Chapter

Retreatment of Polymer Wastes by Disintegrator Milling

Priit Kulu and Dmitri Goljandin

Abstract

Global introduction of waste utilization techniques to the polymer market is currently not fully developed but has enormous potential. Before reintegration of used material into a new product, it normally requires grinding, that is shredding, crushing, or milling. In traditional grinders, the generated stresses in the material to be ground are equal to or less than the strength of the material. If by traditional methods, the stresses generated are compressive + shift, so by milling based on collision are tension + shift. Due to the high stress-material strength ratio at collision, it is possible to crush not only brittle materials but also ductile materials. This process allows easily combining the grinding of composite materials with their separation into individual constituents. In the current study, the mechanical recycling of the following groups of polymer materials was studied: pure brittle and soft polymers (PMMA, HDPE and IER), blends of plastics (ABS+PMMA, PC + ABS), reinforced plastics (PMMA+GFP); elastomers (rubber and tyres), and printed circuit boards (PCB).

Keywords: mechanical recycling, disintegrator mill, selective milling, separation, plastic scrap, composite plastic waste, polymer wastes, plastic powder, electronic wastes, crumb rubber, tyres recycling, electronic wastes, printed circuit boards

1. Introduction

Plastics are inexpensive, easy to mold, and lightweight. These and many other advantages make them very popular candidates for commercial applications. In many areas, they have suppressed traditional materials. However, the problem on recycling still is a major challenge [1, 2]. There are both technological and economic issues that restrain progress in this field. Different types on recycling, primary, secondary, chemical, and biological recycling are discussed with related issues [3, 4].

Global introduction of waste utilization techniques to polymer market is currently not fully developed, but has an enormous potential [5]. Before reintegration of used material into a new product, it normally requires grinding, that is shredding, crushing, or milling. These processes make the material more homogeneous and easier to blend with additives. In traditional grinders like jaw crushers, roller mills, vibration and ball mills the generated stresses in material to be grinded are equal to or less than strength of material. If by traditional methods, the stresses generated are compressive + shift, so by milling based on collision are tension + shift.

Due to the high stress-material strength ratio at collision, it is possible to crush not only brittle materials but also ductile materials.

In current study the mechanical recycling of following groups of polymer materials were studied:

- pure plastics as examples of brittle (ABS and PMMA) and soft materials (HDPE);
- blends of ABS + PMMA, and PC + ABS plastics;
- glassfibre reinforced plastics, (PMMA + GFP);
- compounded plastics - circuit boards, (PCB);
- pure rubber and tyres.

2. Bases of disintegrator milling

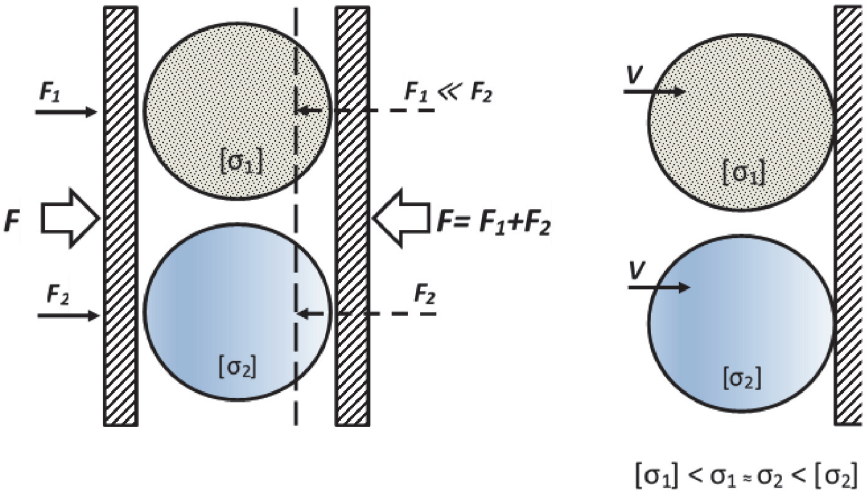
2.1 Theoretical aspects of impact milling and separation in disintegrator

In traditional grinding using traditional jaw crushers, ball mills etc., the grindable material remains between the two grinding bodies (**Figure 1a**) and will be crush by pressure or shear stresses [6]. The generated stresses in the particle are equal to or less than the strength of the material.

At traditional grinding methods, will take place uniform size reduction. The selectivity of process is low. It means that by bring weak and strong particles in the active grinding zone both, the strong and weak pieces will be broken up.

In the disintegrator, grinding occurs when piece collides with the moving working blades. This collision is an unlimited hit with some speed. The speeds used in the disintegrator range from 30 to 200 m/s [7].

At the moment of impact of the moving piece on the grinding body, an intensive wave of compression stress begins to propagate in the piece from the area of contact with the working body. The piece remains as a whole, intact during the propagation of the compression wave, until it reaches the opposite side of the piece, where it is reflected as a stretching wave of the same intensity from which the destruction of the piece to particles occurs [8]. The stresses arising in the material in this case (**Figure 1b**) exceed its strength by an order of magnitude.



(a) (b)

Figure 1.
Stresses in material: (a) Traditional milling; (b) Impact milling (image based on [8]).

Only by impact milling in the disintegrator, i.e. at a certain speed of impact, allows to implement a mode in which a less robust piece will collapse, and a more robust (or defect - free) one will remain intact, and selective size reduction occurs. The degree of selectivity of grinding depends on the materials properties (density, strength) and on the defectivity of the material crystallographic structure. It enables to grind selectively with disintegrator mills and is suitable for processing multicomponent materials and components, like polymer composites, blends and PCBs.

2.2 Developed disintegrators, separators and disintegrator milling systems

The DS series of multi-functional disintegrators were developed at Tallinn University of Technology (TalTech) for the processing of various materials, including polymer materials and plastic scrap [9]. The disintegrators of this series include:

- Special multipurpose disintegrator milling system – the laboratory disintegrator DSL-175 with a combined inertial-centrifugal classifier;
- Semi-industrial multi-functional disintegrator milling system DSL-115 of direct, separative and selective modes of treatment;
- DSA-series industrial disintegrators (DSA-2, DSA-158);
- Device SD-15 – an experimental module for studying the processes of the high-speed grinding of complexly deformed rubber composite;
- Device SD-25 – a module for simulating the processing of solid car tyres into rubber chips, with simultaneous separation of textile and metal cord at room temperature.

These systems are designed for processing various types of materials and are characterized as follows (**Table 1**):

- treats materials by collision at high rate velocities,
- operates on direct, separative or selective systems in portional or steady conditions,
- is flexible in transfer from one system to another,
- increases a material's chemical activity.

Disintegrators	Purpose	Productivity t/h
DSA-158	Precrushing	up to 1.5
DSA-2	Multistage direct milling	up to 5.8
DSL-115	Direct / Separative / Selective milling	up to 1.3 / 0.25 / 1.3
	Separators/classifiers	
Inertial (IC)	For separation of ground product	0.25[a]
Centrifugal (CC)	The same, higher sensitivity to compare with IC	0.15[a]

[a] *Set for separative milling by disintegrator DSL-115.*

Table 1.
Characteristics of used disintegrators and classifiers.

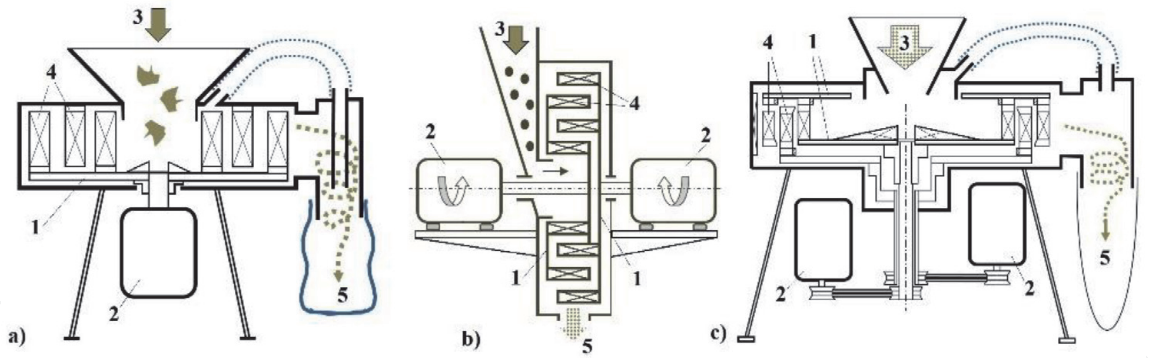


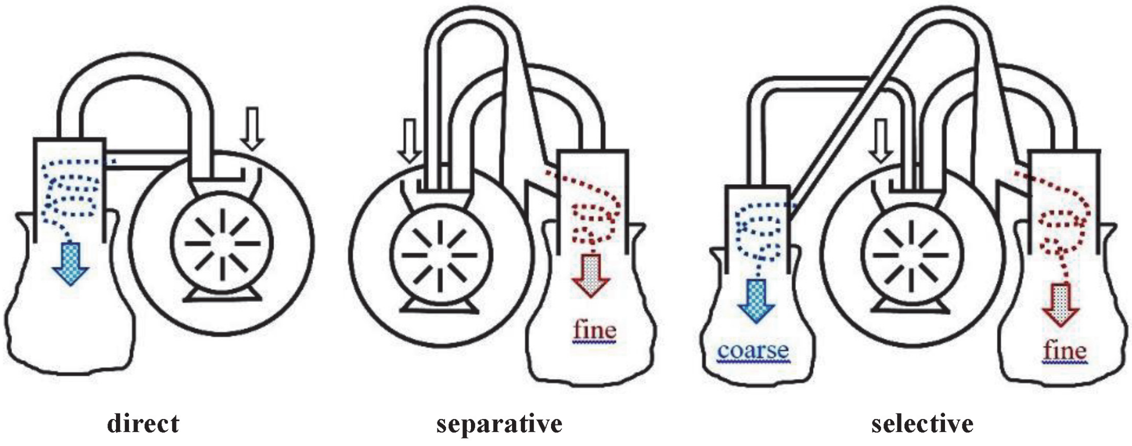
Figure 2.
Schematic representation of disintegrators DSA-2 (a), DSL-115 (b) and DSA-158 (c): Disintegrator equipment: 1 – Rotors; 2 – Electric drives; 3 – Material supply; 4 – Grinding elements; 5 – Output (authors image).

The developed disintegrators can work in the system of direct, separative (closed) or selective milling (**Figure 2**).

In the direct milling system, all the material passes through the disintegrator, undergoes a series of high-intensity impacts and flies out into the collector of the final product. Direct grinding is intended for the: testing the properties of materials, production of materials with a wide granularity, processing dry, wet or liquid materials, to combine the impact effect with other technological operations, such as mixing, homogenizing, cleaning and drying.

To obtain a product with a very fine grain, it is necessary to use the separative milling mode of disintegration, in which the crushed mass repeatedly passes the disintegrator and a classifier separates the product with the required grain size. Switching from one grinding mode to another requires only one simple operation - changing the classifier. An example of separative milling is the final operation of recycling acrylic waste – pre-crushed scrap is milled into particles of an adjustable size from 2 mm to 50 microns. The resulting powder can be effectively used as a starting material for creating new powder mixtures and for electrostatic spraying as a new coating.

Selective milling is a variant of separation in process, in which small and large particles are sent to different collectors. It allows to separate (enrich) materials with different density, strength, or get rid of microdefects in the structure of the material (**Figure 3**).



direct **separative** **selective**

Figure 3.
Modes of disintegrator milling (authors image).

The purpose of this option is:

- treating multicomponent materials by refining one component mainly while leaving the other components as intact as possible,

- industrial wastes in order to extract valuable components.

3. Disintegrator treatment of different polymer-based materials

3.1 Treatment of brittle plastics

The plates of polymethyl methacrylate (PMMA) plastic preliminarily cut into pieces with the dimensions of length 100 mm, width 100 mm and thickness 3–5 mm were reprocessed by the disintegrator milling.

The reprocessing technology of the PMMA plastic in the disintegrators consisted of three stages:

- preliminary milling by the DSA-158 disintegrator in the conditions of direct milling;

- intermediate milling for the size reduction in the DSA-2 disintegrator in the conditions of multi-stage milling;

- final milling by the DSL-115 disintegrator system applying the direct or/and separative milling conditions.

The main kinetic parameter when processing the material in disintegrators is the specific treatment energy of E_S in kWh/t, which is important both from the point of view of the milling effect (grindability) and from the economic side of the process [10]. Grindability of PMMA plastic as a function of particle size of the specific energy of treatment was analyzed.

The powder particles size after preliminary, intermediate multi-step and final millings of the PMMA plastic are briefly given in **Figure 4**.

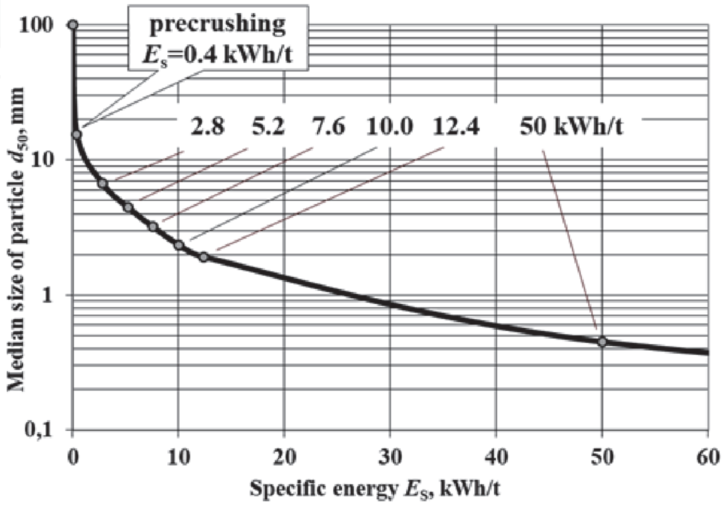


Figure 4.
Dependence of the median particle size d_{50} of the PMMA plastic powder on the specific energy of treatment E_S [9].

3.2 Treatment of ion exchange resins

Ion-exchange resins (ion exchangers – ionites, IER) are polymers of spatial three-dimensional structure and are divided into cationites and anionites.

By chemical nature, ionites are high-molecular "weight" "crosslinked materials" (polymers such as phenol-formaldehyde resins or "copolymers" of styrene and divinylbenzene), having functional groups-carriers of ion-exchange catalytic properties.

Resins that exchange positive ions are called cation - exchanging (cationites), and those that exchange negatively charged ions are called anion-exchanging (anionites).

Due to the ability to reversibly exchange their ions for an equivalent amount of other ions in solution, ionites have found wide application in various fields of technology (in hydrometallurgy for the separation and purification of rare elements, in the processing of radioactive waste, in the chemical and pharmaceutical industries, to eliminate water hardness; as catalysts for organic reactions of various types: alkylation, esterification, condensation, cyanethylation, hydrolysis, etc.)

In particular, cationides with a particle size of 1.5 mm ÷ 300 μm (**Figure 5**) are widely used as a component of water purification filters, and the remains of this material can be used as a catalyst in petrochemistry, provided that at least 90% of the material has a particle size of less than 125 μm.

The study of the possibility of obtaining a cationide powder with a dimension less than 125 μm by disintegrator grinding is the topic and task of this part. To obtain such a material, it is proposed to use a disintegrator mill with a separation mode of operation. The results of separative milling is presented in **Figure 6a, b**.

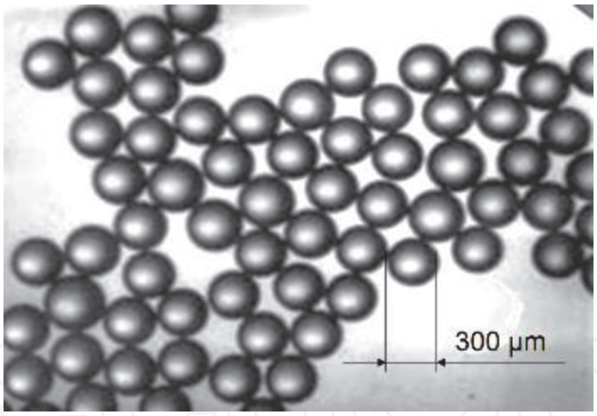


Figure 5.
Ion exchange resin (IER): Initial granules (authors image).

3.3 Treatment of soft polyethylene

High-density polyethylene (HDPE) is an example of thermoplastics.

Representing widespread polyethylene applications, HDPE offers excellent impact resistance, lightweight, low moisture absorption, and high tensile strength. HDPE is non-toxic with a high strength-to-density ratio. HDPE is used in the production of plastic bottles, corrosion-resistant piping, geomembranes, and plastic lumber.

Milling experiments to assess the grindability of HDPE (grade 277–73) were conducted in a semi-industrial disintegrator DSL-115. The parameter of grinding – the specific treatment energy E_S – was used to estimate grindability.

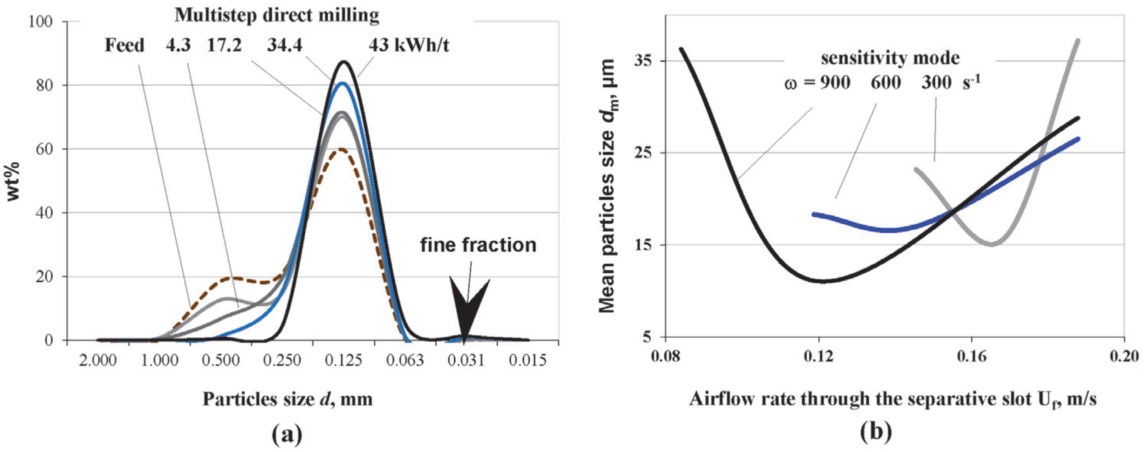


Figure 6.
Dependence of the particles size of the IER on the specific energy of treatment E_S and the grinding scheme: (a) Direct multistep milling, (b) results of separative milling with a centrifugal separator (authors image).

Initial HDPE granules with sizes from 1 to 10 mm were used as initial material for the following millings (**Figure** 7 and **Table** 2).

The studied HDPE is difficulty to process with traditional disintegrator mills due to its plasticity, because most of the kinetic energy will be consumed in the deformation and heating of the material. However, still, there is a grinding method that allows obtaining a powder of the desired size and will turn out to be much less energy-consuming than melting.

Small particles of material are formed through low cyclic fatigue fracture, in which they peel off the surface in places of repeated plastic deformations formed during impact, therefore, this process requires a large number of impact cycles. Despite this, a fine fraction <355 μm of polyethylene can be obtained using separation grinding. It has been found that the size of the product can be adjusted within the required limits.

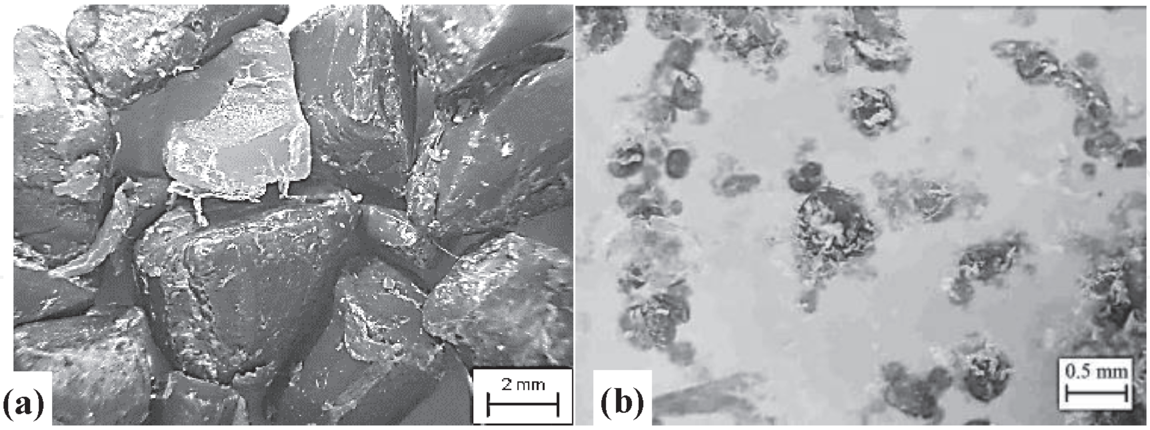


Figure 7.
HDPE particles: (a) initial; (b) disintegrator milled.

Sieves, mm	11.2	5.6	2.8	1.4	0.710	0.355	< 0.355
wt %	0	15.3	75.6	6.3	1.9	0.7	0.2

Table 2.
Granule size distribution of initial HDPE [9].

The reprocessing technology of the HDPE in disintegrators consisted of two stages:

- intermediate direct multi-step milling for the size reduction in the semi-industrial disintegrator DSL-115,
- final separative milling by the DSL-115 disintegrator with an inertial classifier.

Considering that the reason for the formation of small particles of the material is low-cycle fatigue failure, and the fact that small particles peel off from large ones in the places of repeated plastic deformations formed upon impact, it was assumed

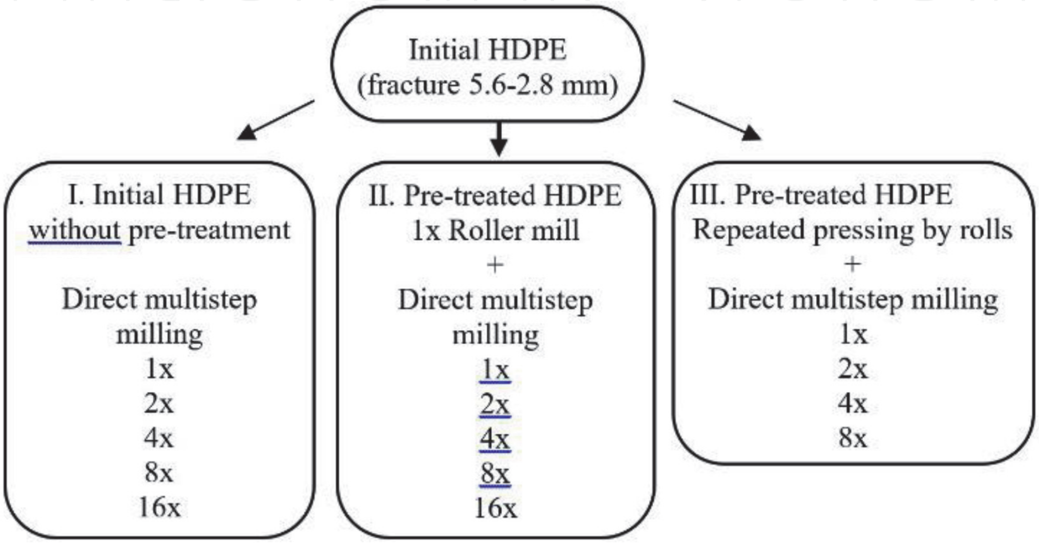


Figure 8.
Modes for treatment of HDPE by disintegrator DSL-115: I – Direct multiple grinding; II – Direct multiple grinding of single-deformed material; III – Direct multiple grinding of repeatedly deformed material (authors image).

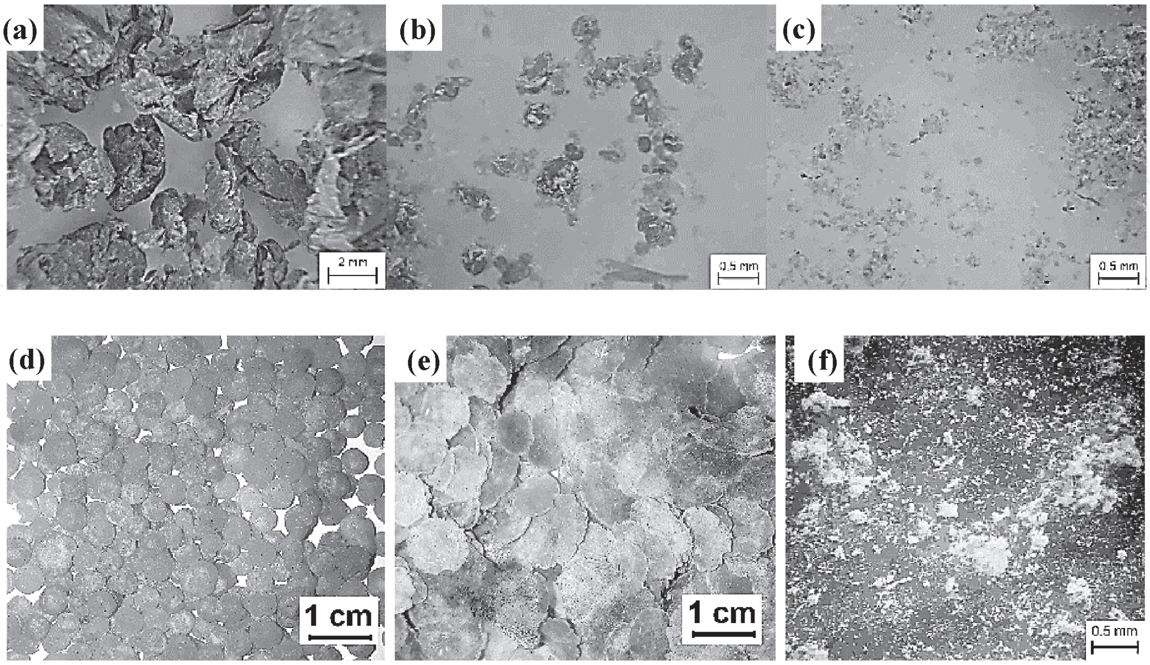


Figure 9.
Particles size and shape of HDPE after milling in DSL-115: Mode II (a – c) – Direct multiple milling at a single plastic deformation by rolls (a – 6.7 kWh/t, b – 33.5 kWh/t and c – 107.2 kWh / t; mode III (d – f) – Direct multiple milling after a multiple plastic deformation by rolls (e – Initial granules, f – Deformed by roll mill granules, g – Deformed and milled powder – 33.5 kWh/t) (authors image).

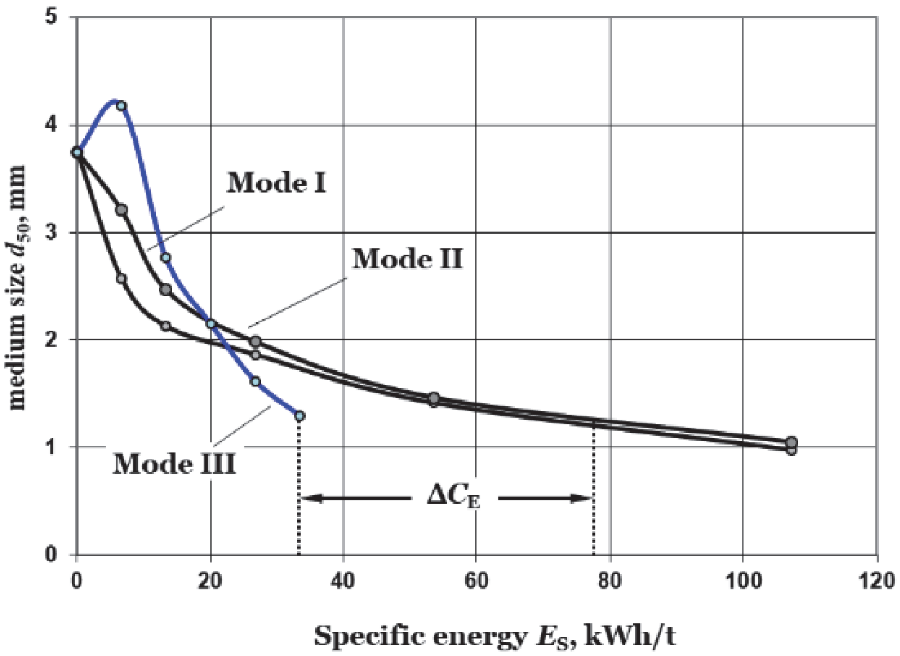
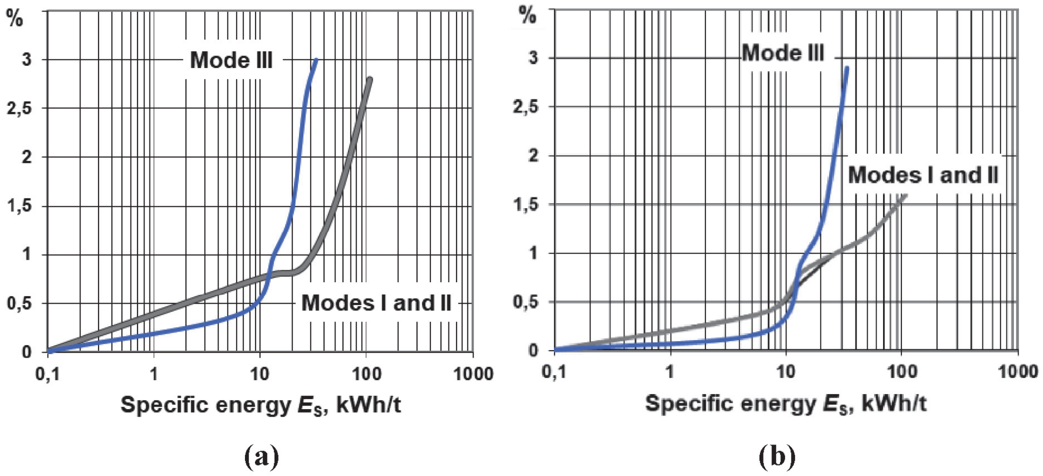


Figure 10.
Dependence of the average size of the HDPE particles on the specific energy of treatment E_S *and the grinding scheme: I, II, III – Modes of treatment (see* **Figure 8***);* ΔC_E *– The difference between the energy costs of different treatment modes (authors image).*



(a)

(b)

Figure 11.
Dependence of fine fraction of crushed HDPE at different milling modes on the specific energy of treatment E_S*: (a) fraction −355 + 180 μm; (b) fraction <180 μm (authors image).*

that a preliminarily strongly plastically deformed material could exhibit the property of easier formation of small particles. It is proposed to preliminarily pass the raw material through the rollers. The testing scheme will look like this in **Figure 8**.

Particle size and shape are given in **Figure 9**. The dependence of the particle size of multiple direct milling on the specific energy of treatment E_S is shown in **Figures 10** and **11**. Dependence of particle size at separative milling on the specific energy of treatment E_S is given in **Table 3**.

The production of powders of a given fine fraction is a process with an energy consumption of the order of $E_S = 450 \; \ldots \; 1350$ kWh/t of specific energy of treatment and total energy $E = 700 \; \ldots \; 2200$ kWh/t, depending on the separation mode.

A specific problem, arising at milling is the heating of the material during processing (on average 0.25 C° / kWh/t), which significantly reduces productivity. Possible options for solution of problem and increasing productivity may be:

Classifier type and specific energy of treatment E_S, kWh/t		Sieves, mm						d_m, mm
		1.4	0.710	0.355	0.180	0.090	< 0.090	
IC mode1	450	0	18.1	24	44	9.2	4.7	0.26
IC mode 2	690	0	5.5	12.5	30.7	31.9	19.4	0.13
CC mode 1	1350	—	0	6	32	39	23	0.11

Table 3.
HDPE powder particles distribution at disintegrator milling DSL-115 with an inertial (IC) and centrifugal (CC) classifiers.

1. Cooling of material to be treated
    a. air cooling (open-loop system of inertial or centrifugal separation, when air after separation of fine material does not return back to the working chamber) or
    b. adding water or dry ice to the material or
    c. pre-cooling of polyethylene in liquid nitrogen (perhaps the most effective option, which can notably increase the productivity of milling by an order of magnitude, since it will lead to embrittlement of a viscous particles).
2. Combination of disintegration with forming, for example, rolling between rolls (in this work the thickness reduction ratio R was 6:1). Increase in the yield of fraction −355 + 180 μm was up to 200%.
3. Effective non-impact (shear) crushing of material using compressive and friction loads. For this purposes was designed an experimental setup SD-25. The treatment is highly efficient and productive (50–75 kg/h with an energy consumption of 2–5 kWh/t); is technically easier to realize, but requires mandatory cooling (for example, with water). Results of shear crushing of HDPE are given at **Figure 12**.

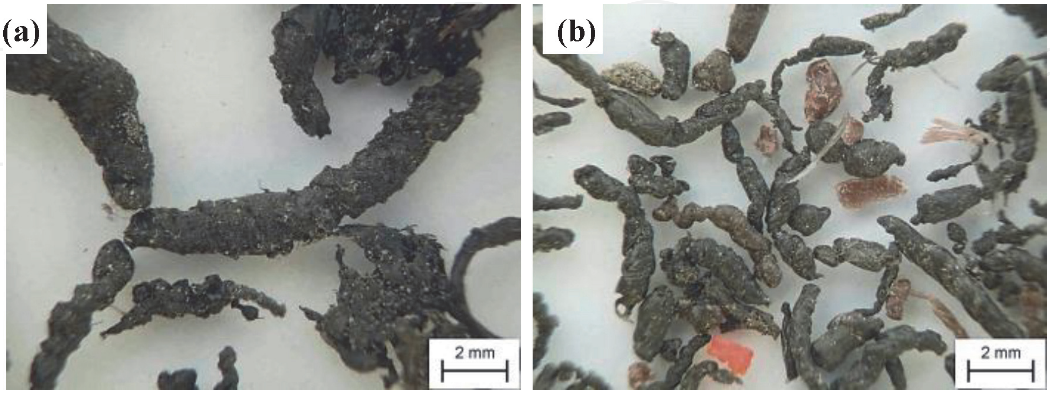


Figure 12.
Shear crushing of HDPE by the SD-25 with the gap of the cones: (a) 1 mm; (b) 0.5 mm (authors image).

3.4 Treatment of blends of plastics

Compounded plastics of the waste from electrical and electronic equipment (WEEE) subjected to recycling from dismantled personal computers (**Figure 13a**)

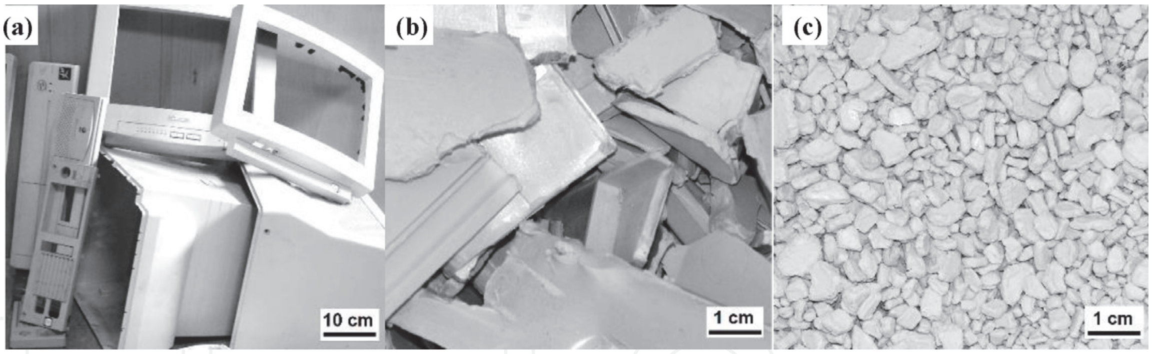


Figure 13.
Compounded plastics from dismantled personal computers: (a) initial material; (b) precrushed by disintegrator DSL-158 with the specific energy of treatment E_S = 1.4 kWh/t; (c) multistage milled by disintegrator DSA-2 with the specific energy of treatment E_S = 20.6 kWh/t [9].

Plastic	Tensile strength *R*m, MPa	Modulus of elasticity *E*, GPa	Impact strength kJ/m^2	Density *p*, kg/dm^3	Elongation after fracture *A*, %
HDPE	25	1.03	20	0.96	500
PMMA	78	3.33	12	1.2	4
ABS	56	3.40	17.5	1.17	30
PC + ABS	88	3.40	34	1.20	150

Table 4.
Physical and mechanical properties of the studied plastics of the WEEE at RT [9].

are forming from processors (4 wt%), monitors (21 wt%) and keyboards (79 wt%). Physical and mechanical properties of the compounded plastics are given in **Table 4**.

Compounded plastics polycarbonate-acrylonitrile butadiene styrene (PC + ABS) and polyphenylene ether – polystyrene (PPE + PS) from monitor housings, forming 21 wt% of the monitors, were used as the compounded plastic waste.

The compounded plastics were cut into pieces and then preliminarily milled for the feed of DSA-158 (**Figure 13b**). The multistage milling (up to 16x) of the compounded plastics was performed in the DSA-2 disintegrator (**Figure 13c**). The final milling was performed in the semi-industrial disintegrator system DSL-115 with an inertial separator. The results of the preliminary and final milling of the compounded plastics from different parts of computers are given in **Table 5**. The photos of different precrushed and milled compounded plastics are given in **Figure 13b** and **c**.

Regarding the physical and mechanical properties of the plastics to be milled, the tensile strength and impact strength of the ABS plastics are lower than those of the compounded plastics PC + ABS (**Table 5**). After the fracture, the elongation of the PC + ABS plastics exceeds that of the ABS plastics for five times. These PC + ABS plastics were easy to cut with the guillotine shears for feeding the DSA-158 (100–150 mm). After the precrushing in the DSA-158, it was obvious that the material has a ductile fracture mechanism. The crushed material was appropriate for the feed of the DSA-2.

The multistage preliminary milling of the compounded material was performed by the DSA-2 disintegrator in the direct milling condition. The results of the preliminary crushing and milling of the compounded plastic PC + ABS in the disintegrator are shown in **Figure 14**.

As follows from **Table 5** and **Figure 14a**, the best results of size reduction were achieved after the two-stage preliminary milling in the DSA-158 when the mean

Milling step and type of disintegrator	Precrushing	Preliminary multi-stage milling						Final separative milling
Multiplicity of milling	2	1	2	4	8	12	16	1
	Median size d_{50}, mm							
PC + ABS monitors	12.0	7.5	6.2	5.3	4.1	—	3.6	2.2
ABS processors	9.9	—	—	—	4.1	3.9	3.8	2.3
ABS+ HIPS[a] keyboards	15.0	—	—	—	6.0	4.3	3.6	1.7
PS + ABS+PPE monitors	9.7	—	—	—	4.0	3.8	3.6	2.4
Specific energy of treatment E_S, kWh/t	1.4	3.8	6.2	11	20.6	30.2	39.8	67.0

[a] *High Impact Polystyrene (HIPS).*

Table 5.
Dependence of the milled compounded plastic powder particle medium size d_{50} on the disintegrator type and the specific energy of treatment E_S.

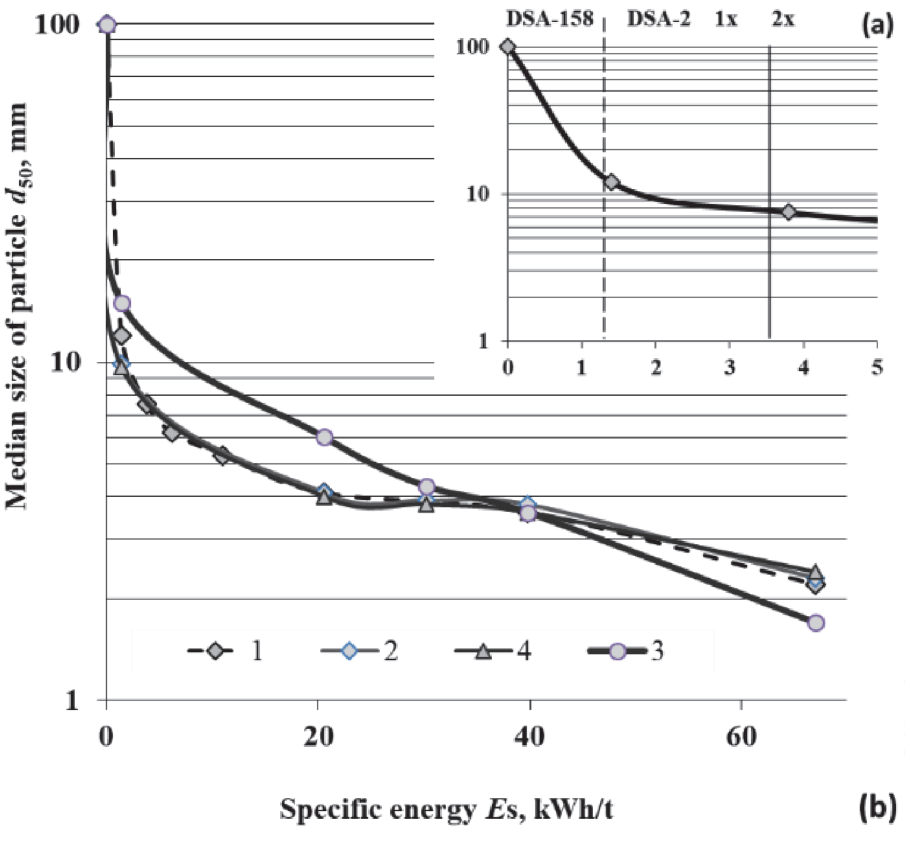


Figure 14.
Dependence of the median particle size d_{50} of compound plastic PC + ABS on the specific energy of treatment E_S after precrushing (a) and final multistage milling (b): 1 – PC + ABS monitor housings; 2 – ABS processors; 3 – ABS+HIPS keyboards after precrushing (a) and final multistage milling (b); 4 – PS + ABS+PPE monitor housings [9].

particle size was about 11 mm, and after milling in the DSA-2 – particles size was about 5 mm. During the next stages of milling in DSA-2 (from stage 4 to 8), the size reduction is less effective (from 5 mm to 4 mm only). Thus, for fracturing the particles of the ductile material, the number of collisions must be more than ten.

After eight stages of preliminary milling in the DSA-2, the particle size was in the range of 4–6 mm (**Figure 13c**). The final direct milling in the DSL-115 disintegrator reduced the particle size by 40–50% (from 3.6–3.8 to 2.4–1.7 mm).

Parameter		**Fraction, mm**			
		+ 1.25	**+ 0.63**	**+ 0.315**	**< 0.315**
Median size d_{50}, mm		1.88	0.94	0.47	0.16
Mean size	d_m, mm	2.63	0.37	0.69	2.63
	d_m^v, mm	3.60	0.46	0.74	3.60
Aspect, *AS*		1.52	1.55	1.62	1.67
Roundness, *RN*		1.32	1.43	1.53	1.57

Table 6.
The size, aspect and roundness of the milled PC + ABS powder fractions [9].

As follows from **Table 6**, the shape parameters (roundness and aspect) of powder particles milled PC + ABC compounded plastics are slightly increasing when the size of particles is decreasing.

Different types of plastics have their own dynamics of size reduction with the same applied crushing energy. If this dynamics are considerably different, it is possible to separate different types of plastics during grinding from each other in one-step.

Different particle size reduction rates allow to separate soft/ductile and hard/brittle plastics from each other in a single operation. When the viscous plastic pieces is crushed into relatively large pieces, at same time the brittle plastic pieces has time to reduce considerably in size and can be separated by a separation system (**Figures 15** and **16**).

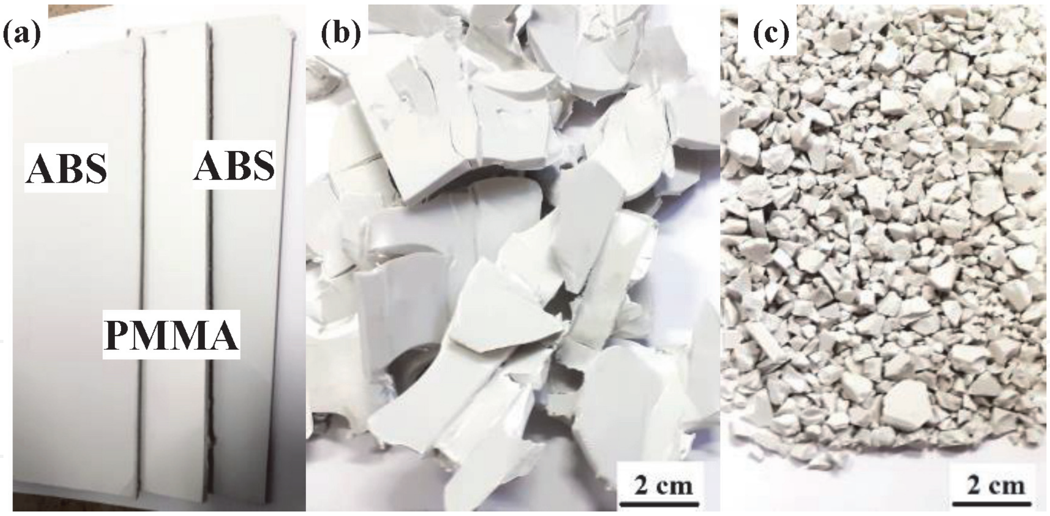


Figure 15.
Difference in particle size of the different types of plastics at the same crushing energy E_S = 4.8 kWh/t: (a) sheets of material; (b) ductile (ABS); (c) brittle (PMMA) plastics (authors image).

3.5 Treatment of glass fiber reinforced plastic composite

Glass fiber reinforced plastic (GFP) scrap consisted of acrylic plastic (PMMA) with glass fiber reinforcement in polyester resin matrix.

Polymethylmethacrylate (PMMA) sheets were formed in vacuum (shower trays and bathtubs) and reinforced with fiberglass in a polyester resin matrix. At the end of the life cycle, the product was disassembled (cut into pieces). A scrap of this composite was used for experiments (**Figure 17**).

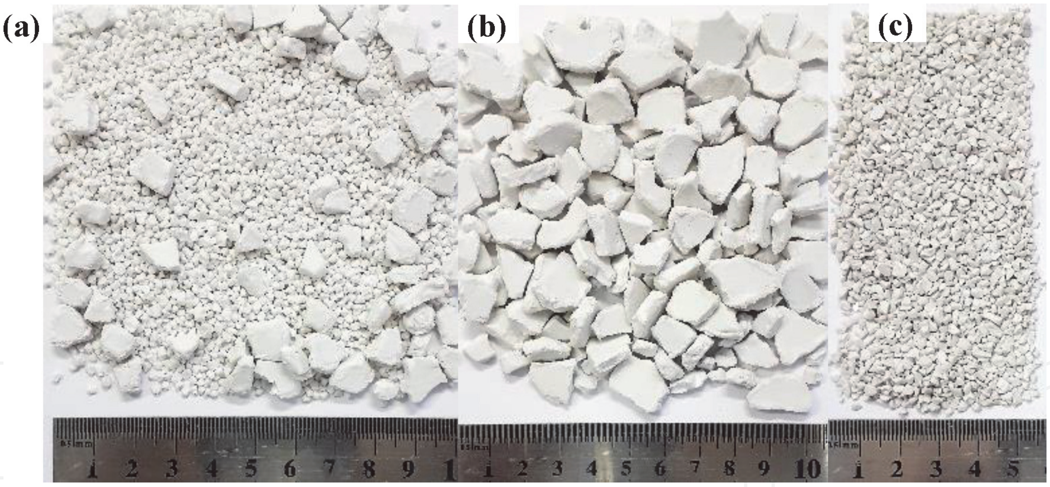


Figure 16.
The result of simultaneous grinding-separation of a mixture of ductile (ABS) and brittle (PMMA) plastics: (a) ground mixture of ABS+PMMA; (b) separated ABS; (c) separated PMMA (authors image).

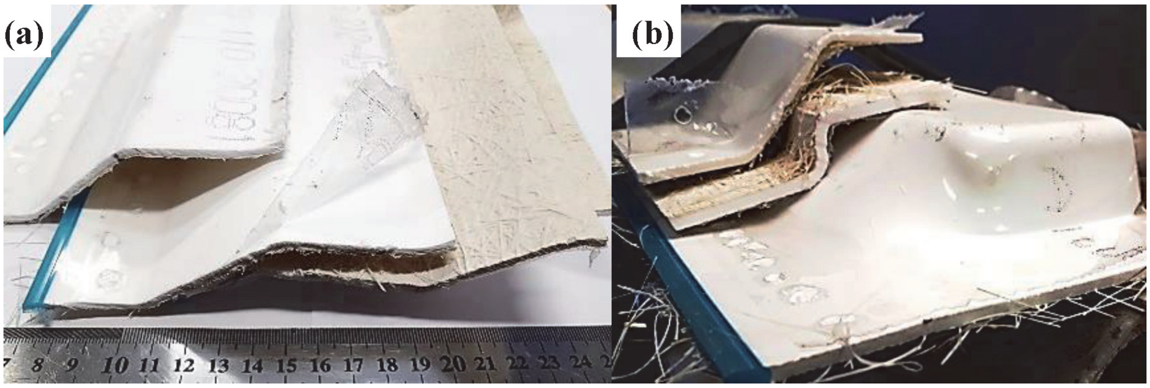


Figure 17.
Initial plastic composite scrap of PMMA+GFP (authors image).

For the milling of composite scrap, different disintegrator mills were used. To treat composite plastic scrap, the focus was on the size reduction of the acrylic plastic constituent and on the separation of the glass fiber constituent.

Disintegrator milling enables size reduction with simultaneous separation of components of low toughness. Composite plastic strips (PMMA+GFP) with dimensions of 100 × 100 × 5 mm were retreated.

The reprocessing technology of composite plastic scrap in disintegrators consisted of two steps:

1. Preliminary direct milling of reinforced acrylic strips with the disintegrator DSL-158 with following separation of glass fiber from the milled material by sieving or multi-stage milling with the semi-industrial disintegrator DSA-2. Sieve analysis were taken and the percentage of the separated glass fiber was determined;

2. Direct or separative final milling with the DSL-115 disintegrator milling system to remove glass fiber from the milled material.

3. The results of separation glass fiber and acrylic plastic are presented in **Figures 18–20**.

The results obtained during the preliminary grinding of PMMA + GFP composite plastic in disintegrator mills are shown in **Figures 18** and **20 a, b**. The particle

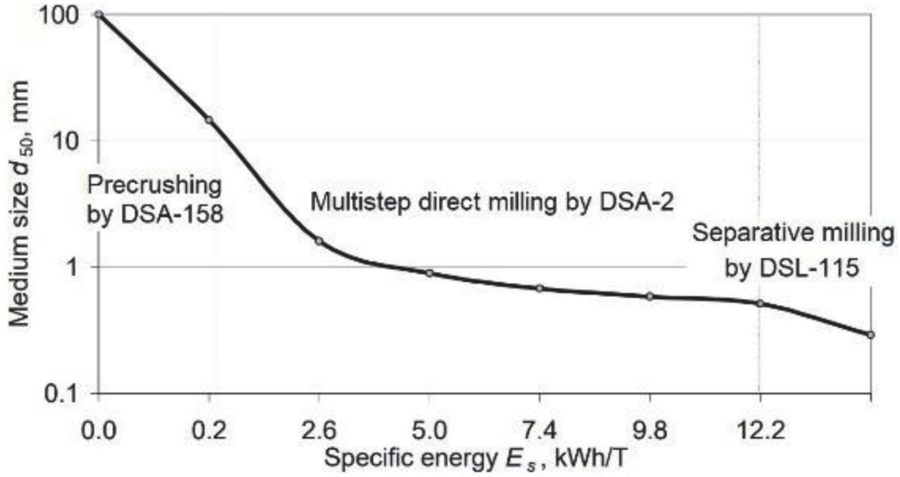


Figure 18.
Dependence of the particle size d_{50} of the milled composite plastic PMMA+GFP on the specific energy of treatment E_S [11].

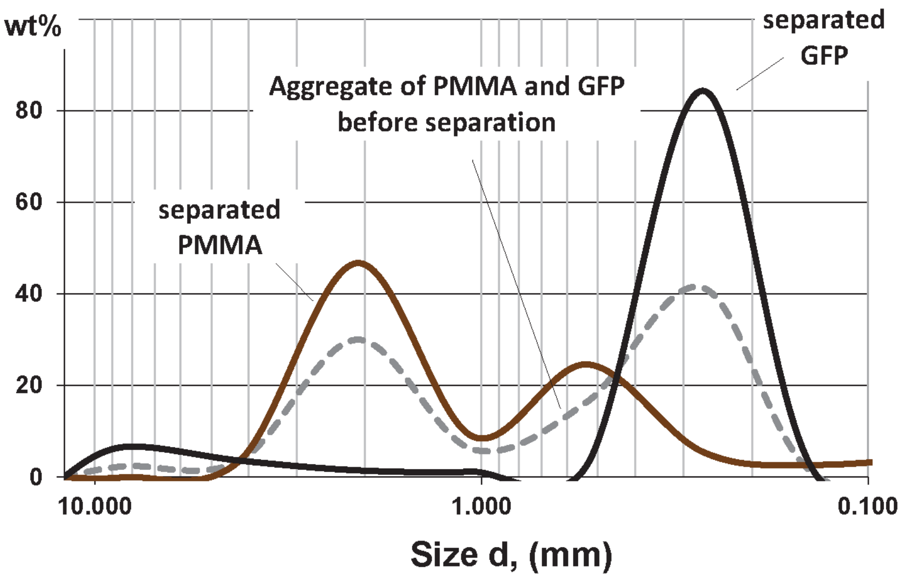


Figure 19.
Separation of glass fiber from acrylic plastic [12].

size at the outlet of the DSA-158 disintegrator was approximately 13–25 mm. The precrushed material is suitable for direct grinding in the DSA-2 disintegrator.

Before the separation, the curve of mixed plastics has two modes (**Figure 19**). After separation the acrylic plastic has two main fractions: 46% of 1.4 mm and 25% of 0.355 mm and glass fiber has one main fraction 0.180 mm, which is more than 85%. The results of separating glass fiber from scrap of composite plastic are shown in **Table 7**.

As follows from **Table 7**, the total amount of separated GFP was 45 wt%. As a result, 55% of acrylic plastic from composite plastic scrap can be reused. GFP can be reused as reinforcement in the production of polymer concrete products.

Plastic powder with a particle size of about 1–2 mm can be obtained by two-stage grinding, and 95% by weight of the glass fiber content can be separated by final selective grinding.

The recovered material can be reused in the same production process in which it was obtained. The crushed PMMA powder is applicable as a filler in the casting technology.

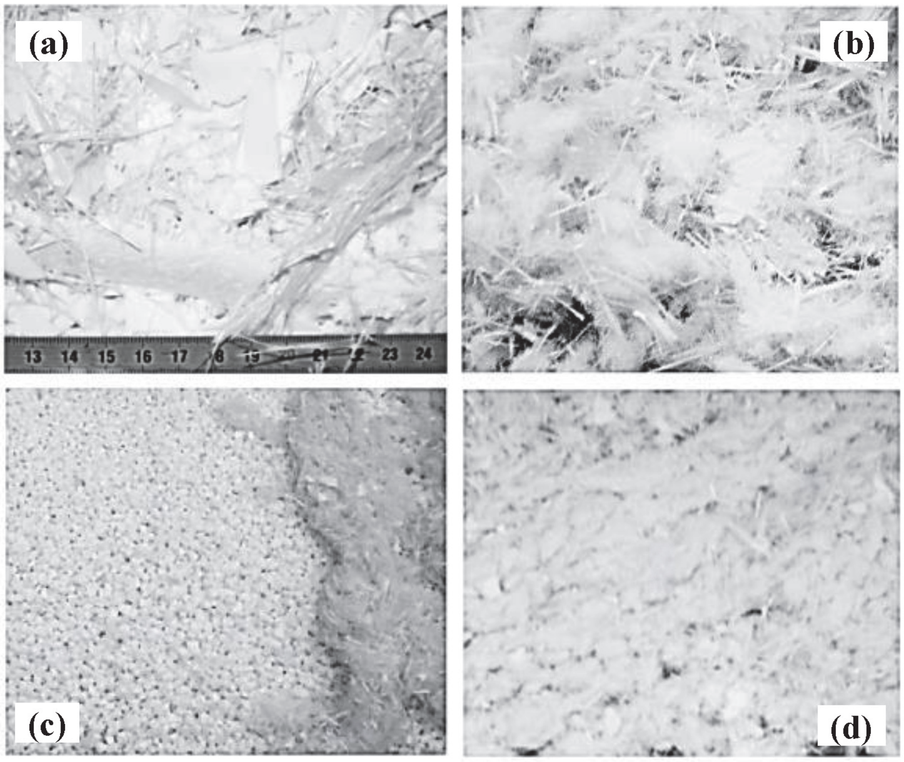


Figure 20.
The results of milling-separation of the glass fiber and acrylic plastic: (a) pre-crushed plastic scrap, (b) separated GFP large fraction, (c) finally milled mixture, (d) separated GFP fine fraction (authors image).

Milling stage	Milling device	Separation method	Separated GFP, wt.%
I	DSA-158	Sieving	16.3
	DSA-2	Sieving	12.2
II	DSL-115	Air classifying	16.5

Table 7.
Results of GFP separation during selective milling by different disintegrators [13].

3.6 Retreatment of tyres and pure rubber

Tyres can be utilized by disintegration in two ways (**Figure 21**):

- preliminary cutting of the tyres to the pieces of 50–150 mm, and size reduction to pieces of 10–30 mm by special devices, or

- direct milling of whole tyres to the rubber powder of size 1–2 mm by special units.

Collars with wires will be removed before cutting-off in both cases.

At the first stage, the pieces of tyres sized 50 to 150 mm can be used in the pyrolysis technology for oil production. The pieces sized 10 to 50 mm can be used as additional fuel in furnaces. Fine powder of fraction 1–2 mm can be used for producing asphalt-concrete of road pavement, which is most beneficial.

At the second stage, further milling of rubber assumes separation of pure rubber from textile and wire fiber. The technology of treatment pure rubber to reclaimed rubber, used instead of caoutchouc, is cost-effective. The ultra-fine rubber powder

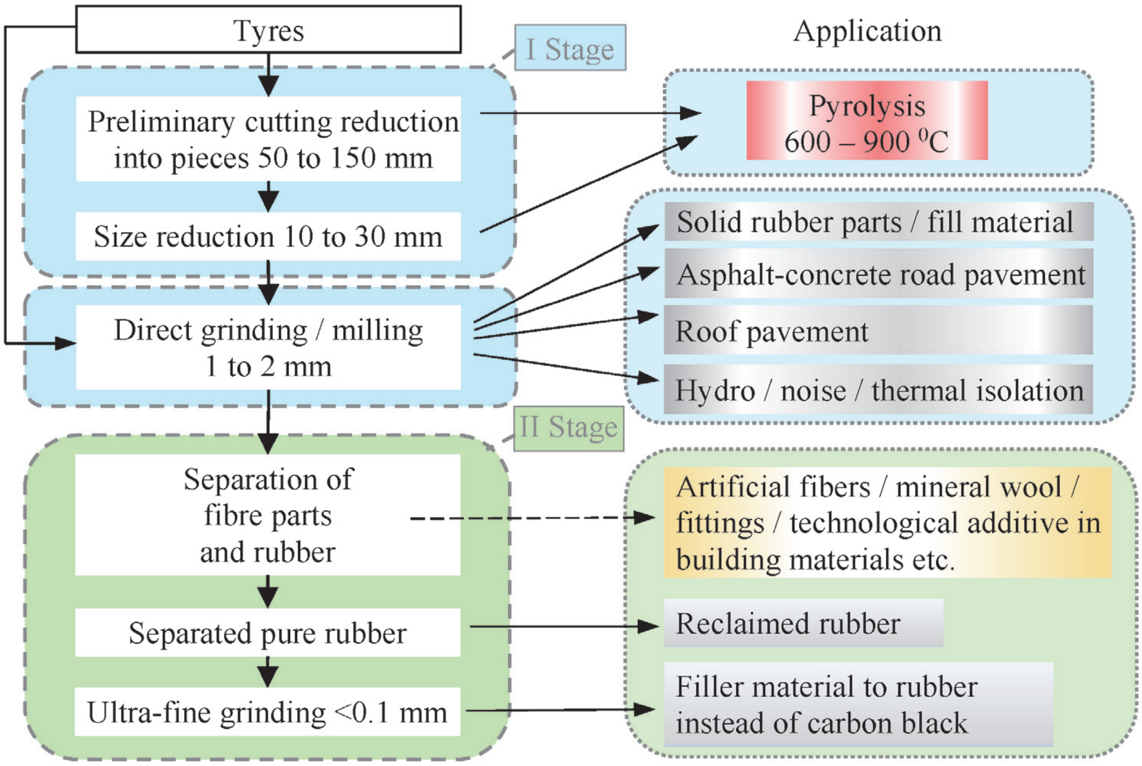


Figure 21.
Principal scheme of utilization of tyres at room temperature (authors image, based on [14]).

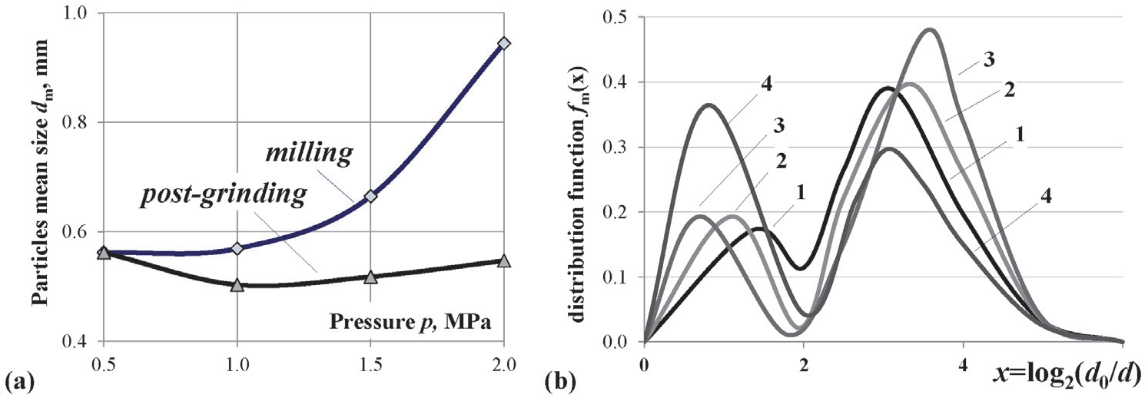


Figure 22.
Dependences of particles size on the feed pressure:(a) of tyre strips in the process of milling and post-grinding; (b) of rubber particles at different feed pressures: 1–0.5 MPa; 2–1.0 MPa; 3–1.5 MPa; 4–2.0 MPa (d_o = 5 mm) [9].

with particles less than 100 μm can be used as carbon black in the production of new rubber. However, this rubber powder is more expensive than carbon black.

Direct milling of whole tyres to the powder of 1–2 mm is more effective. In this case, the disintegrator system consists of two special devices - units for milling and post-grinding.

The granularity of the product depends on the pressure of the tyres against the milling tool. The mean size of the product after milling is shown in **Figure 22a**.

The dependence of particle mean size on the pressure is notable. Post-grinding evens the size of the particles. The granularity of the final product is shown in **Figure 22b**. It can be seen that the distribution function is two-modal.

Comparative grinding of pure rubber at normal and low temperatures was also conducted. The results are shown in **Figures 22** and **23**. The effectiveness of grindability of pure rubber at normal temperature is very low but as it follows from

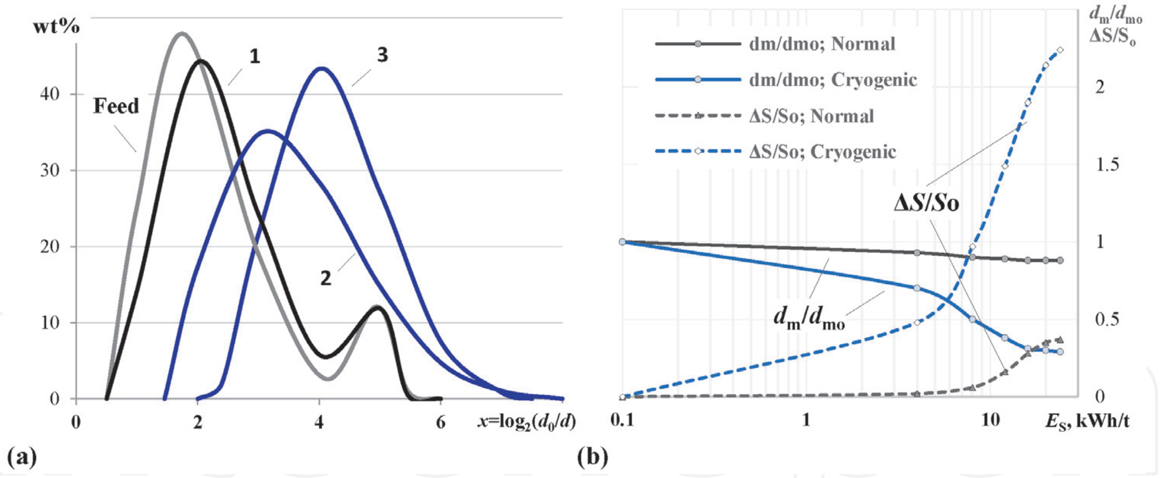


Figure 23.
Dependence of the granularity (a) and specific surface area (b) of ground rubber particles on the specific energy of treatment E_S *at normal temperature and cryogenic treatment: 1 – Milling at normal temperature with energy 25 kWh/t; 2 – Cryogenic milling with energy 8.3 kWh/t and 3–27 kWh/t* (d_o = 5 mm)*, (authors image).*

Figure 23a, the cryogenic grinding is more effective. Results of normal and low temperature grindings for comparison are grouped together graphically in **Figure 23b**.

As can be seen, the relative mean particle size d_m/d_{m0} and the relative increase in the specific surface area $\Delta S/\Delta S_0$ of rubber particles depend on the specific energy of treatment E_S. **Figure 23** shows that cryogenic grinding is more effective, particularly due to the increase in the specific surface area of rubber particles.

Highly alloyed steel wire found in car tyre collars may have its own value. To find out this value, it was necessary to study first the possibility of separating the wires.

Selective collar size reduction was achieved by using cryogenic grinding in disintegrator DS-158, during which the separation of wire, textile and rubber particles will take place (**Figure 24**).

The amount of rubber in the collars is relatively low, so the main value is probably in the metal. In addition, the amount of liquid nitrogen needed for cryogenic grinding has been determined theoretically and experimentally.

However, the economic efficiency of selective grinding has not been determined. It depends on the market value of alloy steel, amount of tyre collars to be treated and methods used to separate materials. The alternative is disposing collars to landfills, which is currently allowed.

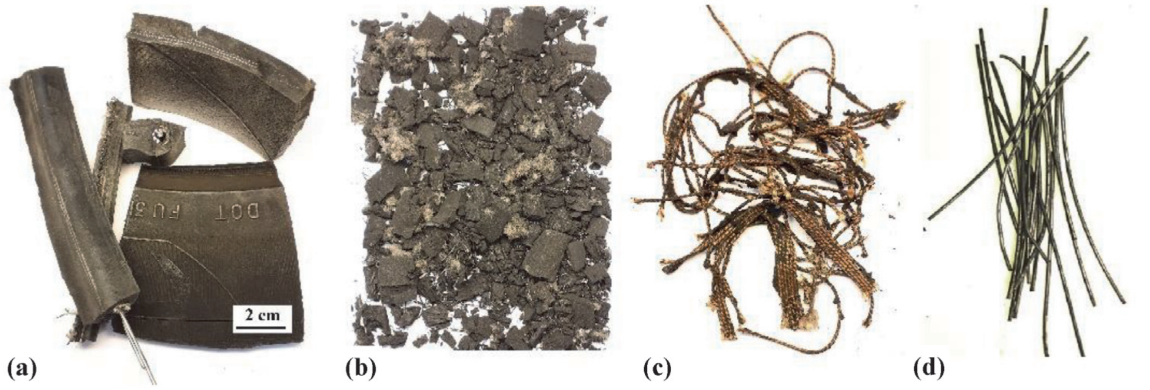


Figure 24.
Cryogenic grinding by the disintegrator DS-158: (a) initial pieces of collar; (b) ground and separated rubber; (c) textile; (d) steel wire (authors image).

Grinding of pure rubber and composites using classical disintegrator mills at normal temperatures is much (by orders of magnitude) less effective than grinding in liquid nitrogen (**Figures 22** and **23**). This is due to high elastic properties of rubber. Under impact, kinetic energy leads to strong elastic deformations of rubber pieces, which preferentially heat the material but do not destroy it.

Therefore, it is rational to use such a grinding method for the following:

- precrushing of rubber agglomerates (**Figure 25**) and
- changing in material properties.

Changing in material properties consists:

a. creation of interstitial composites (e.g., rubber-iron powder);

b. spheroidizing to reduce the surface and improve the free-flow properties of the powders (**Figure 25c**);

c. changing chemical properties under the influence of strong shear shock loads (devulcanization).

The degree of grinding is directly related to the energy consumption of the process. Thus, the feasibility of energy costs for the rubber grinding process is associated with the minimum particle size obtained during grinding. **Figure 25** shows the dependence of the average diameter of rubber particles on the specific grinding energy.

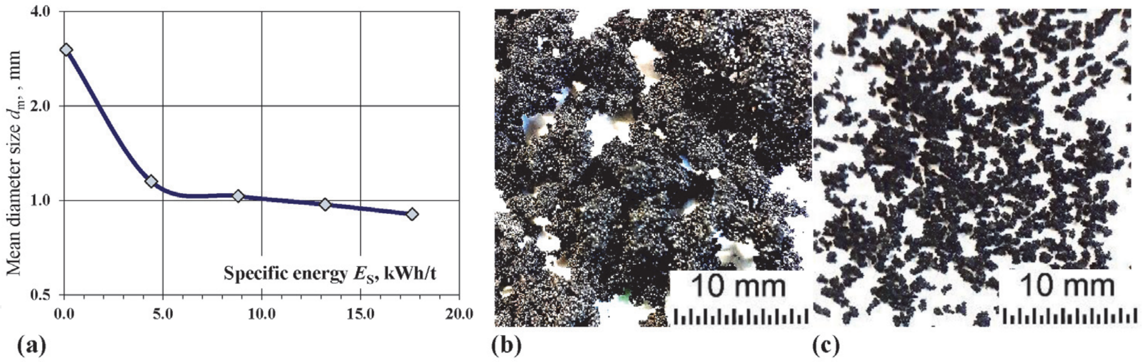


Figure 25.
Dependence of the mean diameter d_m (a) and shape (b, c) of treated at normal temperature pure rubber on the specific energy of disintegrator milling E_S: b – Pure rubber particles before and c – After milling [14].

3.7 Treatment of printed circuit boards

Disintegrator milling as a prospective recycling method to relieve metallic components of current generation of printed circuit boards (PCB) (**Figure 26**).

The PCBs stands out with one of the highest concentrations of the rare and precious metals (RPMs). Therefore, the PCBs can/should be become a sustainable source of the RPMs for future generations of technology.

Traditionally, PCBs are processed using cutting processes (shredder mills) and a combination of low intensity impacts with shear and abrasion (hammer mills).

Both methods have disadvantages. During cutting, the knife must pass through all layers of material, which consist not only of epoxy resins and glass fiber, but also of strong and ductile metals and alloys, as well as ceramics (**Figure 27a, b**). This leads to high wear on the cutting edges. The cutting process also does not effectively

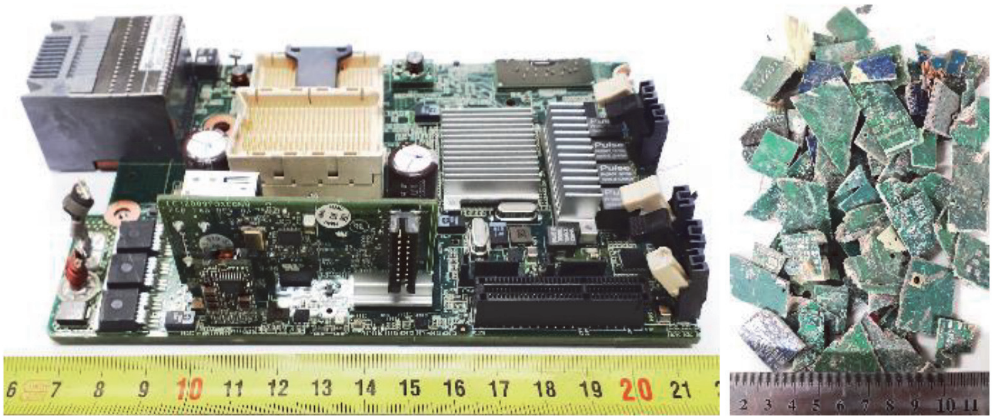


Figure 26.
Initial PCB plates (authors image).

separate into its constituent components. During abrasion and shearing, the largest and most protruding pieces are affected, and these are basically the same metal and ceramic components. As a result, a lot of extra energy is wasted on grinding media wear, unnecessary grinding of metal and ceramic components and unnecessary movement of the entire mass of material.

The high-intensity impact that is used in disintegrator mills generates high stresses in the composite materials, breaking primarily the weakest constituents of them. This should lead to the fact that in the first place the bonds between metallic fraction (MF) and non-metallic (NMF) phase will be destroyed, which will allow further classification to achieve high-quality metal concentrate (**Figure 27c, d**). Thus, due to the selectivity of the impact action, a high level of fragmentation can be achieved – this is the main factor for mechanical enrichment. Such selective disassembly is less energy consuming, relieves the rest of the components of the PCB composite from excessive grinding, and, consequently, transformation into technological emissions.

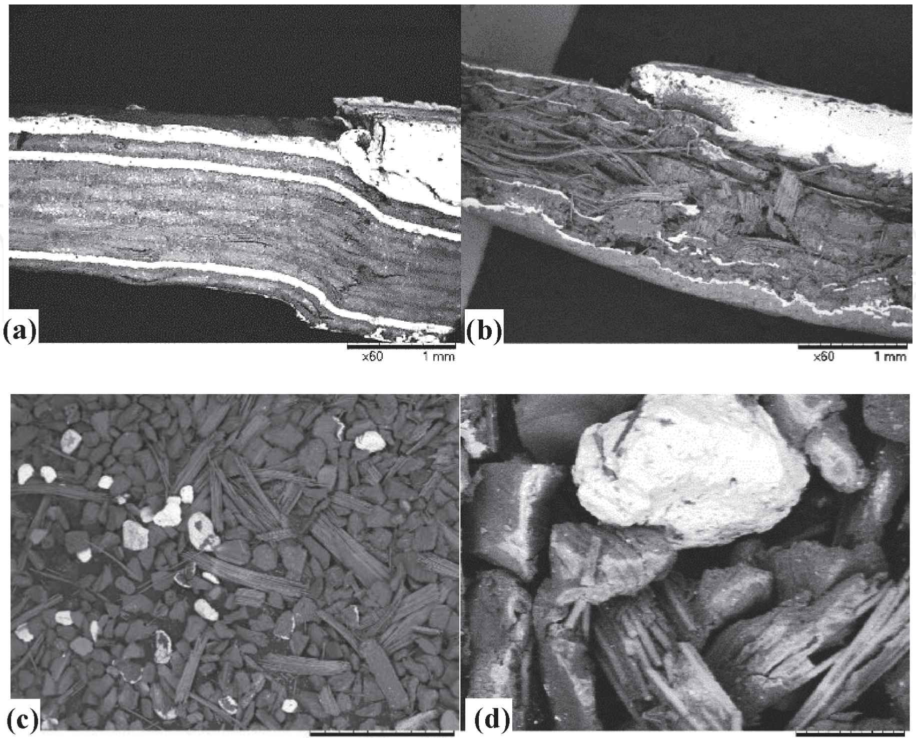


Figure 27.
Structure of the PCBs plate: (a) initial material, (b) after cutting with a shredder and (c, d) ground by the disintegrator DSA-2 (author's image).

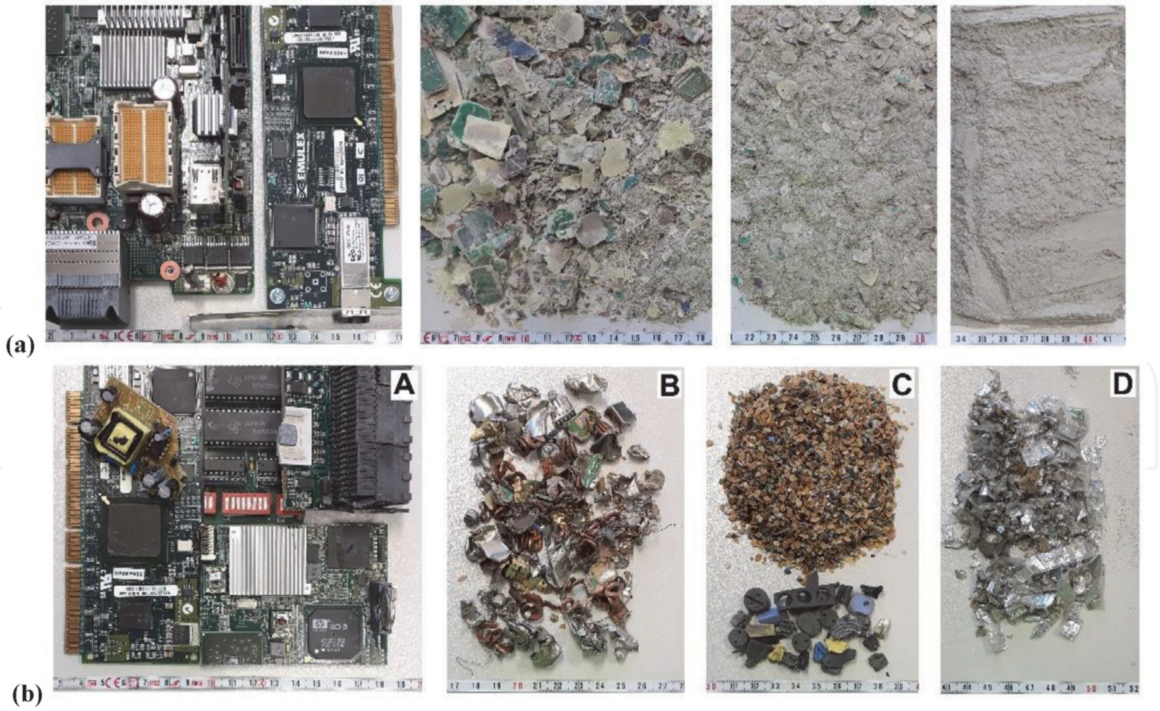


Figure 28.
Example of possible PCB disintegration degrees with existing disintegration systems (a) and PCB milling and separation stages (b): raw PCB (A), metal- (B), plastic- (C) and foil- (D) enriched components after disintegration and separation (author's image).

In this part, the integration of a high-energy impact milling for PCBs recycling comminution state-of-the-art was reviewed from the fragmentation (relieving) process perspective. Modifying PCBs wastes through delamination and fragmentation turning it to MF and NMF mixture and makes it possible to further efficient classification into enriched concentrates (**Figure 28**).

A printed circuit board is a multicomponent metal-plastic multilayer composite material that has a complex structure with brittle and plastic components. The mechanisms for reducing the particle size of plastic and brittle materials are different.

At the pre-crushing stage, large pieces of the composite plates quickly disintegrate into their component parts. The result of milling is a direct fracture. Each separated component is then crushed at its own different speed.

Then each of these components is crushed at a different speed:

- slow reduction in the size of metallic parts as a result of fatigue failure and
- rapid reduction in the size of brittle non-metallic components as a result of direct fracture.

4. Summary

Based on the study of impact milling by disintegrators different polymer materials the following conclusions may be drawn:

1. Impact milling an effective way for retreatment of polymeric materials due to the stresses initiated in materials due be ground. For impact milling DS-series of multi-functional disintegrators and milling systems operating at direct, separation and selective modes were developed.

2. Impact milling is effective way for treatment as brittle as well soft plastic wastes. The grindability of ABS, PMMA, HDPE plastics, based on different fracture mechanisms (direct fracture, low cyclic fatigue), and characteristics of ground product were clarified. Disintegrator retreatment of blends of plastics (e.g. WEEE components) enables to ground and separate plastics of different mechanical properties due to the considerably different dynamics of size reduction.

3. Retreatment of multicomponent materials (e.g. polymeric-metallic PCBs) enables to relieve the valuable metallic components, grind and separate plastic components; by tyres pretreatment (at room and cryogenic temperatures) ways/possibilities were studied and different components (rubber, textile, wire) will be separated.

4. The potential areas of application retreated plastics wastes are shown. The recovered polymer materials can be used in the same process in which they were obtained; valuable metals will be separated and enriched, and used in new PCBs production.

Acknowledgements

This work was supported by the Estonian Research Council grants PRG643 and PRG1145 and by the Environmental Investment Centre Foundation grant KIK19019.

Author details

Priit Kulu and Dmitri Goljandin*
Tallinn University of Technology (TalTech), Tallinn, Estonia

*Address all correspondence to: dmitri.goljandin@taltech.ee

References

[1] Pivnenko K, Jakobsen L, Eriksen M, Damgaard A, Astrup T. (2015). Challenges in Plastics Recycling.

[2] Hopewell J, Dvorak R, Kosior E. Plastics recycling: challenges and opportunities. Philos Trans R Soc Lond B Biol Sci. 2009;364(1526):2115-2126. DOI:10.1098/rstb.2008.0311

[3] Goodship V. Plastic recycling. Sci Prog. 2007;90(Pt 4):245-268. doi: 10.3184/003685007X228748

[4] Rudolph N, Kiesel R, Aumnate C. (2020). Understanding Plastics Recycling: Economic, Ecological, and Technical Aspects of Plastic Waste Handling. DOI: 10.3139/9781569908471.

[5] Ignatyev IA, Thielemans W, Vander Beke B. Recycling of polymers: a review. ChemSusChem. 2014;7(6):1579-1593. DOI:10.1002/cssc.201300898.

[6] Yokoyama T, Inoue Y. Chapter 10 Selection of Fine Grinding Mills. Handbook of Powder Technology, 2007, Vol. 12, 487–508. DOI: 10.1016/S0167-3785(07)12013-3

[7] Tümanok A, Kulu P. Treatment of different materials by disintegrator systems. Proc. Estonian Acad. Sci. Eng., 5/3, 1999, 222–242.

[8] Tamm B, Tymanok A. Impact grinding and disintegrators. Proc. Estonian Acad. Sci. Eng., 1996, 2, p.209–223.

[9] Goljandin D, Kulu P. Disintegrators and Disintegrator Treatment of Materials. Germany: Lap Lambert Academic Publishing; 2015. 162.

[10] Tymanok A. About the energy of material to be treated in multistep rotor milling device. Proc. Techn. Univ. Tallinn, No 393, 1976, p. 139–147.

[11] Kers J, Majak J. (2008). Modelling a new composite from a recycled GFRP. Mechanics of Composite Materials, 44 (6), 623–632. DOI: 10.1007/S11029-009-9050-4

[12] Kers J, Kulu P, Goljandin D, Mikli V. Reuse of reinforced acrylic plastic waste in new composite material development. In: Proceedings of the 5th International Conference of DAAAM Baltic “Industrial engineering - adding innovation capacity of labour force and entrepreneur”, 20-22 April 2006, Tallinn, Estonia; 2006. Ed. Küttner, R. Tallinn: Tallinn University of Technology; p. 267—271.

[13] Kers J, Kulu P, Goljandin D, Mikli V. Reprocessing technology of composite plastic scrap and properties of materials from recycled plastics. Proceedings of the Estonian Academy of Sciences Engineering, 2007, 13 (2), p. 105—117.

[14] Lapkovskis V, Mironovs V, Kasperovich A, Myadelets V, Goljandin D. Crumb Rubber as a Secondary Raw Material from Waste Rubber: A Short Review of End-of-Life Mechanical Processing Methods. Recycling; 2020 Dec 1 5(4):32. Available from: http://dx.doi.org/10.3390/recycling5040032