogging of the slag tapping hole, etc.
4. Co-current flow combustion of pulverised fuel (Koppers-Totzek). The reactor is simply a combustion chamber, into which several pulverised coal burners inject their incomplete combustion products. Temperatures of 1400–1600 °C are reached, so that tar conversion and a fully slagging operation is ensured.
5. Montecatini, Texaco, and Shell all developed processes for converting heavy fuel oil (or any pumpable fuel), injecting these together with oxygen and steam into a combustion chamber at sufficiently high temperatures. Their designs differed mainly in the methods used to separate soot particles and to recover sensible heat from the gas.

Producer gas from Winkler units consists of a mixture of drying vapours, pyrolysis, and gasification products arising from flash drying, fast fuel pyrolysis, and gasification of carbonised residues, respectively. The presence of pyrolysis products is unproblematic in case the producer gas is fired directly, without intermediate cooling, in a furnace or a gas turbine. In synthesis gas applications, however, pyrolysis products must be removed carefully, e.g. by scrubbing with deep-cooled methanol (Lurgi), or by thermal or catalytic conversion of tars, hydrocarbons, etc. to more gas. Obviously, treating tar considerably complicates the plant and inflates its cost so that almost all waste gasification units opted for direct firing of gas. Producer gas from Lurgi units also contains the full range of low- and high-temperature feedstock pyrolysis products. Only the Koppers-Totzek units operate at sufficient temperature for destroying virtually all pyrolysis products.

Gasification has been widely applied for coal and heavy oil; it has been studied and tested for municipal solid waste, biomass, and plastics. In the 1970s both Andco–Torrax and Union Carbide failed to launch vertical shaft gasifiers, dedicated to MSW. Nippon Steel was more successful. During the nineties, Japanese authorities decided that there was a need for innovative thermal techniques that would be leaner in their total dioxin output. So, Nippon Steel secured part of this new market using its already proven technology. Ebara proposed gasification/melting, combining two proven units: an internally circulating fluidised bed followed by a slagging fixed combustion chamber. The combined concept, *TwinRec*, was first demonstrated in a privately owned and operated plant at Aomori. The plant accepts a wide range of waste, yet its base load consists of ASR, provided on captive basis. Complete mass balances were established during ad hoc test runs for numerous chemical elements. The granulated glassy residue was subjected to leaching tests, not only according to Japanese, French, and Swiss test procedures, but also following the highly demanding Dutch test methods [77]. Another unit treats ASR using a fluidised bed gasifier (590 °C) followed by a cyclonic afterburner (1400 °C); it is another example of sequential gasification and combustion [78, 79].

Tests with ASR were conducted at Schwarze Pumpe (SVZ; Global Energy), at the site of a traditional producer of methanol by gasification of lignite [43]. Sequential gasification/incineration tests were reported in a rotary kiln operating between 850 and 1120 °C as a first stage [44]. Combustion is completed in a second stage, featuring an afterburner chamber with waste heat boiler (steam at 4.3 MPa, 430 °C) and steam turbine. Tests were conducted at a capacity of 2400 kg/h. The composition and the characteristics, the combustion properties and the ash analysis of the ASR, as well as the composition of the exhaust gases are all well documented; some operating problems, data on residue management, and energy efficiency figures are reported on. It was concluded that the process still needs minor modifications.

Catalytic gasification of ASR was conducted in a lab-scale fixed-bed downdraft unit to generate hydrogen [48]. A 15 wt% NiO/Al_2O_3 steam reforming catalyst was used at 760–900 °C. Optimistic up-scaling figures are presented.

Pyrolysis

Polymerisation of reactive monomers, such as vinyl compounds or olefins and dienes, or gradual polycondensation to form polyesters and polyamides, is thermodynamically favoured at low temperature. With rising temperature this trend gradually reverses and monomers become more stable than their polymers. Above 500 °C all organic compounds turn unstable and tend to carbonise, i.e. convert into their elements carbon C, hydrogen H_2 and possibly some simple stable molecules, such as water vapour H_2O, carbon dioxide CO_2 and monoxide CO, hydrogen fluoride HF, hydrogen chloride HCl, hydrogen bromide HBr, ammonia NH_3, hydrogen cyanide HCN, etc. Even though they are unstable, some compounds still stay sufficiently robust to survive thermal treatment for hours (low-temperature pyrolysis, at 400–500 °C) or seconds (high-temperature pyrolysis, above 750 °C). Methane CH_4 is the most stable member paraffin, and ethylene C_2H_4 the most stable olefin. Monocyclic as well as some polycyclic aromatic compounds (PAHs), such as diene compounds (e.g. butadiene, isoprene), acetylene compounds (C_2H_2 and higher acetylenes), and allene ($CH_2{=}CO$) all show some thermal stability. Stability declines faster for paraffins > olefins > diolefins > aromatics.

Only few polymers (e.g. polymethylmetacrylate, polytetrafluorethylene and polymethylstyrene) decompose along lines that really reverse their synthesis and produce monomer in good yields. Most plastics decompose by very complex mechanisms that are profoundly influenced by reaction conditions (temperature, pressure, gas phase composition, reaction time) and the presence of impurities (additives), and catalysts. Pyrolysis first forms primary products, with structures close to those of the fragmented parent molecules. Primary products convert into secondary, tertiary… products. Ultimately, the nature of the pyrolysis products becomes virtually independent of the parent molecules and depends mainly on their relative thermal stability.

Pyrolysis of pure plastics is most commonly run in the liquid phase at moderate temperatures (400–500 °C) in the absence of oxygen. Organic vapours, condensable to pyrolytic liquids, are formed, together with permanent gases (H_2, CH_4, C_2H_4, C_2H_6, C_3^+, CO, CO_2) and a carbonising solid residue, continuously losing weight with rising time and/or temperature. The relative yield of liquids, gases, and solids depends on nature and composition of the feedstock and on operating conditions.

Polyolefins, such as polyethylene, polypropylene… convert almost entirely into condensable volatiles. Their boiling range lowers in temperature with rising pyrolysis temperature: low-temperature pyrolysis (400 °C) yields paraffinic waxes (PE) and vaselines (PP). Rising temperatures give rise to lighter hydrocarbons, boiling in a range of gas-oil (240–350 °C), kerosene (180–240 °C), or even naphtha (<180 °C), as pyrolysis temperature rises from 450 to 600 °C. Cracking severity can be artificially increased by mounting a reflux cooler on top of the pyrolysis reactor, so that condensing liquids return to the reactor.

Styrenics and vinyl compounds leave more char than polyolefins. Thermosets and rubbers form mainly solid residues. Char production from ASR ranges from 33 to 68 wt%, thus largely exceeding the char amounts commonly encountered when pyrolysing single plastics or even biomass [25]. Charring of heat transfer surfaces jeopardises supplying the required reaction heat.

Product distributions are impossible to predict accurately and should thus be tested empirically, at first at small scale (e.g. thermogravimetric analysis), later under more realistic operating conditions. Differences in heating rate, in solid/vapour contact, as well as wall effects all influence upon the extent of secondary reactions and hence upon the resulting product distribution. Vacuum pyrolysis [80] minimises the occurrence and extent of secondary reactions; technically, these conditions are unsuitable, given their reduced throughput and difficult heat transfer conditions.

Industrial pyrolysis reactors are basically controlled by their heat input capacity. In industrial reactors the initial rate of heat transfer (and hence reactor capacity) is soon curtailed by coking of the reactor walls. This problem may still be tractable in the case of pure polyolefins feed; yet, it is insurmountable in cases of strongly coking feed, such as a mix of ASR plastics. Technical solutions are reactors with self-cleaning properties, or apt to remove coke, soot or residues out of the reactor.

Difficult feedstock can best be processed in fluidised bed (or rotary kiln) pyrolysis units. At laboratory scale, these are externally heated. At full-scale the most common type are the double fluidised bed unit inspired by the Fluid Catalytic Cracking units first developed (1942) by MIT and Esso, with catalyst as a heat carrier circulating between the pyrolysis bed and a combustion bed, in which carbon is burnt off and the necessary heat to sustain the entire process is generated. Tsukishima Kikai applied such techniques at Chiba and Ebara at Yokohama (the Stardust project). Both differed in the method of circulating sand between the beds. The first used the patented K + K method (Prof. Kunii & Kunugi, Tokyo University). Both were soon scrapped, yet the second still achieved its aim: demonstrating 30 days of full-scale operation using automatic control systems.

Catalytic cracking [81] has been discussed during many ISFR (International Symposium on Feedstock Recycling of Polymeric Materials) presentations. Since the contact between macromolecules and catalyst is precarious it is often proposed as part of a two-step process: thermal cracking in the liquid phase, followed by catalytic cracking. Fouling and coking are problematic; used Fluid Catalytic Cracking and various mineral waste streams were proposed as inexpensive catalyst.

Fundamental research studies mostly make use of the following:

- Thermogravimetric analysis (TGA), e.g. to define the temperature windows of fast reaction and develop kinetic data and models (yet, heat transfer controls capacity!). TGA is possibly coupled with evolving gas analysis (EGA) by means of mass spectrometry (MS), gas chromatography (GC) or fourier transformed infrared (FTIR) analysis.
- Differential scanning calorimetry (DSC) or thermal analysis (DTA) establishes the heat effects associated with (sequentially) solid phase recrystallisation, melting, and decomposition of polymers.
- Small-scale pyrolysis reactors, to establish mass balances and obtain aliquots of solid, liquid, or vapour phase products.

Pyrolysis kinetics may be derived with great care and are at times accompanied by elaborate conjunctures regarding the steps and mechanisms of chain scission, transfer, or termination. Minor amounts of impurities are greatly important, just like accidental chain defects. Relevant studies in such scientific work are those of int. al. Rausa and Pollesel [82], de Marco et al. [83], Day et al. [84], Zolezzi et al. [85], Chiarioni et al. [86] and Joung et al. [87, 88]. Pilot-scale experiments (Galvagno et al. [89]) are rather scarce.

In practice, pyrolysis reactors are controlled by the rate of heat transfer. In industrial ethylene production doubling the rate of heat input almost exactly halved the length of the tubular reactor required [77].

At laboratory and pilot scale most reactors are electrically heated. Some industrial units are using molten salts as heating agent; these techniques are rather problematic, because of salts sublimating. Low temperature salts are often mixtures of nitrates and nitrites; these are strong oxidisers, possibly reacting explosively upon the introduction of reducing agents. Also molten lead units have been used in a weekly production schedule: the lead cools and solidifies over the week-end and is cleaned from residue before restarting.

Donaj et al. [90, 91] conducted microwave pyrolysis experiments to separate metals from carbonised residues. This illustrates another potential purpose of pyrolytic processes: to recover valuable metals covered by plastics, rubber, or coatings. Liquid and solid residue from microwave pyrolysis was converted by a gasifying agent (steam, air or an air/steam mixture) heated to temperatures above 900 °C, providing the heat needed for gasification. Cleaning metal objects also proceeds by immersion into a fluid bed cleaner, or else by batch by batch cleaning in heated chambers.

Two factors jeopardise any future plastics pyrolysis projects: the economy of scale (annually, pyrolysis units typically process 4–15 ktonnes; an ethylene cracker converts 1 M tonnes of naphtha); the logistic cost to collect these plastics rise with distance.

Hydrolysis, solvolysis, glycolysis, acidolysis, or transesterification

Numerous polymers (polyesters, polyamides, polycarbonates, polyurethanes, etc.) are manufactured by means of stepwise synthesis, involving gradual growth of polymer molecules. In principle, such polycondensation or polyaddition reactions are reversible under appropriate conditions (suitable solvents, catalysts, and enhanced temperature). More information on hydrolytic processes is found in [12], with references to processes developed by Ford Motor Co., Troy Polymers, Inc. and Delphi.

Breaking down the structures of thermoset and rubbers also inspired a lot of research. Mechanical, thermal, and solvent treatment and combinations therefrom have been proposed to depolymerise and/or size reduce such materials. Rubber has been milled at ambient temperature and under cryogenic conditions; rubber crumb is applied in sport fields, as filler, in vibration dampening, etc. Rubber reclaim has been introduced into some formulations [1].

Although these reverse processes are technically feasible, the cost of collecting and processing adequate amounts of sorted plastics, thermosets, or rubber, the uncertain specifications and limited purity of the resulting products all have hampered the practice of solvolytic chemical recycling.

Solvent-based separation

The Rensselaer Polytechnic Institute, Argonne, Solvay, and Delphi Automotive Systems all developed solvent methods to separate plastics [12] by, e.g. selective dissolution, followed by precipitation. Mixtures of plastics can be separated for analytical purposes by such methods [1]. Separating pure plastics is feasible relatively well. In real life systems numerous difficulties appear. Moreover, solvent-based methods are relatively expensive, when compared to mechanical or electrostatic separation methods.

Solvay has operated its Vinyloop plant in Ferrara since the start of the century, to recover PVC from composites, such as cable scrap. A larger plant was built in Kobe, yet could not secure raw materials at economically justifiable conditions.

Mechanical recycling

Mechanical recycling of waste plastics proceeds via mainly mechanical processes (cutting, washing, drying, separating, cryogenic grinding, compounding with additives, re-granulating), producing recyclate that can be converted into new plastics products, thus substituting some virgin plastics (though often in less critical undemanding applications).

Thermoplastics may be re-melted and re-processed into products, via usual techniques such as injection moulding or extrusion. Thermosets and other cross-linked materials (rubbers, XPE…) can no longer be softened or melted without destroying molecular structures; they may be converted back into feedstock (chemical recycling) or used as fuel in cement kilns, power plants, or diverse co-incineration units.

Thermoplastics represent a wide variety of polymers with different structure, Molecular Weight, and physical and mechanical properties. Prior to their use they are compounded with a host of additives required to tailor their properties and for ensuring thermal stability during processing and photochemical stability during their use.

A major hurdle for mechanical recycling is the host of different polymers and assorted additives used. Differences in molecular weight and structure lead to distinct melting behaviour and rheological properties, causing problems of flow adaptation during conversion also when processing the same resin with different Melt Index. Even worse is a lack in compatibility: a mixture of different polymers habitually forms systems with one continuous phase and one or more dispersed phases (cf. oil in water suspensions). Such mixes have far inferior mechanical properties than the parent compounds (being brittle, still lacking stiffness): inclusions indeed lead to marked defects in solid structure, each insertion acting as a notch or a hole, lowering the resistance to internal and external strain, finally making those recyclates unsuitable for almost any application. Therefore, mechanical recycling is only feasible for pure and homogeneous, single polymer resin-and-additives streams or for undemanding applications.

Most mechanical recyclers obtain their input material from known sources: trusted collecting and sorting organisations. Since recyclate partly substitutes virgin polymers in existing applications, their market value is directly linked to virgin prices. Market value of recyclates not necessarily supports the actual costs of collecting and sorting. Collecting and handling lightweight plastics is onerous. Each processing step easily costs 50–100 €/tonne. Operating several operations, such as shredding, washing,

Table 3 Overview of some post-shredder technologies [25]

	Argonne	Galloo	MBA-polymers	Salyp process	Stena	R-plus (WESA-SLF)	VW-Sicon
Separation techniques							
Air classification	X	X	X		X	X	X
Magnetic separation	X	X	X	X	X	X	X
Eddy current separation	X	X	X	X	X		X
Screening		X		X	X	X	X
Trommel separation	X	X		X	X		
Optical sorting				X			X
Manual sorting				X	X		
Drying						X	
Sink/float separation		X		X	X		X
Froth flotation	X						
Thermo-mechanical sorting				X			
Wet grinding			X				
Hydrocyclone			X				
Static separation tanks		X					
Heavy media separation					X		

sorting, and drying in series may raise costs by 500 €/tonne.

Post-shredder technologies

Post-shredder technologies separate a plastics-rich stream first from ASR, then try and separate it into distinct resins. Systems differ in the techniques used for eliminating extraneous matter (wood, board, sand…) and for classifying and separating plastics. These differ in density, electrostatic charging, and surface properties. Successive sink/float separation steps in water, water/methanol mixtures, brine, or solid suspensions are commonly used for grading mixed plastics by density. Density-based separators include, e.g. settling tanks, hydrocyclones, and jigs. Some other techniques, as applied by Argonne/Salyp, Galloo, MBA Polymers, R-Plus and VW-Sicon are conveniently summarised in Table 3. Obviously, using different techniques for grading improves the purity of the resins separated. The original Table also cited the plants in actual operation at that time and the results achieved. Operators are most secretive both on technology and its achievements, so reliable information is hard to get.

Future evolution

Automotive industry faces significant challenges, as vehicles cause considerable environmental impacts at all stages of their life cycle, in particular while consuming motor fuels. These results in the increased use of lighter, composite materials to build a car: fuel consumption over the lifetime of a car is much more important than the oil-equivalent recovered by recycling car plastics!

Under increasing pressure from national governments and the green movement the automotive industry still signed various voluntary agreements to achieve higher recycling and recovery rates and to accept responsibility for the treatment and recycling of ELVs. To date, many countries worldwide have introduced legislation encouraging the reuse, recovery, and recycling of ELVs or even, to make these mandatory. By 2015 all EU member states must meet targets of reuse and recovery and of reuse and recycling of 95 and 85 %, respectively. Similar targets were set in Japan. These ambitious criteria are difficult to meet, as well as to monitor.

The techniques applied to classify and concentrate the different types of materials at the shredder plant are mainly air classification, magnetic and eddy current metal separation, and screening or trommel separation. Post-shredder technologies have been developed, designed, and tested to treat the residual ASR material stream, remaining after depollution, dismantling, and shredding of an ELV. Primary recovery techniques, based on mechanical or physical separation, are capable at present of recycling some 75 % of current ELVs, leaving 25 % of automotive shredder residue. New cars with a rising part of synthetic materials and of light metals makes this target more difficult to reach.

The present review assesses the fate of plastics from ELVs and ASR. Their characteristics show the presence of a wide range of contaminants. Additional post shredder systems, isolating fractions of sufficiently pure plastics, would allow meeting the reuse and recycling targets of 85 %, but only with difficulty. The plastics recovered can be recycled, yet only in undemanding applications. Splitting up ASR into low-value fractions of sufficient quality for integration into manufactured products, such as composites, concrete or asphalt also meets with severe limitations as to product quality and characteristics. A further reduction in ASR calls upon either incineration (waste-to-energy plant, cement kilns, metallurgical processes) or to the use of dedicated thermo-chemical processes, such as pyrolysis or gasification. Plastics pyrolysis processes have been developed and tested since the seventies, in particular in Japan. A critical review of developments in the full-scale pyrolysis of plastics at large [92] and of ASR in particular [43] has been presented.

Co-incineration of ASR together with, e.g. MSW, sewage sludge, etc. may be conducted, e.g. in grate-furnaces or in fluidised bed or rotary kiln units. In cement industry, only low percentages of ASR can be used, unless ASR is previously upgraded to reduce its halogen content. All energy-applications of ASR indeed need to address several environmental as well as technical issues. Applying ASR in metallurgical processes has been found troublesome, due to the variable quality of ASR, along with the presence of certain contaminants.

All these treatment methods result in environmental benefits compared to present landfill practice [93].

Shredder operations are a mature technology at present. Truly, there have been numerous innovations and improvements but no great developments are to be expected at the level of both pre-treatment and shredder technology. Moreover, any innovations in this field do not really pertain to the theme of plastics from ASR.

Plastics from ASR have been separated consequently for the past 10 years only. Today, shredder operators are still confronted with the wide variety of plastics as used in the past 25 years. The techniques used in separating plastics are only moderately innovative, since a majority of these was known already at the time of our first review [1]. The challenge is (1) to separate fractions of sufficient quality to be of use and (2) to recycle a maximum of low-quality material flows, while still ensuring environmental safety.

ASR is classified as a hazardous waste, yet it has been largely landfilled. Responsible management of hazardous compounds and components should be given ample priority. Conversely, shredder operators regard this step as supplemental cost; adequate depollution still should be ensured permanently and monitored adequately.

In summary, industry is confronted with realising conflicting goals: building lighter cars to reduce fuel consumption, using performing plastics to ensure high quality of car components, yet ensure second life to fractions that are of limited purity, of doubtful value, and potentially contaminated with hazardous substances. Since the introduction of the ELV Directive much has been tested and obvious progress has been made. The challenge is now to select the best methods realising more recycling at a sustainable cost.

Conclusions

The concept of mandatory mechanical and thermal recycling of plastics from ASR in the EU is inspired by the European ELV Directive, since neither dismantling, nor post-separation of ASR could ever be economically viable. Positive is the mandatory control of hazardous compounds entering in a car; landfill levies or bans somewhat ameliorate the picture.

Recycling all plastics from a given make of cars would be extremely challenging. ASR plastics are indeed composed of plastics from all possible car makes, models, and vintages! Recovery of all thermoplastic materials from ASR is unfeasible, due to the large number of resins and extraneous materials present, their heterogeneity, mutual entanglement and the presence of coatings, composites, and different additives. The fractions resulting from separating ASR, mainly PP, ABS, and possibly PUR, still contain embedded extraneous matter and are of markedly lower purity; yet they can be reprocessed into undemanding automobile applications. It is still questionable whether enough of such outlets could be created, should all shredder enterprises follow the excellent example of, e.g. Galloo Plastics.

Thus, thermal recovery has a guaranteed future. Until now the adage has been to dilute and disperse: adding 1 to 10 wt% of ASR to more conventional waste fed into conventional incinerators has proved environmentally acceptable, since all emission threshold values were respected. The alternative of operating dedicated plant has even larger potential to recover more deeply the residual metals in ASR and the various valuable elements built into modern cars. Pyrolysis paves the way to enhanced metal recovery. Low-temperature gasification restricts both the oxidation and volatilisation of metals.

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