by **Andreas Mäurer and Martin Schlummer**

Good as new

Recycling plastics from WEEE and packaging wastes

Waste electrical and electronic equipment often contains huge quantities of plastics. An innovative process offers an exciting solution to the problem of recycling them. This solvent-based process purifies polymers contaminated with brominated flame retardants or other additives, and produces pure recycled polymers also from other plastic waste streams including expanded polystyrene.

The huge amounts of waste electrical and electronic equipment (WEEE) in the European Union contain large quantities of plastics. Directive 2002/96/EC[1] on waste electrical and electronic equipment specifies strict recycling quotas, which will only be met with an additional recycling circle for plastics. The plastic waste in WEEE consists mainly of polystyrene-based thermoplastics with high additive content from the equipment housings. The Fraunhofer Institute for Process Engineering and Packaging, together with its partner CreaCycle GmbH, have developed the CreaSolv® process to produce recycled polymer of high quality from these mixed and contaminated thermoplastics. This article describes the application of this new recycling process to acrylonitrile-butadiene-styrene (ABS) and polystyrene copolymer from post-consumer electronic and packaging wastes.

Different views on recycling technical polymers from WEEE

Plastic waste generally consists of polluted mixed fractions with some composites. The presence of other materials requires a greater effort to recycle technical polymers without an unacceptable deterioration in quality. High-value creation via recycling is only achieved when the recycled polymers are free of any impurities.

However, there are strong concerns against the recycling of plastics waste from WEEE. The quote of a German chief executive expresses this clearly: 'Have you ever seen such complex waste mixtures? Don't tell me that it's worth separating and cleaning the polymer for another application.' And the plastics industry lobby obviously shares this view: 'We already recycle our pure production wastes. Let's use the commingled WEEE plastic fraction for energy recovery. Let's burn it.'

This may be a logical statement from an industry group standpoint, because higher recycling rates of high quality plastics will automatically compete with existing or new production capacities. Incineration will not carry this conflict, but we need to ask what is better for the society and the environment.

The recycling plant at RUAG AG, Switzerland, deals with the mechanical recycling of WEEE. It was planned and constructed by MeWa Recycling Maschinen und Anlagenbau, Germany. PHOTO: MEWA

But the more important opinion – the neutral 'Voice of the Customer', electrical and electronics (E&E) producers will be the new driving force. They state: 'We will use recycled materials for our products as long as they fulfil the same technical specifications as virgin polymers and are constantly available at a reasonable price.'

An additional challenge is the huge discrepancy between the composition of waste plastics delivered to the recycler and the demand for high quality for recycled plastics.

The challenge

Although external pollutants can be easily removed, impurities in the plastic matrix are not yet accessible in a purification stage. Such impurities consist of:

- degradation and reaction products of additives or the polymer
- substances which have migrated into the plastic matrix during use or collection
- mixtures of various additives.

The desire for recycled plastics of high purity from WEEE plastic waste cannot be satisfied by conventional mechanical recycling methods. It requires a modified new recycling technology. The Fraunhofer Institute and the CreaCycle GmbH co-operated in solving this fascinating challenge of developing an economic process with high purification-efficiency by developing the CreaSolv process. This process allows the production of recycled plastics of high quality from mixed and contaminated plastic waste.

TABLE 1. WEEE processing quotas

	Processing quota (%)	Recycling quota (%)
Large domestic appliances	80	75
Small domestic appliances	70	50
IT and telecommunications equipment	75	65
Entertainment equipment	75	65

The main task was to clean up the polymeric macromolecules by removing substances which can negatively influence the quality, while conserving the primary morphology and functionality of the polymers. Based on well-founded current knowledge and technology regarding solvent-based recycling methods, the work was aimed at the following goals:

High-quality recycled plastics can be produced from mixed and contaminated plastic waste

- formulate a solvent-based concept
- develop technical equipment specifications
- set up and test a continuous process line with a throughput of 10 kg/hour at the Fraunhofer Institute.

The next step is to offer the process to the industry with the aim of building an industrial-scale plant.

Plastics in WEEE

The WEEE Directive defines strict quotas (see Table 1), but in respect to plastic consumption, only the top four groups of applications in Table 2 are relevant.

TABLE 2. Plastics consumption in E&E in Western Europe by sub-sector, 2000

	Percentage of total plastic consumption (1483 kilotonnes)	Percentage of total theoretical plastic waste (777 kilotonnes)
IT and telecommunications	40	34
Large household appliances	32.5	41
Consumer equipment	14.5	14
Small household appliances	10	9
Others	3	2
E&E tools	1	
Automatic dispensers	1	
Medical equipment	0.5	
Toys	0.5	

Source: *Insight into consumption and recovery in Western Europe: a material of innovation for the electrical and electronic industry 2000.* Association of Plastics Manufacturers in Europe (APME), 2001.

Bromine-containing compounds in the plastic component of WEEE

WEEE in Western Europe contains annually some 230,000 tonnes of thermoplastic materials. Half of these contain brominated compounds and the other half are bromine-free. If suitable sorting methods were available, this mixture could be separated to allow recovery of the bromine-free fraction (calculated from Bromine Science and Environmental Forum [BSEF] data for 2000). Plastic scrap containing brominated additives is currently excluded from material recovery because of the risk of exceeding mandatory threshold values for toxic polybrominated dibenzo-p-dioxins (PBDDs) and polybrominated furans (PBDFs) in the recycled plastics.

After PBB and PBDE had been recognized as a possible source of PBDDs and PBDFs,[2] producers of electronic equipment started to phase out their use as flame retardants. However, the long lifetime of some electronic items creates the potential risk that present WEEE may still contain PBB and PBDE, together with PBDD/F in the parts per billion (ppb) range.[3] (See box on facing page for details of different flame retardants and their potential for PBDD/F formation.)

Apart from the technical problems, the likely existence of high PBDD/F levels in electronic waste polymers is the main issue that prevents any mechanical recycling approaches to treat this waste fraction[4] even if the available amounts of waste and the mechanical quality of the recycled plastics would support economical recycling processes.

Post-consumer plastic materials with brominated flame retardants can technically be treated for bromine recovery. According to the European Brominated Flame Retardant Industry Panel (www.ebfrip.org), approximately 10,000 tonnes/year of bromine is potentially available from WEEE. With the development of the CreaSolv process, the possibility of returning this amount of bromine into the industrial bromine cycle appears promising.[5]

Different brominated flame retardants have varied potential for PBDD/F formation. For example, the presence of the state-of-the art flame retardant, tetrabromobisphenol A (TBBP A) – in which the bridging oxygen between the aromatic ring is replaced by a blocking alkyl group – leads to the formation of only a small amount of PBDD/F. On the other hand, there is risk of quantitatively relevant amounts of PBDD/F formation when using flame retardants that are structurally similar to PBDD/F such as polybrominated diphenylether (PBDE), polybrominated biphenyls (PBB), bis-[dibromopropoxy-dibromophenyl]-propane (OBPE) or bis-(tribromo-phenoxy)-ethane (TBPE).

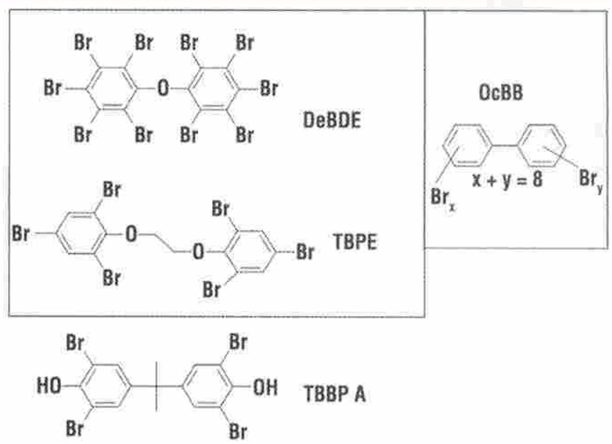


FIGURE A. **Structure of different flame retardants with varying potential for PBDD/F formation**

The CreaSolv process

Concept

The process (see Figure 1) consists of three main stages:

- dissolution of the targeted plastic in a selective solvent
- separation of pollutants/impurities from the produced polymer solution
- precipitation of the targeted plastic from the purified polymer solution.

To refine the plastic, the first step is to dissolve the polymer matrix in a suitable solvent. Factors such as the polarity of the solvent, non-hazardous classification, pH value, temperature, etc. must be clearly defined for this stage of the process. Once dissolved, the plastic polymer exists in the form of individual macromolecules. Now it is possible to eliminate undesired externally adherent materials and, unlike methods employing melt technologies, remove pollutants from within the original plastic matrix.

These interfering substances can be categorized into three groups according to their interfering effects:

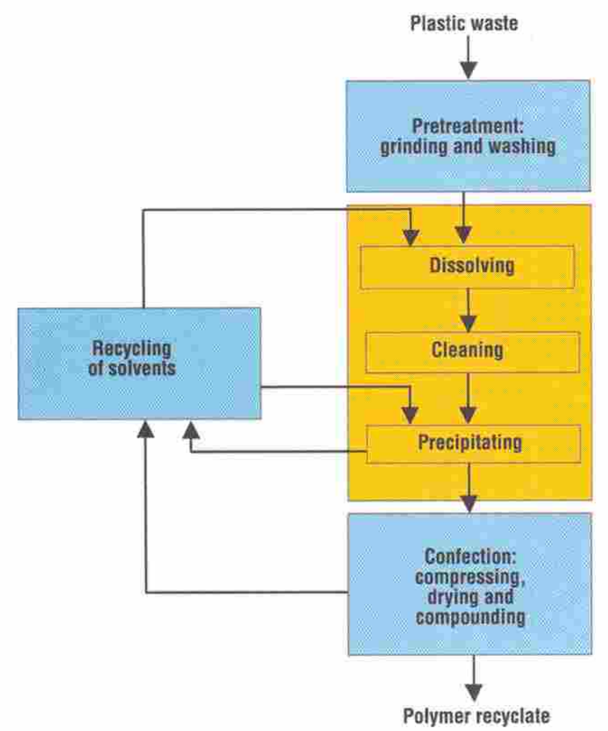
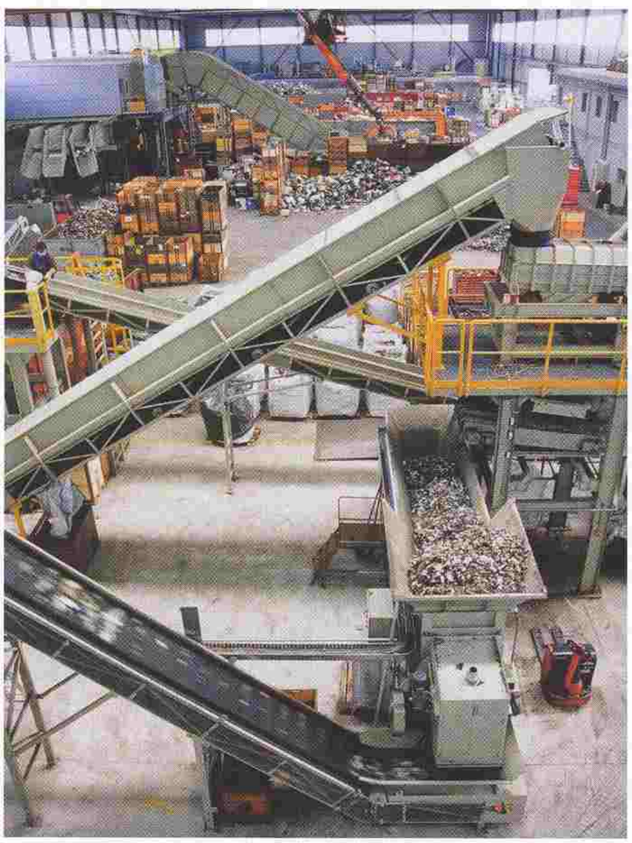


FIGURE 1. **Recycling of polymers from electronic waste with the CreaSolv process**



At the RUAG plant, the feed material goes through an input control stage at which pre-sorting takes place. PHOTO: MEWA

- those with toxicological effects (polyaromatic hydrocarbons, polychlorinated aromatics, heavy metals)
- those causing interference to processing (foreign materials, low molecular polymer fractions, radicals, cross-linking agents causing changes in rheological properties)
- those causing optical effects (coloured materials, oxidants)

After the dissolution step, interfering substances are removed from the solution by a combination of separation methods. The precipitation step is then carried out on a purified polymer solution.

If necessary, the three main stages are supplemented by a preliminary grinding and mechanical separation as well as by final processing for mechanical concentration, drying and compounding.

Development

For the process development, stepwise process engineering was combined with analytical evaluation of the optimization steps.

Analytical characterization of waste input

Spectroscopic (infrared), chromatographic (both gas and liquid chromatography with mass spectrometry) and thermoanalytical methods (differential scanning calorimetry) were successfully used for the characterization of both the waste material and the product-samples (pellets, test bars, etc.).

Table 3 shows the results from the material screening of several TV sets and PC monitor housings, and highlights the dominance of high impact polystyrene (PS) and ABS in these waste fractions. Analysis has shown that brominated flame retardants, which have been shown to react to PBDD/F during extrusion (DeBDE, OcBDE and TBPE; see Figure A) account for over 50% of all input materials tested so far, indicating the increased caution with which these waste materials should be handled.

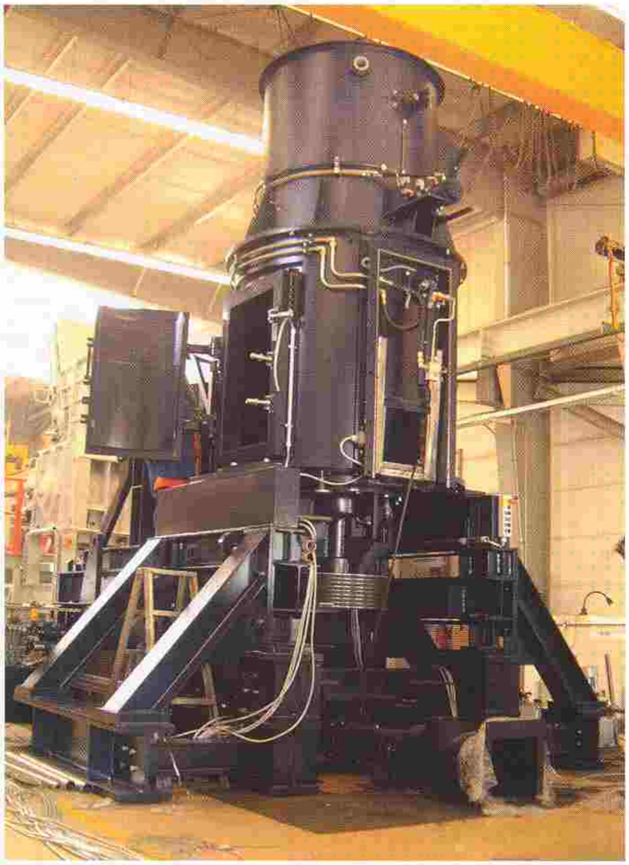
TABLE 3. Material screening of TV set and monitor housings

	Percentage
High-impact polystyrene (HIPS)	34
Acrylonitrile-butadiene-styrene (ABS, without PVC)	24
Polyphenylene oxide/polystyrene (PPO/PS)	24
ABS/polyvinyl chloride (ABS, with PVC)	9
ABS/polycarbonate (ABS/PC)	9

Pilot plant and sample production

The analytical information about the waste input and the mid-term samples allowed the process steps to be optimized and finally led to the implementation of our first small pilot plant, which had a throughput of 10 kg/hour.

Due to the high purification potential of this recycling method, the product specifications correspond well with those of virgin ABS and it is possible to use the recycled products in an array of applications (including their original use).



Machines such as this heavy-duty Universal-Querstromzerspaner (UNI-CUT QZ 1400) at the RUAG plant are used for dismantling composite materials. Interestingly, the QZ machines operate without cutting or crushing tools: materials are opened up as rotational acceleration forces them to impact onto each other, producing various output sizes depending on material properties.
PHOTO: MEWA

Our analytical data proved that the process had achieved high elimination rates (e.g. above 90%) for heavy metals, flame retardants, PBDDs and PBDFs (see Figure 2). The recycled ABS complies with all relevant threshold values as regulated by German law.

Scaling up the process

In order to scale up the process to an industrial level and to adjust the single process steps to the polymer types, present in the plastic fractions of electronic waste, we carried out the following projects:

- screening of typical waste fractions for polymer types, levels of PBDD/F and levels of flame retardants
- process optimization to handle the main polymer types and to prepare mass balances for brominated flame retardants and PBDD/Fs
- tests of different toxic waste treatment approaches to decontaminate the separated waste fraction containing brominated flame retardants and PBDD/Fs
- development of analytical tools for efficient and inexpensive quality assurance.

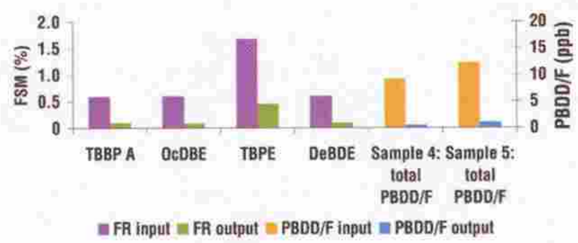


FIGURE 2. Pilot plant achieves high reduction rates for undesired flame retardant (FR) additives and PBDD/F

The small pilot plant at the Fraunhofer Institute has a throughput of 10 kg/hour

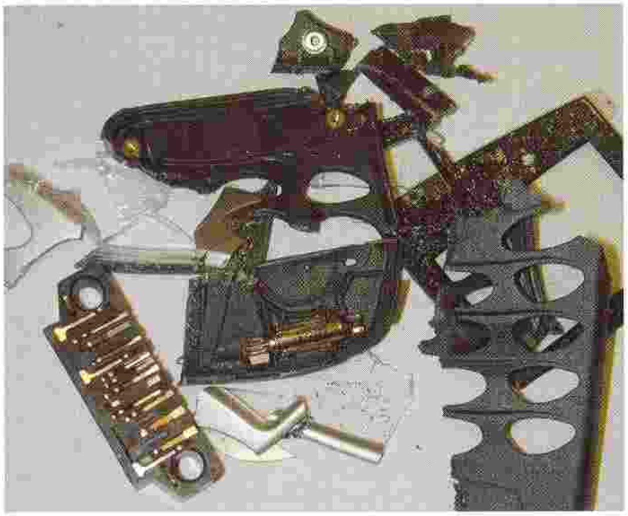
The results from these four projects provided the necessary process engineering data for a scale-up to demonstration plant quantities (500 tonnes/year). The energy demand and the conversion costs of an industrial plant (4000 tonnes/year) were calculated based on measured mass balances and energy flows. These results are now available to be incorporated into a business plan.

Multi-purpose process

The process is applicable to different polymers (PVC, PC and PS) and copolymers such as polyacrylonitrile-butadiene-styrene (ABS), polyacrylate, polymethacrylate (PMMA), polyethylene terephthalate (PET) and polyvinylbutyral (PVB). It can separate and process polybrominated additives and separate plasticizers from plastic materials.

PVC recyclates from soft PVC floor coverings

To demonstrate the technical flexibility of the pilot plant, PVC was recycled from soft PVC floor covering. Test bars were produced to evaluate their mechanical and application data. These data corresponded well with those for original PVC and it is possible to use the recycled PVC in an array of applications, including their original use. The analytical data revealed elimination rates for heavy metals, PBDDs and PBDFs of greater than 97%. The recycled PVC also complies with all relevant thresholds values regulated by German law.



ABOVE Even as mobile phones become smaller and smaller, serious concerns remain about the end-of-life management of these devices due to small amounts of many different materials **BELOW** Recycled polystyrene from this solvent-based process is clean and pure when it leaves the initial production line, regardless of whether it came from EPS packagings for TVs or from insulating materials for the construction industry

Recycling of PVB

A further proven example of the reprocessing of plastic-containing waste using the process is the recycling of PVB scrap from automotive vehicle windscreens. The removal of the various PVB plasticizers is essential if the recyclate is to be used for products with defined properties. PVB film producers and raw material suppliers rejected plasticizer separation at the beginning of the 1990 for economical reasons. Today the process offers the possibility of recovering recycled PVB of high quality, which has a low plasticizer content, to be used in the original application.

The process offers a convenient solution for recovering recyclable materials from mobile phones

Economic recycling of mobile phones

The mobile phone sector is one of the fastest growing markets in the IT business. But even as mobile phones become smaller and smaller, serious concerns remain about the end-of-life management of these devices due to small amounts of many different materials. The dismantling and recovery of recyclable materials from mobile phones is both time-consuming and expensive, and the value of the recovered secondary raw materials is low. However, the WEEE Directive prescribes challenging high recycling quotas for mobile phones, thus forcing recyclers of post-consumer electronic equipment to find economical and ecologically feasible solutions for mobile phone treatment after use.

An adaptation of the CreaSolv process offers a convenient solution to this problem. This solution is based on the fact that a mobile phone housing is made of a soluble polymeric material (ABS/PC) and that most mobile phone parts are directly fixed to the housing. The treatment of post-consumer mobile phones with a suitable organic solvent produces two fractions:

- a mixture of separated non-housing materials, which can be treated by established metal recovery processes
- a polymer solution from which recycled polymers of high quality can be obtained using the CreaSolv process.

The solvent used is recycled within the process and dismantling rates of 1 tonne/day can easily be achieved.

Packaging waste

In November 2003, a consortium of 12 partners launched the 'EPS–Loop' project. This project aims at using the CreaSolv process to develop and optimize a recycling process for expanded polystyrene (EPS) packaging waste. A concept for a cost-efficient transport of EPS waste will also be developed.

Interim results show that the CreaSolv polystyrene approach achieves a volume reduction factor 3–4 times higher than one can obtain with grinding and compressing. Both ground and compressed EPS readily dissolve in the CreaSolv solvent, while other plastics or impurities do not dissolve and can therefore be easily separated. In addition, the recovered polystyrene is easier to handle, and transport in solution is more cost-effective.

Using the CreaSolv process to recycle a collected EPS volume of 1 million m^3 with an average density of 6.5 kg/m^3 offers a significant potential for cost savings. The EPS volume reduction reduces the necessary transport movements by a factor of 10. The process also offers the additional benefits of lower carbon dioxide emissions and greatly reduces road traffic movements.

The process is a major step forward in the quality of the recycled polystyrene product. Recycled polystyrene from this solvent-based process is as clean and pure as if it leaves the initial production line – regardless of whether it came from EPS packagings for TVs or from insulating materials for the construction industry.

Conclusion

The CreaSolv process allows the manufacture of recycled polymers of high quality with defined properties from plastic waste containing pollutants, polymer mixtures and composites. The recycled plastics have a high value because they can be used instead of virgin materials.

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Interested companies are invited to contact the Fraunhofer Institute IVV and can join the development team through a common business plan. We are looking for industrial partners, interested in the CreaSolv® process. Expert support is available based on several years development in the field of solvent-based plastic recycling. A step-wise development offers improved assurance, so that plans can be turned into reality with minimal development costs.

Volume reduction of polystyrene is 3–4 times higher than with grinding and compressing

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