THESIS ON MECHANICAL AND INSTRUMENTAL ENGINEERING E31

Recycling of Composite Plastics

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Herewith I declare that this doctoral thesis is my original investigation and achievement. All ideas, major views and data from different sources by other authors are used only with a reference to the source. The thesis submitted for doctoral degree at Tallinn University of Technology has not been submitted for any degree or examination in any other university.

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List of symbols and abbreviations

A	area of the studied particle, mm^2 or elongation after fracture, %
A_o	initial cross-section area
A_u	final cross-section area
ABS	acrylonitrile-butadiene-styrene
AS	aspect ratio, ellipticity of the particle, ratio between major and minor axes of the Legendre ellipse
ASTM	American National Standards Institute
bcc	body centred cubic frame
BMC	bulk moulding compound
CRT	cathode-ray tube
d_{50}	the median diameter d_{50} (the median of the mass density function of Rosin-Rammler)
d_i	the diameter of the circle with an equal particle area
d_m	the arithmetic mean of length of number of diameters taken at 5° intervals around the center of the mass blob in ImagePro system
d_m^{v}	volume diameter is calculated on the basis of the plate shape formula of the particles
DP	dispersion, irregularity of the particle
dW	the mass increment in grams
E	modulus of elacticity, MN/mm^2
E_s	specific energy of grinding in disintegrator mill kWh/t; depends on disintegrator construction
EL	elongation, ellipticity of the particle
ELV	end of life vehicles
EPRO	European Association of Plastics Recycling and Recovery Organization
F_v	cumulative distribution function of particles volume
F_m	maximal force, N
f_m	modified Rosin-Rammler distribution function
FTIR	Fourier transform infrared spectroscopy
GF	glass fibre
GFP	glass fibre reinforced plastics
HBS	Brinell hardness
HDPE	high density polyethylene
HIPS	high impact polystyrene
HRE	Rockwell hardness
IA	image analysis
IP	irregularity and ellipticity parameter, ratio between minimum circumscribed and maximum inscribed circles of the particles cross-section
IRM	integrated resource management across industries
JIS	Japanese Industrial Standard
K	the proportionality constant for the particle shape
LDPE	low density polyethylene
m	mass, kg
MIR	modular infrared Fourier spectrometer
MPIF	metal Powder Industries Federation
MSW	municipal solid waste
NN	neural networks
OM	optical microscopy

P	perimeter of the studied particle
PE	polyethylene
PC	polycarbonate
PCB	printed circuit board
PET	polyethylene terephthalate
PMMA	polymethylmethacrylate
PS	polystyrene
PP	polypropylene
PPE	polyphenylene ether
PVC	polyvinylchloride
R_m	tensile strength N/mm^2
RN	roundness, irregularity of the particle
RNF	roundness factor, irregularity of the particle
ROHS	restriction of hazardous substances
RP	reinforced plastics
SA	sieving analysis
SF	shape factor
SMC	sheet moulding compound
SSA	specific surface area of the particle, m^2/g
TP	thermoplastics
TS	thermosets
UP	unsaturated polyester resin
V	volume, cm^3
WEEE	waste electrical and electronic equipment
x	logarithmic size of the particles
X	natural size of particles obtained in the sieve analysis
X_m	median particle diameter, the median of the mass density function of the Rosin-Rammler distribution in the sieve analysis
ε_B	elongation after breakage, %
ε_v	relative wear resistance
η	compactness of the structure
ρ	density, g/cm^3
ρ_a	apparent density, g/cm^3
ρ_t	tap density g/cm^3

INTRODUCTION

Recycling of materials are becoming increasingly important as industry response to public demands that resources be conserved and the environment be protected. Recycling of composite- and compounded plastic scrap as well as multi-materials has become an urgent issue. One of the ways of recycling plastic scrap is to produce powdered materials from technological scrap and end-of-life products.

Composites are most commonly produced by compression, injection, manual or continuous processes combining thermosetting synthetic resin (polyester, vinylester or epoxy resin) with reinforcing materials (glass, natural and carbon fibres or fabrics).

The generation of wastes takes place in each manufacturing plant. Every producer should manage their waste effectively to protect the environment from contamination.

According to the EU waste directives, the waste management hierarchy is the following: 1) prevention, 2) reuse, 3) recycling, 4) energy recovery, 5) incineration without energy recovery, 6) landfill [1]. As it follows from directives 2000/53/EC on ELV and 2002/96/EC on WEEE, producers are responsible for the environment protection during the product lifecycle [2]. Producers should form the producers` responsibility organizations to manage the collection and recycling of the post-consumer products. Consequently, there are two options to meet these requirements: firstly, to extend the product lifecycle combining durable materials with the durable design; secondly, to extend the lifecycle of materials by reducing the environmental impacts related to the manufacture and transportation of these materials.

The second aspect of it is to use it as a secondary raw material for producing sustainable products [3]. Finding a solution to the recovery of the plastic scrap from production will help to recycle the end-of-life products in the future [4]. Re-use and recycle strategies for plastic-based products can yield significant environmental benefits [5]. Plastics make an increasing part of our lifestyle (way-of-life) and those who learn properly to utilize plastics based on the design concept applied through the end-of-life recycling will gain a competitive advantage in the global marketplace [6].

The theme of this thesis was set up for the industrial interest of the Estonian manufacturing companies of bathroom equipment. According to their viewpoint, composite plastic scrap is of a low volume weight and thus has to be previously crushed to save the transportation and landfill costs.

Main objectives of the current study

1. To carry out the study on the reprocessing of problematic plastic scrap (composite plastics, compounded plastics and multi-materials) using mechanical methods, milling by collision. Recovery of the obtained secondary plastic product with optimisation of technology for the production of plastic powders with the determined properties (granularity, morphology and technological properties).

2. To develop a new filler material of the PMMA plastic powder with optimal density and also good properties of flowability for designing new products by using the technologies of numerical modelling, mechanical testing and experimental manufacturing.

3. To establish the collecting, sorting, reprocessing and recovery system of plastic wastes in Estonia; achieve the expected targets of the material recovery provided by the WEEE directive of the European Union.

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1. REVIEW OF THE LITERATURE

1.1. Plastic composite wastes

Almost 85% of the polymers produced worldwide are thermoplastics (TP). They can be divided into two broad classes, amorphous and crystalline thermoplastics, depending on their characteristic transition temperature. Over 70% of the total production of thermoplastics is formed by a large amount of low-cost commodity resins: polyethylene (PE) of different density, isotactic polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC). Next in performance and cost are acrylics, acrylonitrile-butadiene-styrene (ABS), terpolymers, polymethylmethacrylat (PMMA) and high-impact polystyrene (HIPS). Engineering plastics, such as acetals**,** polyamides, polycarbonate, polyesters, polyphenylene oxide and the blends thereof, are increasingly being used in high-performance applications [7, 8, 9, 10, 11].

Common thermosetting resins are unsaturated polyesters, phenolic resins, amino resins, urea/ formaldehyde resins, polyurethanes, epoxy resins and silicones [7, 12, 13, 14].

Many combinations of reinforcements and plastics are used by the plastic industry to affect the diversity of performance and cost characteristics. They may occur in a layered form, such as the typical thermosetting (TS) polyester-impregnated fibreglass mat, or in the form of fabric and moulding compounds, such as fibreglass or cotton-filled/TS polyester. The inline compounds are prepared by injection moulding or extruding with the short and long glass fibre (and other fibres). For an example, a chopped glass fibre can be fed into an injection moulding machine or a single to twin screw extruder where the thermo plastic (TP) is melted and bonded to the fibres providing an excellent mix. Reinforcements can significantly improve the structural characteristics of TP or TS plastics. They are available in the continuous and chopped forms of different length or they appear in the non-continuous form (whiskers, flakes, spheres, etc.) to meet the needs of different properties and/or processing methods. Fibreglass represents the major material used in reinforced plastics (RP) worldwide. The reinforcements can allow the RP materials to be tailored to the design or the design tailored to the materials**.** The reinforced fibres used in a large-scale production today are those of glass, cotton, cellulose, sisal, jute and nylon. The fibres reinforcing special properties are those of carbon, graphite, boron, aramid, whiskers and steel. The fibres of RP are primarily used for reinforcing resins by transferring stresses under the applied load from the weaker resin matrix to the much stronger fibre. Plastics provide a valuable and versatile material for matrices, but other materials, such as metals, ceramics and cements, are also used as matrices for the reinforcement of fibrous composites. For an efficient stress transfer of RP, the elongation of the fibre must be less whereas its stiffness modulus higher than those of the matrix. The stress transfer along with the all-important fibre/matrix interface can be improved by the use of sizing, binders or special coupling agents. The diameter of the fibre also plays an important part in maximising the stress transfer. Smaller diameters provide a greater surface area of the fibre per mass unit to aid the stress transfer in a given reinforcement context [15].

Based on the recent research [16-26], it is clear that all types of plastics can be recycled to some extent by one process or another. It is equally clear, however, that there is no single method, which could provide a universally workable solution. A sensible recycling policy (applied to any material) will probably involve different approaches. This leads to a general policy regarding the use of materials and involving a growing support for a cascade philosophy, which assumes that materials have a high-grade first use, followed (possibly) by a lower-grade second use, after which they may be disposed of by safe incineration with the recovery of energy, thus providing a three-fold benefit [15, 27].

Thermoplastic matrices can be re-melted and re-compounded with some possible loss of mechanical properties due to the thermal degradation of the matrix and the breakage of the fibre reinforcement into shorter lengths. Thermosets, such as GFP (the polyester resin matrix with the glass fibre reinforcement), are cross-linked and therefore cannot be re-melted, but can be ground to a fine powder, which is an effective filler in new compounds [15, 28].

If the reinforced plastic is granulated, its processability and performance when reprocessed into any other product are significantly or insignificantly reduced. Thus, it is important to evaluate the properties of the recycled material. A reduction in size and uniformity exerts a substantial influence on the quality of recycled reinforced plastics [15].

1.2. Recycling of plastic materials

After the first oil price shock of the 1970s, plastic recycling gained attention, and soon it became apparent that the definitions and nomenclature of recycling commodities did not apply adequately to the collection and recycling of plastics. Recycling refers to reprocessing and re-fabrication of the material that has been used and discarded by a consumer and would otherwise be destined for disposal as solid waste. The regrind is normally consumed (reused) within the manufacturing process and thus, it is not part of the solid waste problem [29].

An authoritative source of terminology and definitions applicable to plastic recycling is ASTM International standard D5033-00, which describes the following [15, 30]:

Primary recycling is the “processing of a scrap plastic product into a product with the characteristics similar to those of the original product”.

Secondary recycling is the “processing of a scrap plastic product into a product with the characteristics different from those of the original product”.

Tertiary recycling is the “processing of basic chemicals or fuels from segregated plastic scrap or plastic material that is part of a municipal waste stream or of any other source”.

Quaternary recycling is the “useful retrieval of the energy content of scrap plastic by its use as a fuel to produce products, such as steam, electricity, and so forth”.

Primary and secondary recycling are often referred to as mechanical recycling since the principal recycling processes involve the cleaning and separation of plastics by mechanical rather than chemical or thermal means.

Finally, there is the subject of the post-consumer and pre-consumer plastics whose definitions are included in ASTM D5033-00:

*Post-consumer plastic-”*the plastic material of a finished product that has served its independent purpose, and has been diverted or recovered from the phase of disposal, having completed its lifecycle as a consumer item [15, 30]”.

Pre-consumer plastic- “the plastic materials diverted from a waste stream following an industrial process, but excluding the reutilization of the materials, such as the rework, regrind or scrap generated in the process and capable of being reclaimed within the same process [15, 30]”.

The term *plastic scrap* is sometimes used to cover both, the post-consumer and pre-consumer plastics. The definitions above are in line with the position of recycling industry, assuming that scrap destined for recycling is not waste [15, 27, 29]”.

When plastic scrap serves the purpose of producing chemical energy or meeting the material needs of another industry, the final result is called the integrated resource management across industries (IRM). This is becoming a long-term goal in the world. The goals of IRM are resource conservation, waste minimization and progress towards the sustainable development. Figure 1.1 shows a general classification of resource management options of plastic scrap.

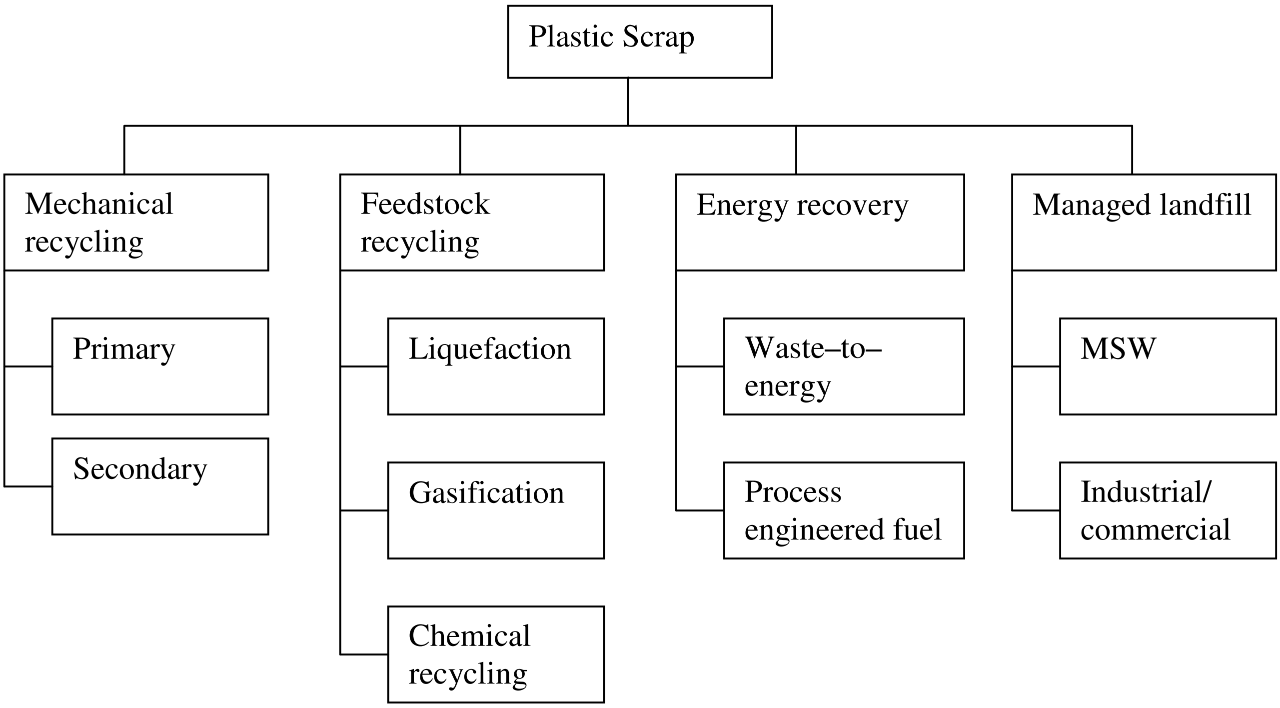


Figure 1.1 Resource management options of plastic scrap [3]

Many of the recycling technologies [31, 32, 33, 34] covered in previous Fig. 1.1 are applicable only to thermoplastics. Since thermosetting plastics cannot be readily dissolved, melted, re-compounded and reshaped like thermoplastics, different mechanical recycling strategies are required [35]. During the past decade, the following strategies have been extensively studied with a significant success: (1) the grinding of thermosetting plastics to fine powders that can be used as the fillers in new thermosetting resins or thermoplastic composites; (2) similar to (1) with an additional or alternative strategy of using a controlled-size reduction in liberating and recovering the original fillers or reinforced fibres in the thermosetting resin that may, in fact, be of a higher value than the original matrix resin. The approaches to mechanical recycling, such as these, contribute much to the chemical recycling, the recycling process of thermal feedstock, such as gasification and liquefaction and the processes of energy recovery [16, 36, 37].

1.2.1. Trends in the technologies of plastic recycling

The reprocessing and re-fabrication of post-consumer plastic scrap into useful products requires several steps: collection, handling/sorting, reclamation/cleaning and end-use fabrication [29]. The cost of separate collections is considerably higher than those of traditional mixed collections. Some plastics are easy to sort by shape, number or physical characteristics and can be reformed into the original application. Much interest has been expressed in the possibility of developing safe and reliable additive systems which could act as markers in post-consumer plastic products [38, 39, 40]. The relatively pure materials (one type of plastic resin) can also be used to make products that are not the same as originals, but are nevertheless quite valuable. The bottles made of polyethylene-terephthalate (PET) are being recycled to fibres. The bottles made of PET could also be used as the partial and complete substitutes for sand in concrete composites [41]. PVC floorings as post-consumer plastic waste can be mechanically recycled from the form in which it was recovered, without upgrading and without the addition of a new plasticizer. Gluing makes mechanical recycling less favourable thanks to a troublesome dismantling and to a high-degree contamination with the glue [37].

After sorting, the plastic materials are usually chopped into small flakes of about the size of 1-2 centimetres. The flakes are then treated by solvents and washed to remove contaminants.

The environmental pollution generated in the cleaning of these plastics is strongly influenced by their source, i.e. a non-selective collection of MSW. Therefore, the collection of wastes by the curbside system (selective collection) could reduce the need for cleaning agents and reduce the emission of some pollutants, thereby reducing the costs of the treatment units [26].

The carefully sorted recycled material is often extruded into pellets, so that the re-fabricator will be able to use the traditional processing equipment for making the finished parts [29].

Most recycled plastic types are inferior substitutes to virgin materials used as input in the original manufacturing process, and markets for virgin resins and recycled scrap plastic are somewhat different [42]. In contrast to most high-income countries where environmental legislation has usually been the driving force behind the development of the recycling industry, the recycling sector in India has developed autonomously because of the particularly low cost of labour and on account of the fairly large market for second-grade (lower-quality) products. Recycled products are available at a 20–40% lower price than the same products manufactured from virgin plastics [43].

Thermosetting materials can also be part of the “other” category in the plastics recycling system. Most people consider thermosetting plastics to be non-recyclable because they cannot be re-melted and therefore, reformed into the original products. Many materials are recycled into products other than their original form and moreover, some are recycled without ever been melted, therefore the possibility of recycling the thermosetting plastics still exists [29, 34]. This type of recycling has been demonstrated by using several different types of thermosetting materials. In these applications, thermosetting plastics are ground into powders and then used as the fillers in other products. For instance, the ground rubber tires are used successfully with polyolefin blends and the cross-linked PE is used with the virgin HDPE to make tote bins, the wheels for carts, trays and pallets. The mechanical properties of the products produced by using thermosetting fillers are only slightly less than those superior to the properties of virgin materials [29, 44].

Composite materials can also be recycled, even though the composites are often made of thermosetting resins and they comprise the added complication of the reinforcing agent. However, the parts made of sheet moulding compound (SMC) and bulk moulding compound (BMC), which are made of thermosetting polyester with the glass reinforcement and inorganic fillers, are recycled by grinding the parts and then using the resultant granules as the filler in other SMC/BMC mixes or in the polyolefin moulding compounds. The presence of this reinforcement does not seem to pose any problems, except for the fact that the grinding of cured material is somewhat more difficult than would be the case with the non-reinforced thermosets. The properties of the parts are generally the same as those of the conventional moulding compounds, provided the amount of recycle is 20% or less. The problems that have not as yet been fully resolved include the processing materials with paint or adhesive contaminants, the control of moisture in the granulated material, the segregation of various SMC/BMC formulations to provide the material consistency and development of viable SMC/BMC collection and the marketing scenarios of recycled products.

Other composite products, such as the advanced composites and non-polyester glass-reinforced plastics, are recycled by grinding the composites and then using the granules in moulding compounds. These moulding compounds are processed by compression moulding and injection moulding [15, 29, 45]. The recycling codes, which are in use worldwide and the main application areas of recycled plastics are presented in Table 1.1.

Table 1.1. The recycling codes of plastics, their applications and recycled products [37, 46]

Code	Description	Application	Recycled products
♳	Polyethylene-terephthalate **(PET, PETE).**	Plastic soft drink bottles, peanut butter, pickle, jelly and jam jars. The ovenable film and ovenable food trays.	Fibre, tote bags, clothing, film and sheet, food and beverage containers, carpets, strapping, fleece wear, luggage and bottles.
♴	High Density Polyethylene **(HDPE).**	Milk, water, juice, cosmetic, shampoo, dish and laundry detergent bottles; yogurt and margarine tubs; cereal box liners; grocery, trash and retail bags.	Liquid laundry detergent, shampoo, conditioner and motor oil bottles; pipe, buckets, crates, flower pots, garden edging, film and sheet, recycling bins, benches, dog houses, plastic lumber, floor tiles, picnic tables, fencing.
♵	Polyvinyl Chloride **(PVC)**	Clear food and non-food packaging, medical tubing, wire and cable insulation, film and sheet, construction products – pipes, fittings, sidings, floor tiles, carpet backing and window frames.	Packaging, loose-leaf binders, decking, panelling, gutters, mud flaps, film and sheet, floor tiles and mats, resilient flooring, cassette trays, electrical boxes, cables, traffic cones, garden hose.
♶	Low Density Polyethylene **(LDPE)**	Dry cleaning, bread and frozen food bags, squeezable bottles.	Shipping envelopes, garbage can liners, floor tile, furniture, film and sheet, compost bins, panelling, trash cans, landscape timber, lumber
♷	Polypropylene **(PP)**	Ketchup bottles, yogurt containers and margarine tubs, medicine bottles, bread and frozen food bags.	Automobile battery cases, signal lights, battery cables, brooms, brushes, ice scrapers, oil funnels, bicycle racks, rakes, bins, pallets, sheeting, trays.
♸	Polystyrene **(PS)**	Compact disc jackets, food service applications, grocery store meat trays, egg cartons, aspirin bottles, cups, plates, cutlery.	Thermometers, light switch plates, thermal insulation, egg cartons, vents, desk trays, rulers, license plate frames, foam packing, foam plates, cups, utensils.
♹	**Other**	Use of this code indicates that the package is made with a resin other than the six listed above, or is made of more than one resin listed above, and used in a multi-layer combination	Bottles, plastic lumber applications.

1.2.2. Reprocessing technologies

When planning to develop a recyclable product, it is important to rely on the life-cycle analysis of the product, which would cover its impact on the environment, from the production stage of raw materials to the processing/fabrication stage, through the useful stages of the product life cycle up to the methods of recovery or disposal. The majority of reprocessing and recycling technologies are developed for plastics packaging materials [47]. Different systems are applied to different materials and products, which are based on the type of plastic, its matrix, thickness, the degree of hardness, etc. These include mechanical granulators, hammer mills, energy recovery systems (the thermal reclamations of energy), the systems of chemical recycling and others. Material separation may be based on magnetic, electrostatic, density, visual, or other characteristics. A series of magnets may be used to remove ferrous metals from conveyors. While separation of ferrous and nonferrous metals is well-developed, the research into glass and plastics separation is urgently needed.

The granulators are used predominantly, they are of a different size and type; they process both, thin and thick reinforced plastics. If large and thick reinforced plastics are to be granulated, two or more granulators are used to reduce gradually their size, so that the minimum damage will occur to the fibres, they ensure that overheating is minimized, particularly with the reinforced thermosetting plastics. A cascading action occurs when granulating the thick scrap, thus the next granulator will further reduce the output from the first granulator. In case of granulating the reinforced thermoplastics or reinforced thermosets, their fibre lengths are reduced. This fibre reduction will reduce the property performance. The reduction is related to the original lengths, the degree of uniformity in cut lengths, the type of resin, etc. The actual amount of the negative influence on processing the recycled material depends on the method of granulation (such as overheating thermoplastics during the cutting action, in particular), the amount of microscopic metal from the cutting metal blades, and the size as well as the shape of granulated material (such as fine to coarse, powdered to shredded, with or without fuzz, the degree of uniformity, etc.).

In addition to chemical, physical (melting) and mechanical direct contact (ball-milling, attritor milling, hammer milling, etc.), plastics can be reprocessed by the collision method.

1.2.2.1. The collision method of processing different materials in the disintegrator mill

The fracture of particles in collision with the milling component of one of the rotating rotors is called disintegration. The theoretical studies on milling by the collision method, which were conducted at Tallinn University of Technology (TUT), were followed by the development of the appropriate devices, called disintegrators, and the different types of disintegrator milling, the DS-series systems [48].

The disintegrator system consists of two rotors installed on the motors, which rotate in opposite directions (Fig.1.2). The rotors consist of more than one processing circle, each of them has the milling elements mounted, the material strikes against them and accelerates for the next collision.

Because of the vertical position of the rotors, the additional systems of classification and transportation of the final product are needed, which in case of using the traditional methods might need a special device that would reserve more space in the production plants. The use of the material motion velocity at the outlet from the working chamber enables to diminish the general size of the equipment, combining the milling, air classification and transportation of the final product (for the height up to 3m) in one production plant.

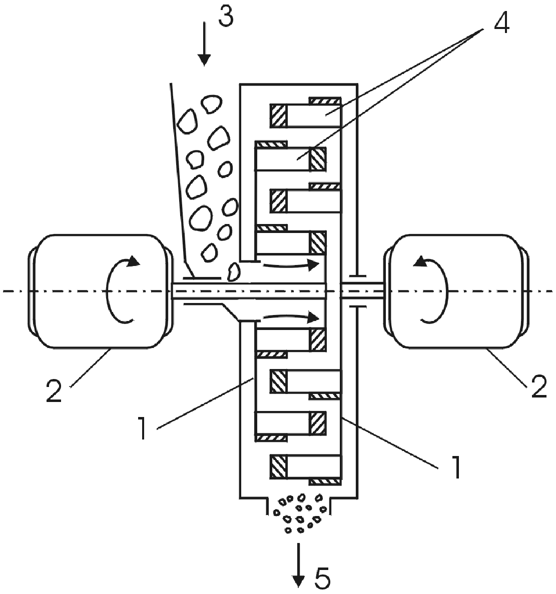


Figure 1. 2. The principal scheme of the disintegrator mill. 1 – rotors, 2 – electrical engines, 3 – material input, 4 – rotor-blades, 5 – output of the milled product

A reduction in the size of materials takes place as a result of fracturing the processed material. Through the collision of the particle with a grinding element, an intensive wave pressure will spread from the point of contact. The stresses are approximately an order higher than the strength of the material. The particle as a whole remains intact until the spreading of the compression wave reaches the opposite side of the particle. It reflects as a tension wave of the same intensity. Behind the tension wave, with a certain delay, the particle falls into pieces. After the particle break-up, the stress waves run in each piece in a very complicated manner, many times back and forth. The main (characteristic) parameters of collision are: the velocity of loading (30-200 m/s) and duration in the active zone (0.01 s) [49]. The main differences between the traditional size- reduction methods and collision methods are given in Table 1.2.

Table 1. 2. Comparison of the parameters of the materials processed by traditional methods and those processed by collision [49]

Parameter	**Traditional method**	**Collision**
Loading velocity, m/s	0.1-10	30-200
Loading time, s	10^{-2}-10^{-1}	10^{-6}-10^{-5}
Time spent in the active zone, s	1-10000	10^{-2}
Ratio of stresses to the material strength, σ / [σ]	≤ 1	≈ 10

Milling by collision means that the mechanisms of the particle size reduction of the ductile and brittle materials are different. The milling of brittle materials by collision results in a direct fracture. When milling the ductile metallic materials at the initial stage, the metal will be hardened and the fatigue fracture will occur [48]. Figure 1.3 illustrates the principal scheme of the disintegrator mill and the formation of powder particles through the milling of brittle and ductile materials.

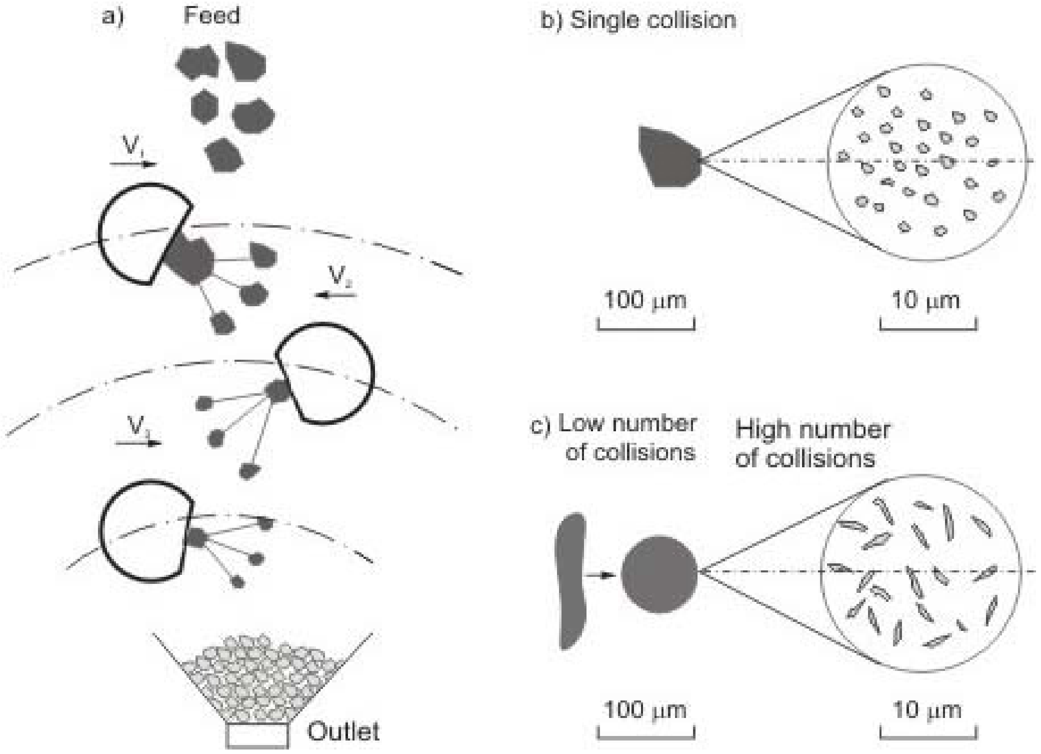


Figure 1.3. Principal scheme of the particle size reduction by disintegrator milling (a) and the fracture mechanisms of brittle (b) and ductile materials (c).

The size reduction is used to increase the bulk density, lower the storage requirements and shipping/transportation cost, it will ease the handling and conveying of the material and will liberate foreign materials.

The size reduction equipment for recycling plastics from the end-of-life durable goods will include the following advantages: it accommodates large amounts of metal, handles tough engineering plastics in reasonable throughputs, liberates molded-in and well-adhered materials, it does not embed or encapsulate foreign materials, it produces the uniform particle shapes and sizes, it is safe for a wide range of operators, it accommodates very large parts or bales of materials, provides a high throughput in the power requirement ratio, minimizes the generation of fines, it can be enclosed or evacuated, it is reasonably priced, requires low maintenance costs, it is easy to clean because of the switch-overs of material, it produces a low noise and has reasonable power requirements [50]. As summarized in Table 1.3, the types of equipment available to meet these challenges include: the hammer mills, ring mills, shear shredders with screens, the four-shaft shear shredders with screens, rotary grinders and granulators. Depending on the types of feed-stocks coming into the plastic recycling plant, there may be a need for several different stages of size reduction and liberation, each involving a different type of equipment.

Table 1.3. Variety of size reduction equipment used in recycling plastics [50]

Equipment	**Advantages**	**Disadvantages**
Traditional granulators	• Can produce fine particle sizes • Excellent liberation of materials • Well-known technology • Can have high throughputs	• Cannot handle metals • Rather high maintenance costs • High speed and can be noisy
Granulators - modified	• Can handle small amounts of metals • Can produce fine particle sizes • Excellent liberation of materials	• Advanced technology and not widely available • Cannot handle large amounts of metal or thick metal
Traditional Shear Shredders	• Can handle large and heavy metal • Large feed hopper • Well known technology	• Poor particle size uniformity • Feed rams not a stock item • Difficult to replace blades • Typically no screens provided
Four shaft shredders and modified two shaft shredders	• Can handle metal • Relatively low power requirements • Good liberation of materials • Better size control	• Not widely available • Much lower throughput rates compared to the shredder with no screen • Feed rams not a stock item • Difficult to replace blades
Hammer mills	• Can handle significant amounts of metals • High throughputs • Very robust designs • Well known technology	• Poor particle uniformity • Relatively high power requirements • Noise can be high
Rotary grinders	• Can handle moderate metal • High throughputs • Blades easily replaced & sharpened • Relatively low power requirements • Automatic ram feed • Comes with screens • Very good liberation of materials • Reasonably easy to clean	• Cannot handle large amounts of hard metals • Cannot easily produce small particle sizes • Higher cost
Cryogenic grinders	• Very fine particles sizes possible • Excellent liberation of materials	• High cost • Low throughputs • Potentially high operating cost due to liquid nitrogen needs • Cannot handle metals
Disintegrator milling	• Can produce materials with high degree of fineness and with narrow granulometry •Material is in active zone very short time (avoids over-heating) •Lower specific energy of treatment • Simultaneous separation of the components with air-classificators	• Remarkable noise level • Feed material should be in suitable size • Less efficient size reduction of materials with high toughness

It should be noted that the represented advantages and disadvantages of the size reduction technology may not hold perfectly well for specific pieces of equipment. Furthermore, the technology in this area is rapidly changing, and it is possible that some of the disadvantages will be overcome with the development efforts already underway.

Disintegrator systems of processing materials

Depending on the design of the disintegrator systems the direct, separative and selective types of milling are available and useful in powder production (see Fig. 1.4). Direct milling suits best for testing the properties of materials or producing materials with a wide granulometry, it is used for the treatment of dry, damp and liquid materials. Separative milling is meant only for dry materials, it yields materials with a high degree of fineness and a narrow granulometry [49]. Selective milling is suitable for the treatment of multi-component materials, such as the components of industrial and domestic wastes, etc. With traditional grinding methods, the selectivity is weak; if the weak and strong particles are in an active milling zone at the same time, then both the strong and weak particles will be broken. In this process, the breaking force is unevenly distributed between the strong and weak particles. In case of treating materials by collision, the stresses that appear in the particles depend on the velocity of collision, and they are independent of the particle size or material. It is possible to use the velocity low enough to keep the particles intact, and if the velocity is high enough, it will cause a break-up of both particles. Between the two extreme velocities, there is a velocity limit when the strong particle remains intact and the weak particle breaks up. The treatment of materials by collision provides the highest range of selectivity. With other methods of grinding, it is unattainable [48].

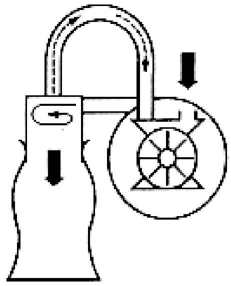
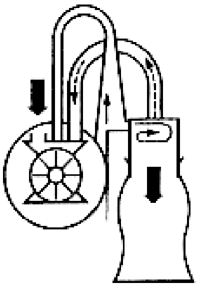
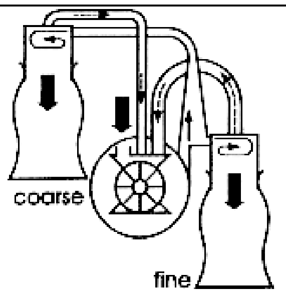
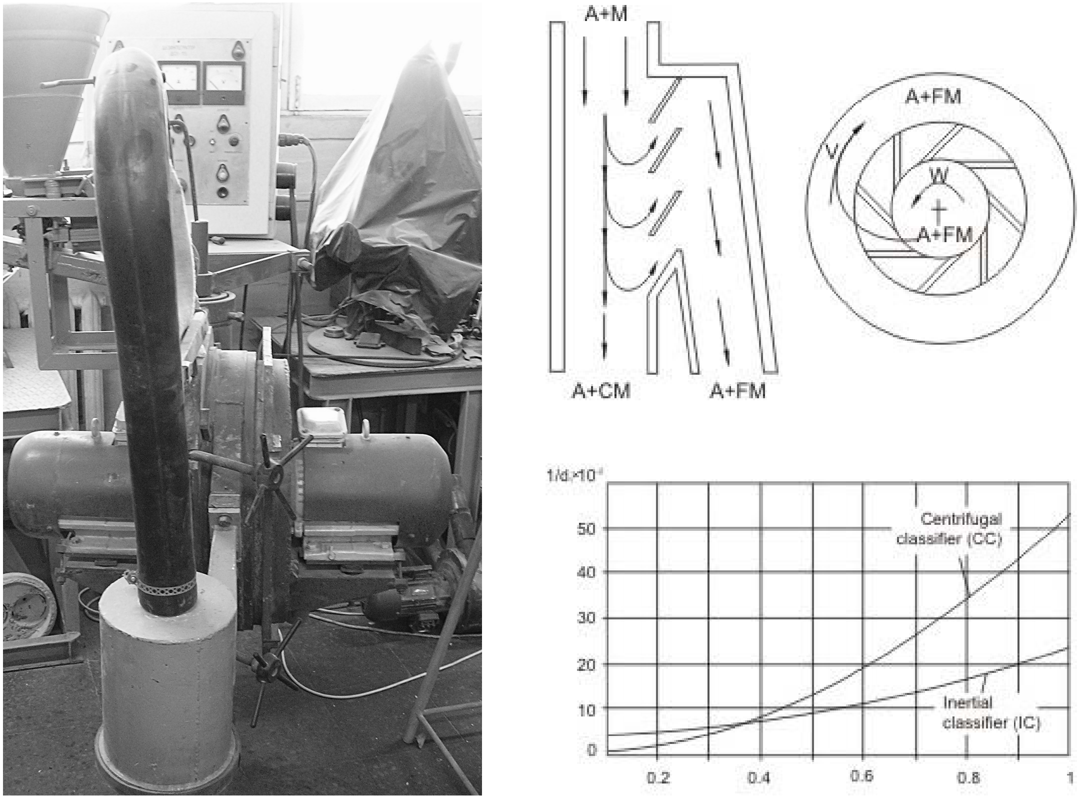
Direct milling	**Separative milling**	**Selective milling**
		

Figure 1.4. Different types of grinding and milling systems used in the disintegrator [49]

The reinforced acrylic plastic wastes have different mechanical properties because of the plasticity of glass fibre, but they can be reprocessed by the selective milling system. The aim of our experiments was to reduce the particle size of acrylic plastic and keep the remaining glass fibre intact for separation. In DS disintegrator systems, the ground material ejected from rotors carries a significant amount of kinetic energy, which can be used for further transportation of the material [51, 52]. The separation systems in the DS-series disintegrators are based on the aerodynamic forces. A special inertial classifier with a closed air or gas system has been developed [52, 53]. This system is autonomous and ecologically clean due to the use of kinetic energy in the output material. The system does not need any additional devices of transportation or fans. For various materials and disintegrator milling systems, different inertial classifiers have been designed and developed as an axial inertial classifier and a classifier with a grid formed by the row of blades (see Fig. 1.5) [54, 55].



a) b)

Figure 1.5. The laboratory disintegrator milling system DSL-115 with the inertial classifier (a) and the principal schemes of the inertial (air) classifier and centrifugal classifier (b)

1.2.3. Waste management legislation and waste handling systems in EU

The directives are the instructions given to the member states of the European Union – currently twenty countries – to alter or supplement their national legislation within some specified time unit in order to achieve the results within another specified time unit. Each EU piece of legislation – including directives – is given a reference number: the first two digits refer to the year when the piece of legislation was adopted and the second set of digits is specific to the piece of legislation and is given in a chronological order of that year.

There are several stages in the policy process of adopting the directives. First, the European Commission writes a working document showing an intention to propose a directive. After discussion, the working document can become a draft proposal for the directive. Then the directive is proposed by the European Commission. Subsequently, it is examined by the European Parliament, the Council of the European Union (made up of relevant ministers, varying according to the subject discussed) and other institutions. It typically takes 2 to 4 years for a directive to be adopted after it has been proposed. Once a directive has been adopted, the member states are required to implement it within a specified time usually two years. The national waste management strategies have been produced to implement the European waste directives.

1.2.3.1. Recycling directives in EU

Framework of waste management directives in EU

The Framework of Waste directives establishes the framework for waste management across the European Union. It provides common definition of waste management across the EU (see Fig. 1.10): 1) prevention, 2) reuse, 3) recycling, 4) energy recovery, 5) incineration without energy recovery, 6) landfill [1]. The hierarchy of waste management, according to the EU directives, is as it follows from Figure 1.10.

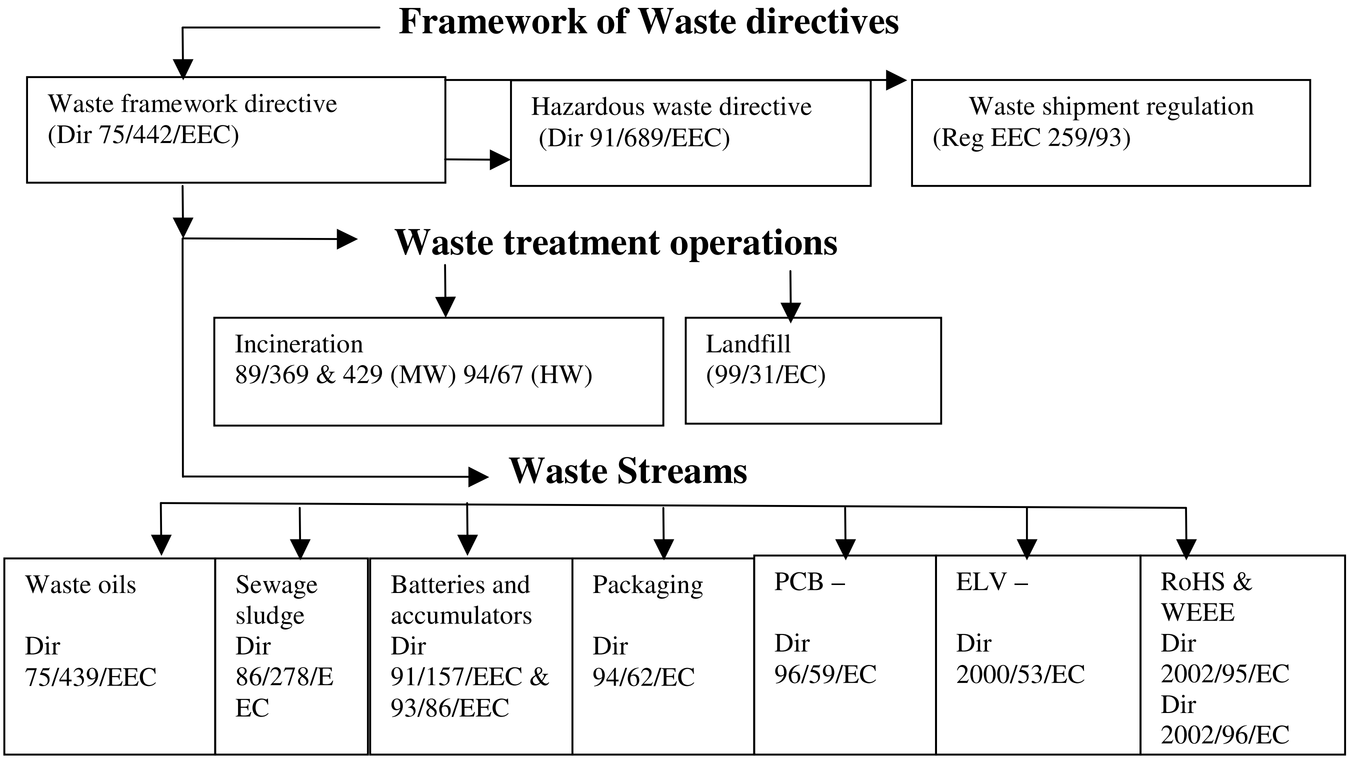


Figure 1.10. The framework of the waste management directives in EU [104]

Some common definitions are associated with the electrical and electronic waste legislation:

Dangerous substance or preparation means any substance or preparation, which have to be considered dangerous under Council Directive 67/548/EEC or Directive 1999/45/EC of the European Parliament and that of the Council [1].
Disposal means any of the applicable operations provided in Directive 75/442/EEC.
Energy recovery means the use of combustible waste as a means of generating energy through direct incineration with or without other waste but with the recovery of heat.
Prevention means that the measures aimed at reducing the quantity and harmfulness to the environment of WEEE and the materials and substances contained therein.
Recovery means any of the applicable operations provided in Directive 75/442/EEC.
Recycling means reprocessing the waste materials for the purpose of reforming the originals or for other purposes, but excluding the energy recovery in the production process.
Re-use means any operation by which WEEE is used for the same purpose for which it was conceived, including the continued use of WEEE, which is returned to the collection points, distributors, recyclers or manufacturers.

Treatment means any activity after the WEEE has been handed over to the facilities of de-pollution, disassembly, shredding, recovery or disposal and any other operation carried out for the recovery and/or the disposal of the WEEE.

Restriction of Hazardous Substances in Electrical and Electronic Equipment (ROHS) Directive (2002/95/EC)

It does affect the manufacturers, sellers, distributors and recyclers of electrical and electronic equipment, which contain lead, mercury, cadmium, hexavalent chromium, polybrominated biphenyls or polybrominated diphenyl ethers [56].

The directive aims at [56]:

- protecting human health and the environment by restricting the use of certain hazardous substances in the new equipment;
- supplementing the WEEE Directive.

Key elements [56]:

- Since 1 July 2006, the new electrical and electronic equipment will not contain lead, mercury, cadmium, hexavalent chromium, polybrominated biphenyls or polybrominated diphenyl ethers.
- Certain applications are exempt from the requirements of the directive including mercury in certain types of fluorescent lamps, lead in the glass of cathode ray tubes, electronic components and fluorescent tubes, lead in electronic ceramic parts and hexavalent chromium as an anti-corrosion of the carbon steel cooling system in the absorption refrigerators. The exemptions will be reviewed every four years.
- Before 13 February 2005, the European Commission will review the terms of the Directive to take into account any new scientific evidence.

WEEE – Waste Electrical and Electronic Equipment (WEEE) Directive (2002/96/EC)

This directive does affect those who are involved in manufacturing, selling, distributing, recycling or treating electrical and electronic equipment (including large household appliances, IT and telecommunications equipment, audiovisual equipment, lighting equipment, electrical and electronic tools, toys, leisure and sports equipment, medical devices and automatic dispensers) [2]. The targets of a minimum reuse, recycling, and recovery are given in Table 1.4.

Electrical and electronic equipment means the equipment, which is dependent on electric currents or electromagnetic fields in order to work properly, and the equipment for the generation, transfer and measurement of such currents and fields falling under the 10 categories, designed for the use with a voltage rating not exceeding 1000 volt for alternating current and 1500 volt for direct current [2];

Electrical and electronic equipment as waste or “WEEE” means the electrical or electronic equipment, which is waste within the meaning of Article 1(a) of Directive 75/442/EEC, including all components, sub-assemblies and consumables, which are parts of a product at the time of discarding.

Table 1.4. Product categories and minimum end-of-life reuse, recycling and recovery targets set by the WEEE Directive

No.	Product category (See Annex 1B of the WEEE Directive for full list of items in each category)	WEEE reuse and recycling targets in %	WEEE recovery target (includes energy recovery in a power plant) in %
1	Large household appliances (e.g. fridges, washing machines, electric ovens)	75	80
2	Small household appliances (e.g. vacuum cleaners, toasters, irons, clocks, scales)	50	70
3	IT and Telecommunication equipment (e.g. computers, photocopiers, telephones)	65	75
4	Consumer equipment (e.g. TV-sets, video recorders, hi-fi equipment)	65	75
5	Lighting equipment (e.g. fluorescent lamps, discharge lamps)	80	N/A
6	Electrical and electronic tools (e.g. drills, sewing machines, lawnmowers)	50	70
7	Toys, leisure and sports equipment (e.g. video games and consoles, train sets)	50	70
8	Medical equipment systems (e.g. radio-therapy equipment, pulmonary ventilators)	No targets set at present	No targets set at present
9	Monitoring and control equipment (e.g. thermostats, control panels)	50	70
10	Automatic dispensers (e.g. drinks machines)	75	80

The directive aims at [2]:

- reducing the waste arising from electrical and electronic equipment;
- improving the environment-friendly performance of all those involved in the life cycle of electrical and electronic equipment

Key elements [2]:

The directive covers the WEEE directive used by the consumers for professional purposes.

By 13 August, 2005 [2]:

- private households will be able to return their WEEE to the collection facilities free of charge;
- producers (manufacturers, sellers, distributors) will be responsible for financing the collection, treatment, recovery and disposal of WEEE from the private households deposited at these collection facilities;
- producers will be responsible for financing the collection, treatment, recovery and disposal of WEEE from users other than private households, it concerns the products placed on the market after 13 August 2005;
- producers will also be responsible for financing the management of WEEE, it concerns the products placed in the market before 13 August 2005. However, it is possible to recover these costs totally or partly by the users other than private households.

By 31 December 2006 [2]:

- producers will be required to achieve a series of demanding recycling and recovery targets for different categories of appliance;

- Estonia must have reached an average WEEE collection rate of four kilograms for each private household annually.

1.2.3.2. Waste legislation in Estonia

Producer responsibility for problematic products is constituted in Waste Act (2004). Producer responsibility covers the following types of waste: packaging waste, the waste of electrical and electronic equipment, scrap cars, car tyres, and scrap paper. As for batteries, the joint responsibility is shared with the municipalities.Packaging waste prevention, handling and reuse is prescribed in Packaging Act (2004).

The types of deposit packaging and excise amounts are defined in Packaging Excise Act (1996). The government regulations are issued for hazardous waste management andrequirements for landfill usage and closing.

1.2.3.3 Waste handling systems in Estonia

Packaging waste

The packaging operators pack the goods or import and sell the packed goods. The obligation in the Packaging Act is: to recycle 50 % of total volume weight of packages, at least 15 % from each group of packages should be recycled [57]. The obligations of the packaging operator can be transferred to the producer responsibility of an organization by contract [57].

Producer responsibility organizations for packaging waste collection are:

- The Estonian Recovery Organization (ETO)
- Estonian Packaging Circulation (EPC)/Eesti Pakendiringlus (EPR)
- ·Eesti Pandipakend LLC (EPP) is a recovery organisation for the recovery of packaging subject to the payment of a deposit.

Recycling of plastic packaging waste

There are two bigger recycling companies (Plastitehase Ltd, Dagöplast Ltd) which are recycling mainly the plastic packaging films (LDPE, HDPE and PP). One company is recycling (Lipland LLC) rigid packaging and technological scrap (LDPE, HDPE, PP and PVC). Recycled plastic granules are mainly sold to China or India.

WEEE:

Producers are responsible for financing the collection, treatment, recovery and disposal of WEEE, required from the users other than private households, from the products placed on the market after 13 August 2005, establishing the producer responsibility for generating WEEE.*MTÜ EES-Ringlus* was established by the producers and retailers of electronic and electrical equipment, including batteries.

Recycling of WEEE:

- Large electric household appliances are collected and recycled in Estonia, by Kuusakoski Oy except for the fridges
- Fridges are recycled in Finland, Forssa
- The collected small electric and electronic appliances are recycled in Lithuania, UAB (Elektroniniu Masinu Perdirbimas – EMP)
- New plant for recycling WEEE is established (Weerec Ltd) to recycle more than 90 percent of WEEE. Plastic fraction will be precrushed and sold to India or China.

1.2.4. Reuse and recovery of plastics

There continues a growing worldwide demand for recycling thermosetting polyester/glass-fibre reinforced plastics in different marketable products, such as the automobile parts. The starting point was the realization that the hammer mill process used for size reduction of the parts, not only grinds the resin to powder but tends to split along the line of fibre, leaving it intact. After grinding the waste to powder, the fibre can be separated for re-use. The recyclate is suitable for the production of new parts as well as for the production of fillers and/or the reinforcement material. The parts containing up to 60 % of the recyclate mass are tested, suggesting that the properties of strength, surface quality and paintability are nearly as good as those of the new parts; and up to 25% of the recyclate mass can be incorporated in a significant loss of properties. The parts of clean SMC (the sheet moulding compound) and BMC (the bulk moulding compound) can be recycled and the material can be used in new SMC parts. Over 20 of the US automotive components contain the recycled SMC from in-plant and post-consumer scrap, using the material, which has 6-25 % of the mass replacement for calcium carbonate (which makes up about 50% of the raw material of SMC) [15].

In our case, the recycling of PMMA+GFP composite scrap is very complicated because they contain both the thermoplastic PMMA and thermosetting polyester resins with the glass fibre reinforcement.

1.3. Characterization of powdered materials

1.3.1. Definition of powder

There are a few terms necessary to understand before having discussion on the characterization of powders. The three definitions of powders are as follows:

- the powder is defined as a finely divided solid, smaller than 1mm in its maximum dimension [58];
- powder is defined as an aggregate of discrete particles that are usually in a size range of 1 to 1000 μm [59];
- the powder production is a process by which the powder is produced, such as machining, milling, atomization, condensation, reduction, oxide decomposition, carbonyl decomposition, electrolytic decomposition or precipitation from a solution [59].

1.3.2. Granularity and morphology

Granularity

Under granularity, the size of particles and fractions are observed and determined by different granulometric methods, such as sieving and image analyses.

To study the granularity of a ground product by the granulometric analysis, the modified Rosin-Rammler distribution function and method are used [51],

$$f_m(X) = \frac{n-1}{m}\left(\frac{x-x_0}{m}\right)^{n-1} e^{-\frac{n-1}{n}\left(\frac{x-x_0}{m}\right)^{n}} \qquad (1.1)$$

where n, m and x_0 are the parameters of distribution, the logarithmic size of the particle x is given as [51],

$$x = \log_k \frac{X_0}{X} \qquad (1.2)$$

where x indicates the natural size of the particles of the material; X_0 is the upper limit on the possible particles size of the studied material; X is the natural particles size of the studied material; and k is the coefficient (ratio) of the sieving system used in the experiments (k = 2, 2^5, 2^{25}).

To characterize the particles size, the following parameters are defined [60]:

- the median diameter d_{50} (the median of the mass density function of Rosin-Rammler) is calculated according to the experimental (results of sieving analyses) cumulative distribution functions;
- the mean diameter d_m (the linear descriptor obtained by IA) is the average length of the number of diameters taken at 5 degree intervals around the centre of the mass blob;
- volume diameter d_m^v is calculated on the basis of the plate shape formula of the particles

$$d_m^{\ v} = d_m \cdot\ d_m \cdot d_m \cdot\ 0.2 \qquad (1.3)$$

Morphology

Geometrical characteristics of powder particles can be divided into two main classes: size and shape parameters [61, 62, 63]. Size parameters (for example, the area perimeter) describe the geometrical object, independent of its shape or location.

On the other hand, shape characteristics characterize mainly the shape without any influence on the size. Mostly, only some approximation of the examined object is used for the shape description – the so-called Legendre ellipse is often used instead of the original object [64]. The Legendre ellipse is an ellipse with the centre in the object centroid and with the same geometrical moments up to the second order as has the original object area.

A lot of these characteristics can be used for describing the ellipse of the original object.

If "*a*" and "*b*" are the axes of the equimomental Legendre ellipse (Fig. 1.6), then elongation is defined by relation $EL = \log_2 a/b$ and dispersion by $\mathrm{DP}= \log_2 \pi \cdot a \cdot b$. The elongation for a circle is EL=0, for ratio of axes 1:2 is EL=1 and EL grows for the ellipses with an increasing ratio of the major and secondary axes – see Fig. 1.7. Dispersion DP is a measure, which allows us to evaluate the range of difference in smoothness. This enables to compare the smoothness of the evaluated object with the ideal ellipse: dispersion of the ellipse is DP=0 and it grows when increasing the object roughness, see Fig. 1.7 [67].

Another well- known and frequently used characteristic is the shape factor (roundness) of the object. If p is the perimeter and a stands for the area of the object, the roundness value is calculated by [65, 67] $RN= P^2/4\pi A$. To characterize the roundness, sometimes shape factor $SF=1/RN$ is used. The roundness factor proposed in [66], RNF is defined by the relationship

$$RNF = P/d_i \qquad (1.4)$$

where P is the particles perimeter and d_i is the diameter of the circle with an equal particle area (see Fig. 1.8).

Then the shape factor was defined as f=4 $\cdot\ \pi \cdot a \cdot b/P^2$. The shape factor for circle is equal to 1, if the object shape tends to the line segment, f tends to =0 [67].

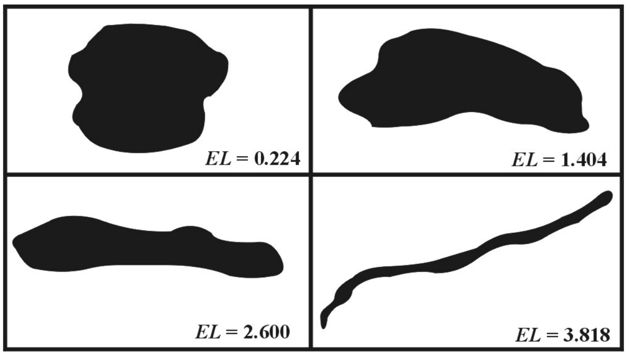


Figure 1.6. Samples with different values of elongation *EL*

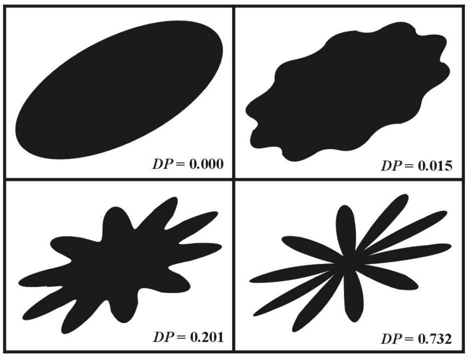


Figure 1.7. Samples with different values of dispersion *DP*

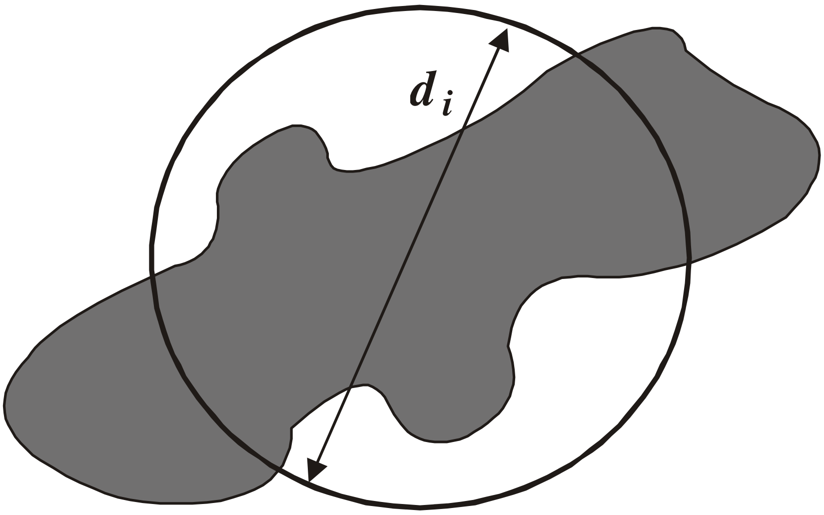


Figure 1.8. The morphology study based on the circle with the same area as the particle

An evaluation of the ellipticity and irregularity of the shape factor is proposed in [68]. The so-called irregularity parameter (*IP*) is defined by the following formula:

$$IP = \frac{D}{d}, \tag{1.5}$$

where *d* and *D* are the diameters of the maximum inscribed, and the minimum circumscribed circles, respectively (see Fig. 1.9).

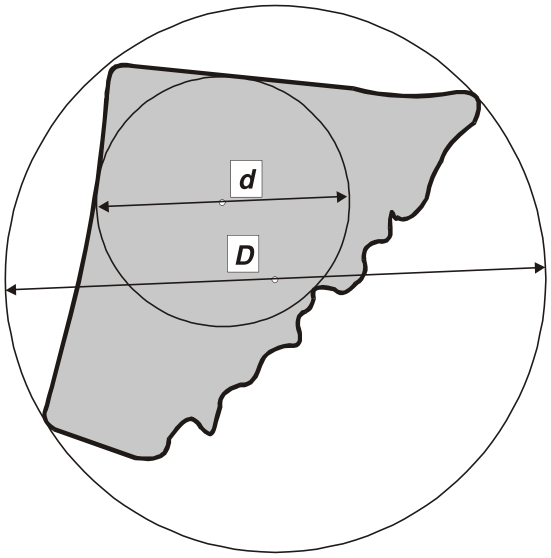
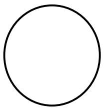
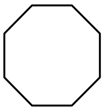
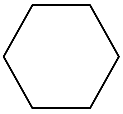
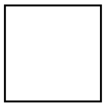
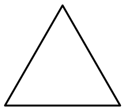


Figure 1. 9. The morphology study is based on the maximum of inscribed and the minimum of circumscribed circles.

As shown in [68], the newly defined shape factor works very well. This shape factor can be applied to elongated or irregular particles, but it does not allow us to decide if the particle is elongated or irregular.

In Table 1.4 the shape factors of regular configurations are given in the order of increasing sharpness: circular, orthogonal, hexagonal, quadrangular and triangular. As it follows from Table 1.5, *IP* is the most sensitive parameter, and parameter *RNF* is the most insensitive one. Whereas *IP* characterizes integrally both, angularity and elongation, it does not allow us to find separately the angularity and elongation [69].

Table 1.5. The shape factors of particles of different form [69]

Shape factor					
Roundness *RN*	1.0	1.055	1.103	1.273	1.654
Roundness factor *RNF*	1.0	1.027	1.050	1.128	1.286
Irregularity parameter *IP*	1.0	1.082	1.155	1.414	2.0

1.3.3. Properties

Density

For non-cellular plastics the density and relative density are determined according to standard ISO 1188:1987 [70]. It cannot be used for density measurements of plastic powder materials.

The bulk density measurements of powders include the apparent density and tap density measurements as described below. The apparent density of powder or the mass of a unit volume of loose powders expressed in grams per cubic centimetres is one of the fundamental properties of powder. This characteristic defines the actual volume occupied by a mass of the loose powder, which directly affects the processing parameters, such as the design of compaction tooling and the magnitude of the press motions required to compact and make dense the loose powder. The apparent density ρ_a of the powder depends on the density of the solid material, the particle size, the distribution of particle sizes, the particle shape, the surface area and particle arrangement. The most common method for determining the apparent density of metal powders uses the Hall flow-meter. Both ASTM B212 and the Metal Powder Industries Federation (MPIF) standard 04 describe this method. The determination of the apparent density is made by pouring the powder into the funnel and allowing it to flow into 25 cm^3 density cup. The apparent density in grams per cubic centimetres is determined by weighing the powder in the cup in grams and dividing it by 25 cm^3 (cup volume) [71].

The tap density ρ_t is defined as the density of the powder when the volume reception is tapped or vibrated under specified conditions. The tap density is always higher than the free-flow apparent density. To determine the tap density, the standard mass (usually 50g) of powder is weighed ± 0.01 g. The powder is poured into a clean, dry graduated cylinder, taking care that a level surface of powder is obtained. The powder is settled in the cylinder by mechanical or hand tapping. The volume of a fully densified powder sample in the graduated cylinder is read and used in the following calculation of the tap density:

$$\rho_t = m/v \tag{1.6}$$

where m is the mass of powder in grams and V is the volume of the tapped powder in cubic centimetres. The results should be reported as the nearest to 0.1 g/cm^3.

Specific surface area

Permeameter is an instrument to measure resistance to the fluid flow through a compact powder bed. This information is used to determine the related properties of a powder, such as the pore size distribution, specific surface area or the average particle size for a packed powder sample [30]. Frequently, the data of the surface area are not given in the suppliers´ literature, and thus, it becomes necessary to obtain an estimate. As a solution, a comparison with the same type of filler having a similar particle size distribution can be done. Another solution is to compute a value by one or two methods: the Gates diagram or the computation based on the equivalent spherical diameter (esd), or the diameter of a sphere having the same volume as the particle [72].

The computational values of the specific surface area are based in the areas of equivalent spheres. Unless the spheres are the subject to calculation, it is obvious that for all other particles the proportionality constant related to the particle shape must be applied. The calculation is simplified by modern data processors and gives a reasonable estimate of the surface area, provided that the deviation from sphericity for the particles is not extremal. Few filler particles are regularly shaped: so the factors for this deviation were empirically derived from the known values of the surface area for shape classes. In effect, the shapes of the most real particles are too variable to be dealt with directly. The difference between the actual and calculated surface areas will be modest for the low-area fillers, but for others there may be a difference greater than two or three times [72].

$$SSA = \frac{K}{\rho}\left(\sum \frac{dW}{\overline{X}_m}\right) \tag{1.7}$$

where SSA is the surface area in m^2/g, K *is* the proportionality constant for the particle shape (spheres = 6, blocky and tabular = about 12), dW is the mass increment in grams, ρ is the density g/cm^3 and $\overline{X}_m$ is the average particle size of mass [72].

Flowability

The flow of metal powders is determined by the standard methods developed by ASTM and MPIF. The flow rate is the time required for a powder sample of the standard mass (50g) to flow under the atmospheric conditions through a funnel into the cavity of a container or a mould. The device, which is most commonly used for measuring the flow rate, is the Hall flow-meter from ASTM B 213 and MPIF 3 (the equivalent standards include ISO 4490, the Japanese standard JIS 7-2505-1966, and the German standard 82-69). The test equipment consists of a funnel with a calibrated hole of 2.5 mm in diameter. With the help of the stopwatch and weighing balance, the flow rate of metal powders can be easily determined. A powder, which does not flow through a 2.5 mm orifice Hall funnel with or without an external impulse, is said to be non-free flowing powder. In this case, the Carney funnel, which is smaller in dimensions than the Hall funnel, except for a larger orifice diameter of 5 mm, can be used to obtain a relative measure of the flowability of non-free flowing metal powders. The Carney funnel is not a standardized test method and it involves no standard calibration procedure [71].

1.4. Summary of the literature

The recycling of thermosetting plastics as well as composite plastics has not been resolved yet and different prospective methods of mechanical recycling are underway. The techniques of mechanical recycling, such as these, contribute to chemical recycling and thermal-recycling processes, such as gasification, liquefaction and energy recovery processes. Mechanical reprocessing exists as the main possibility of recycling thermosetting composite plastics to reclaim the material from composite plastic scrap by producing plastic powders.

For the characterization of the properties of plastic powders (granularity, morphology, technological properties), the same methods and parameters commonly used for metallic powders are acceptable. The parameters, such as granularity and morphology, are important for designing new products and the innovative technologies of their manufacture.

The information about the application of secondary plastic powders reprocessed and produced from the composite or compounded plastic scrap as well as from the multi-materials is not available.

The thermoplastic PMMA (the extruded sheet material used in the vacuum forming of bathtubs) cannot be melted again to recover the material because of the molecular mass, which is more than 1,000,000 in comparison with the recoverable PMMA (granules for injection moulding) whose molecular mass is less than 60,000; the latter is a very different material (for instance, it is used for the production of the coloured back lights of cars). There is only one solution, which can be proposed to recover the extruded sheet material of the PMMA, and it is producing powders as the filler material for the application of designing new products (for example, washbasins etc).

The targets of the material recovery have proven hard to meet under the European WEEE Directive. However, a simple separation and recovery of the metal fraction, no matter how advanced the technology and processing equipment are, is unlikely to meet the requirement of 80 percent. To achieve the required recovery rate, it is recognised that a clean separation, and more importantly the final recovery of the plastic fraction must be seen as paramount.

2. EXPERIMENTAL

2.1. Studied materials

Polymethylmethacrylate (PMMA) sheet is the most commonly used material in the manufacture of bathroom equipment. The material of preheated PMMA sheet has good technological properties for the vacuum forming of acrylic shells in bathtubs and mini-pools. The laminated vacuum-formed acrylic shells combined with the thermosetting synthetic resin, such as polyester or epoxy resin, and the reinforcing material like glass fibre mat or fabric produce reinforced acrylic plastics. The generation of wastes takes place after cutting off the technological edges of vacuum forming. Industrial wastes of reinforced acrylic plastics form about 80% and the acrylic plastic scrap without technological additives account for approximately 20% of the total amount.

Another big group of plastic wastes is formed by the post-consumer electrical and electronic equipment wastes (WEEE) consisting of thermoplastics, compounded plastics (for example, personal computers, household appliances) as well as composite plastics (bathtubs, steam-cabins, capsules, etc) and multi-materials (PCBs).

2.1.1. Composite plastics of the PMMA+GFP

PMMA produced by the radical chain polymerization in mass (molecular mass 1 million) as sheet material was used for the extrusion of sheet materials by vacuum forming. The cut-off technological edges of the PMMA sheet material, then vacuum formed and reinforced with glass fibre in the matrix of the polyester resin, were used as technological waste. The main properties of the studied composites are given in Table 2.1. As it follows from Table 2.1, the physical and mechanical properties of PMMA were the following: tensile strength 78 N/mm^2, the modulus of elasticity 3.33 MN/mm^2, the impact strength 12 kJ/m^2 and density 1200 kg/m^3.

The properties of the hardened unsaturated polyester resin (UP) with 25-30 mass % of glass fibre reinforcement were: tensile strength 75 N/mm^2, modulus of elasticity 7.70 MN/mm^2, flexural modulus 6.70 MN/mm^2 and density 1700 kg/m^3.

Table 2.1. Physical and mechanical properties of the studied plastics (at 23 °C)

Material	**Tensile strength R_m N/mm^2**	**Modulus of elasticity E MN/mm^2**	**Impact strength kJ/m^2**	**Density ,ρ kg/m^3**	**Elongation after fracture A %**
PMMA	78	3.33	12	1200	4
UP (matrix)	50	4.60	5	1200	2.3
GFP	75	7.70	9	1700	3
ABS	56	3.40	17.5	1170	30
PC/ABS	88	3.40	34	1200	150

2.1.2. Compounded plastics

Dismantling processes and recycling of plastics from electronic scrap are discussed in recent studies [73-77]. To detect the brominated or phosphated flame-retardants in compounded plastics the FTIR spectroscopy or mass spectrometry is suggested [76, 78, 79].

Compounded plastics subjected to recycling from dismantled personal computers (see Figs 2.1 and 3.25–3.27) are forming from processors (4.3 mass %), monitors (21.0 mass %) and keyboards (78.6 mass %). The physical and mechanical properties of the compounded plastics are given in Table 2.1. Compounded plastics (PC-ABS) and polyphenylene-polystyrene (PPE-PS) from monitor housings forming 21 mass % of the monitors were used as the compounded plastic waste.

Figure 2.1. Compounded plastics from dismantled personal computers

2.1.3. Multi-materials

Printed circuit boards (PCBs) are common components of many electronic systems built for both military and commercial applications. PCBs are typically manufactured by laminating dry film on clean copper foil, which is supported on a fibreglass plate matrix. Film is exposed to the film negative of the circuit board design and an etcher is used to remove the unmasked copper foil from the plate. Solder is then applied over the unetched copper on the board. Depending upon the use and design of the particular PCB, various other metals may be used in the manufacturing process, including lead, silver, gold, platinum and mercury [80]. PCBs are potentially a difficult waste material to process since they generally have no usefulness once they are removed from the electrical component in which they were installed. In addition, they typically consist of the materials classified as a hazardous or "special" waste stream. They must be segregated and handled separately from other non-hazardous solid waste streams. PCBs that are handled as waste materials must be processed using any of the several available disposal options. Not only are these options expensive, but they also require a significant amount of effort made by the generator. Furthermore, since some of these disposal options do not include the destruction of waste circuit boards, the generator also retains much of the liability associated with improper handling or disposal. As an alternative to off-site disposal, PCBs can be handled and processed to recover the value of the raw materials that are used to produce the boards. Several companies offer recycling services as an alternative to off-site disposal of obsolete printed circuit boards. These services will generally process the boards by systematically removing raw materials of value from the board matrix. The board matrix consists of phenol or epoxy resin with copper sheet covering, paper, and additives such as flame retardant [77]. The materials include metals such as silver, lead, copper, and gold. Depending upon the volume and characteristics of the PCBs processed through a particular vendor, the raw materials can be recovered and the salvage value potentially returned to the generator. PCBs are also containing flame-retardants which should be extracted [79, 80].

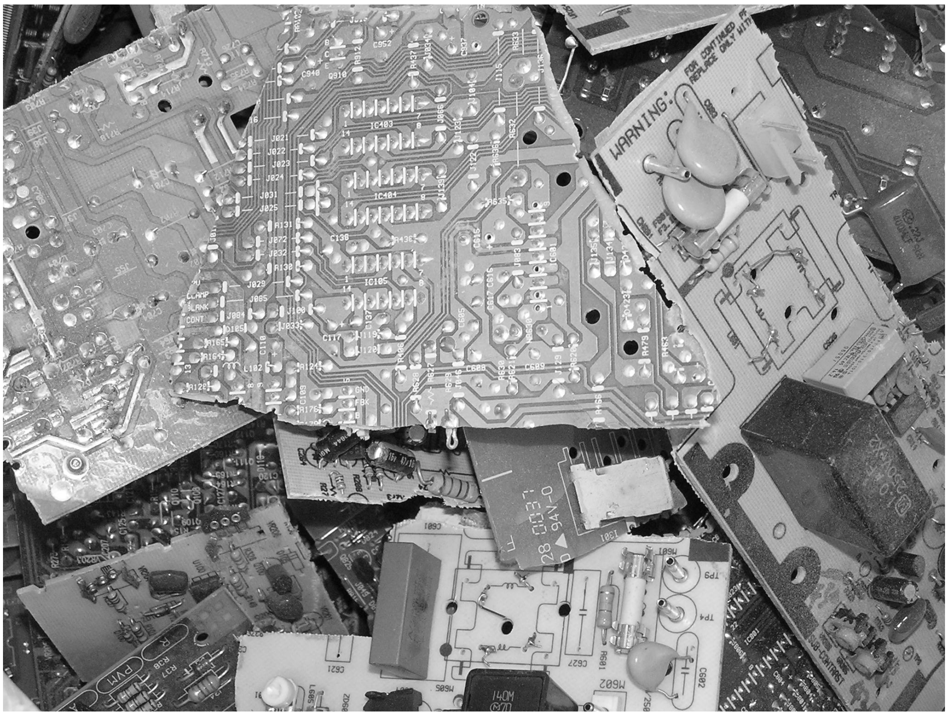


Figure 2.4. Preliminarily crushed PCBs separated from dismantled personal computers

2.2. Technology of disintegrator milling for reprocessing composite plastic scrap

To process different materials, the multi-functional DS-series disintegrators developed at TUT were used. These series include the laboratory disintegrator milling system DSL-175 with the productivity of some kg/h and the DSL-115 with the productivity of some hundreds kg/h. For the preliminary size reduction of the plastics to be retreated, the mono-rotor type experimental disintegrator DSA-158 and semi-industrial DSA-2 were used. The main characteristics of the applied equipment and milling systems are shown in Table 2.2.

Table 2.2. Characteristics of the disintegrators used

Parameter	**Experimental disintegrator DSA-158**	**Semi-industrial disintegrator DSA-2**	**Laboratory disintegrator milling system DSL-115**
Type of device (position of rotors)	Horizontal	Horizontal	Vertical
Rotor system	Mono-rotor	Mono-rotor	Two-rotor
Diameter of rotors, mm	600	480	480
Number of pins/blades roads	1	3	5
Rotation velocity of rotors, rpm	Up to 1500	Up to 3000	Up to 3000
Impact velocity, m/s	Up to 40	Up to 75	Up to 150
Specific energy of treatment E_s, kWh/t	0.2	2.4	6.7
Possible operating system	Direct	Direct	Direct / separative
Input (max particle size), mm	100	50	10
Separation system	–	–	Inertial or centrifugal

The reprocessing technology of the technological waste of composite plastics consists of the following steps:

- preliminary preparation of the material by cutting it into the suitable pieces to be fed by experimental disintegrator type crusher;
- preliminary application of direct or multi-stage milling by the semi-industrial disintegrator;
- final milling under the conditions of the separative milling system of the disintegrator

The main kinematic parameter in the processing of materials is the specific energy of treatment E_s in kWh per ton, both in view of the grinding effect (grindability) and the economic aspect of the process. Grindability of plastics as a function of particle size of the specific energy of treatment was studied.

2.3. Methods of characterizing the milled plastic powders

Granularity and morphology are the main parameters of powders characterizing the particle properties from the physical and technological aspects.

2.3.1. Granularity and morphology of the powder particles

The granularity of powders was determined by different methods: the sieving analysis and image analysis. To evaluate the granularity of coarse powder (having the particle size more than 50 μm), the sieving analysis (SA) that ensures sufficiently good results was used. Before performing the sieving analysis of the milled reinforced acrylic plastic powder, which initially consisted of PMMA plastic powder and glass fibre reinforcements, the glass fibres were separated to evaluate the fractions of the PMMA powder by size. The distribution of the particle size is adequately described by the modified Rosin-Rammler distribution function as the applied method.

To characterize the particle shape, image analysis (IA) was also used. For the characterization of the particles size, the following parameters were studied:

- mean diameter d_m, which is the average length of a number of diameters taken at 5 degree intervals around the centre mass of the blob;
- cumulative volume distribution function F_v of the particles volume;
- median diameter d_{50}, which corresponds to half of the total content of powder obtained from the cumulative distribution function.

For the characterization of the particles size, the following shape parameters were calculated [4]:

- the ellipticity parameter; to characterize ellipticity, the aspect ratio *AS* (similar to elongation in literature) was calculated by

$$AS = a/b, \tag{2.1}$$

where *a* and *b* are the axes of the Legendre ellipse (the ellipse is an ellipse if it has its centre in the object's centroid with the same geometrical moments up to the second order as it is with the original object area).

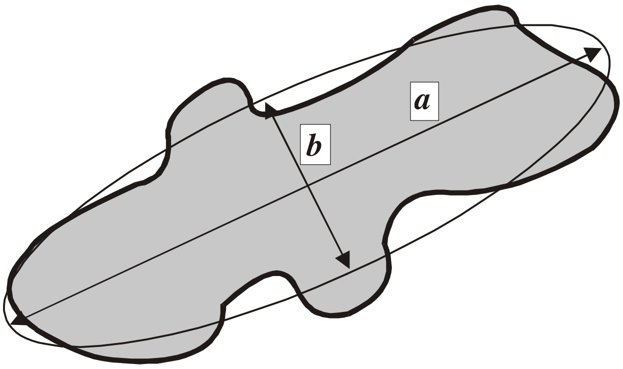


Figure 2.4. Morphology study based on the Legendre ellipse

- the elongation *EL,* which was defined as

$$EL=\log_2 (a/b) \tag{2.2}$$

For example, the elongation of the circle is 0 and that of an ellipse with the ratio of axes 1:2, is equal to 1;

- irregularity or the surface smoothness; the value of roundness *RN* was calculated by [65, 67]

$$RN=P^2/4\pi A \tag{2.3}$$

(The roundness of the circle equals to one, if the object shape approaches the line segment, which approaches zero).

For the linear measurements of the particle size with optical microscope (OM), small quantities of acrylic plastic powders were spread on a well-cleaned glass slide. Several experiments were made to avoid agglomeration and particle adherence with each other. The preliminary treatment of a plastic powder with the particles size less than 315 μm in the wet media gave poor results, because the particles had stuck together. Then we cast the plastic powder into epoxy resin and this gave us good results with particles separation. For the analysis of the particle shape, cross-section polishes were made by the mechanical grinding-polishing procedure. Of course, there were some particles of microscopic metal from the rotor blades, but it was easily observed and thus removable with the graphical design.

The equipment used for the image preparation and analysis was as follows: the optical microscope Nikon-Microphot-FX and the image analysis system *Image Pro* 3.0 [69, 81].
The above mentioned parameters were automatically calculated for every particle and they were stored in files. All characteristics were statistically evaluated. The dependence of all characteristics on the number of disintegration cycles was searched.

2.3.2. Technological properties of powders

The following technological properties of powders were determined:

- the apparent density ρ_a of the powder in kg/m^3
- the tap density (ρ_t) of the powder in kg/m^3
  Densities were determined according to standard ASTM B212;
- the apparent density of the powders with the particle size less than 1.25 mm was determined by using the Scott volumeter described in ASTM B329;
- the specific surface area of the fine fraction (< 0.315 mm) of the plastic powder was determined by using the Blaine permeameter, which is a simplified version of the air permeameter and relies on the various pressure techniques. Vacuum was used to displace the water in a U-tube connected in series with the powder_cell. The resultant pressure caused the air to flow through the powder bed, and the time required for the displaced water to fall back to its equilibrium position was measured. This method resulted in a measured specific surface area, which decreased with porosity [71, 82].
- The specific surface area of the other fractions (0.315-0.63; 0.63-1.25; 1.25-2.5; 2.5-5.0; 5.0-11.2) was calculated on the basis of formula (1.7).

$$SSA = \frac{K}{\rho}\left(\sum \frac{dW}{\overline{X}_m}\right), \tag{1.7}$$

  where *SSA* is the surface area in m^2/g, *K is* the proportionality constant for the particle shape (spheres = 6, blocky and tabular = about 12), dW is the mass increment in grams, ρ is the density g/cm^3 and $\overline{X}_m$ is the average particle size of mass [72].

- the flowability of the powder – when measuring the flow rate in seconds, the powder with the standard mass (50g) was required to flow through the funnel hole with the diameter of 5 mm, (as the powder did not flow through a 2.5 mm hole, a 5 mm hole was used instead) and the relative measure was used; the Hall flowmeter was applied according to standard ASTM B213 (Fig. 2.7).

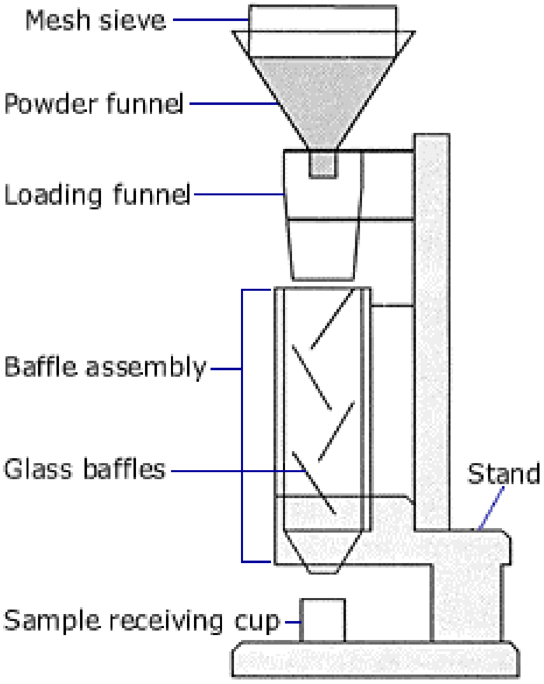


Figure 2. 7. The Scott volumeter

2.4. Characterization of composite plastics

New composite plastic materials were developed from PMMA powder materials by experimental manufacturing technology and their mechanical and technological properties were under the study.

2.4.1. Porosity

Porosity has a large effect on properties. Strength tends to be one of the least sensitive monitors of the porosity or microstructure. Ductility is more sensitive with the impact, fracture and fatigue behaviour having the greatest sensitivities [71].

To study the porosity of the cast composite material, specimens (50 mm length, 50 mm width, and thickness 10 mm) were made. The surfaces of the specimens (top, bottom and cross section) were ground and polished. The images were analyzed by using *Image-Pro Plus* 3.0. Firstly, the surface areas of the matrix and the pores were calculated.

2.4.2. Determination of mechanical properties

The details of ASTM, ISO and IEC test methods to determine the mechanical and physical properties of plastics are given in the different sources [83, 84].

Tensile test

Tensile test of composite plastic materials was performed according to standard ISO 527-1:1993(E) [27, 84, 99]. The mechanical properties, such as tensile strength, elongation, modulus of elasticity and hardness were determined. Tensile test of composite plastic materials was performed according to standard ISO 527-1:1993(E) [83, 84]. The following characteristic parameters were determined:

- tensile strength: R_m, N/mm^2

$$R_m = F_m / A_o \qquad (2.6)$$

where F_m stands for the maximum force and A_o indicates the initial cross-section area

- elongation after breakage: ε_B, %

$$\varepsilon_B = (A_0 - A_u) / A_o \cdot 100\%, \qquad (2.7)$$

where A_0 and A_u are initial and final cross-section areas

- modulus of elasticity: E, MN/mm^2

The tensile tests were performed under the following conditions:

- loading velocity, v = 5 mm/min, tolerance±20 %;
- test specimens without scratches and free of stress concentrators,
- the parallel opposite surfaces;
- type of the specimen, 1B ISO 527-2:1993 (E);
- thickness of the specimen, 4 mm.

The applied testing system was the servohydraulic testing machine *Instron* 8516, which uses both static and cyclic loading (Fig. 2.11, Table 2.3). In this system static tensile tests were carried out at the loading velocity of less than 30 N/mm^2 s^{-1} (EVS-EN 10002-1:1997).

Figure 2.11. Mechanical testing system Instron 8516

Table 2.3. Parameters of the testing system Instron 8516

Parameter	**Value**
Maximum load	100 kN
Stiffness of the frame	350 kN/mm
Position measurement with resolution	0.03% of measurement range
Position measurement accuracy	0.5% of measurement range
Resolution of force measurement	0.2% of measurement range
Force measurement accuracy	±0.05% 1KN if it exceeds the given measurement range
Deformation measuring accuracy	Class B or better by ASTM E83-90
Actuator clearance	+/– 75 mm

The test pieces for the tensile test were prepared according to ISO 527-2/1A/50 type1B (Fig. 2.12). The test pieces were cut out from the material of plastic composite plate and polished. The applied measuring device was the caliper and the measurement accuracy was 0.1mm.

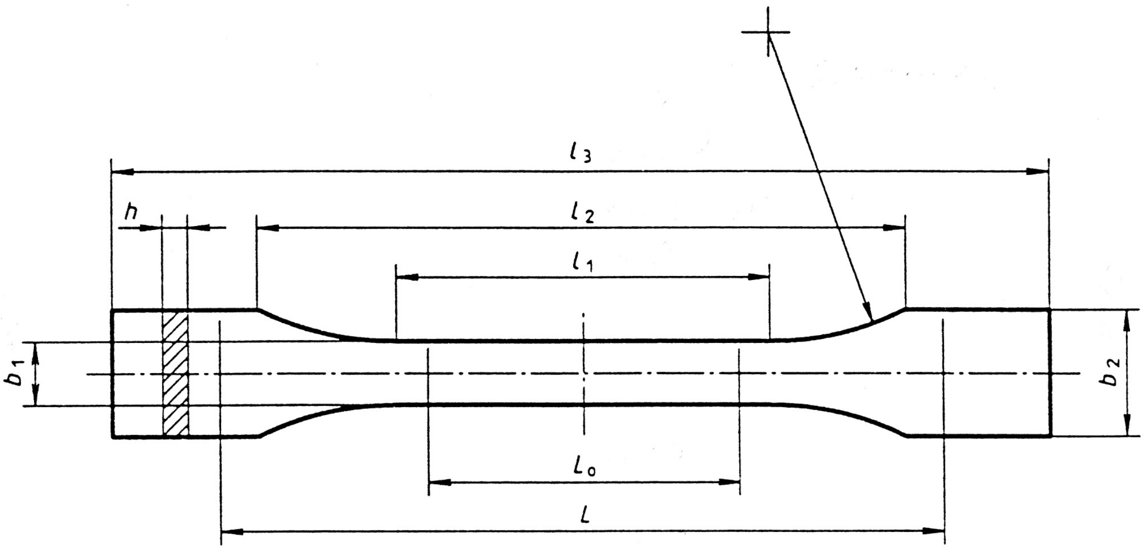
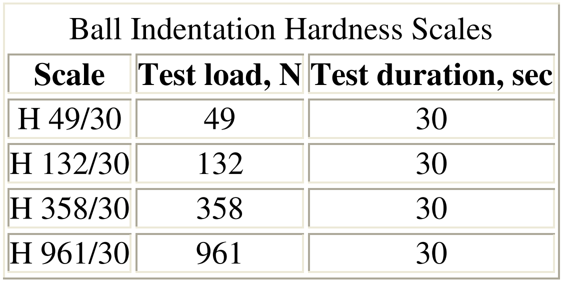


Figure 2.12. Type of the test piece of the tensile test 1B, (measurements of the test piece in mm: $l_3 \geq 150$; $l_1 = 60.0 \pm 0.5$; $l_2 = 106...120$; $b_2 = 20.0 \pm 0.2$; $b_1 = 10.0 \pm 0.2$; $h = 4.0 \pm 0 \pm 0.5$; $L = 115 \pm 0.2$; $L_0 = 50 \pm 1$)

Hardness test

When measuring the hardness of composite materials, the Ball Indentation Hardness method and standard ISO 2039-1 are used [86]. A 5 mm diameter steel ball is forced into a specimen with an initial load of 9.8 N ± 1% and it is increased to a specified load (Table 2.5) for 30 seconds. The depth of the resulting deformation is measured and the surface area of the impression is calculated. The hardness is then expressed as a load in N divided by the surface area of the indentation in mm^2. The choice of the main load depends on the depth of indentation (0.15-0.35 mm). If the preset qualifications are not met, a new main load will be chosen. The measurements of the recommended test piece are 50x50 mm. The minimum number of the measured values is 10. The thickness of the material is recommended to be about 10 mm.

Table. 2.4. Ball indentation hardness [84, 86]



Ball Indentation Hardness Scales

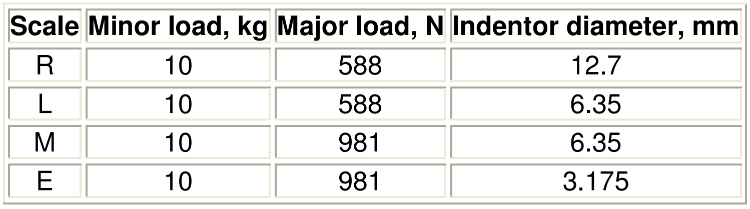
Scale	Test load, N	Test duration, sec
H 49/30	49	30
H 132/30	132	30
H 358/30	358	30
H 961/30	961	30

As the ball indentation hardness method was not available, the hardness tests were performed according to the Brinell hardness test method for metals, EVS-ENISO 6506-1 [87]. The Brinell scale characterizes the indentation hardness of materials through the scale of penetration of an indenter, loaded on a material test-piece. It is one of the several definitions of hardness in the materials science. The typical test uses a 10 mm- diameter steel ball as an indenter with a 29 kN force. For softer materials, a smaller force is used; for harder materials, a tungsten carbide ball is substituted for the steel ball. The indentation is measured and the hardness calculated as:

$$HBS = \frac{2F}{\pi D(D - \sqrt{(D^2 - d^2)})} \tag{2.6}$$

where F *is* an applied force in N, D is the diameter of an indenter in mm and d is the diameter of indentation in mm. The Brinell hardness test of indentation was performed with a 1 mm diameter steel ball and the applied loads were 49 and 98 N, the loading time was 30 seconds.
To compare the results of the Brinell hardness test, the Rockwell hardness test – ISO 2039-2 [88] on plastic materials was made. The Rockwell hardness is a measure of the indentation resistance of the material. Testing is performed first by forcing a steel- ball indentor into the surface of a material by using a specified minor load. The load is then increased to a specified major load and then decreased back to the original minor load.

Table. 2.5. Rockwell indentation hardness [88]



Scale	Minor load, kg	Major load, N	Indentor diameter, mm
R	10	588	12.7
L	10	588	6.35
M	10	981	6.35
E	10	981	3.175

The Rockwell hardness is calculated by subtracting the penetration divided by the scale division from 130.

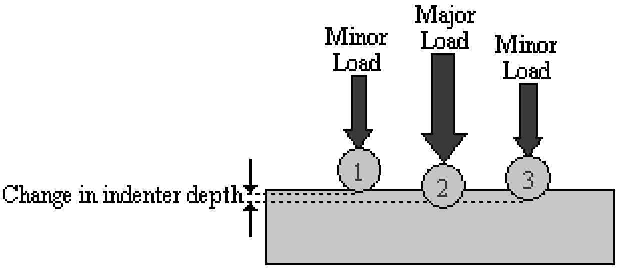


Figure 2.13. The schematic diagram of measuring the Rockwell hardness [88]

Rockwell hardness is a measure of the net increase in depth of the indentor (see Fig. 2.13). Each Rockwell scale division represents a 0.002 mm (0.00008 in) penetration of the indentor.

$$HRE = 130 - \frac{h}{0.002} \tag{2.7}$$

2.4.3. Abrasive wear resistance

The standard methods of testing the resistance of transparent plastics to surface abrasion [89, 90] and abrasion resistance of organic coatings by the Taber Abraser [91] were not available. The evaluation of abrasive wear resistance was performed according to the standard testing method of measuring abrasion by using the dry sand/rubber wheel apparatus [92].

This test method covers laboratory procedures for determining the resistance of metallic materials to a scratching abrasion by means of the dry sand/rubber wheel test. The dry sand/rubber wheel abrasion test involves the abrading of a standard test specimen with the grit of controlled size and composition. The abrasive is introduced between the test specimen and a rotating wheel with the chlorobutyl rubber tire or rim of a specified hardness [93]. This test specimen is pressed against the rotating wheel at a specified force by means of a lever arm while a controlled flow of grit abrades the test surface. The rotation of the wheel is performed so that its contact face moves in the direction of the sand flow. The specimens are weighed before and after the test and the loss is recorded by mass.

The rubber wheel abrasion test was performed according to standard ASTM-G-65-94 [92]. A schematic diagram of the test apparatus is shown in Figure 2.14.

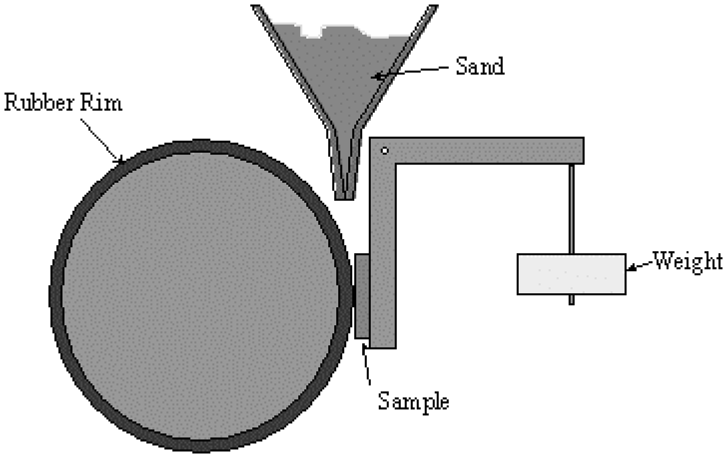


Figure 2.14. The schematic diagram of the original ASTM G65 test system [93]

The tests were conducted at the room temperature in the air. The test procedure consisted of installing the specimen into the holder, running the test and removing the specimen from the holder. All test variables are presented in Table 2.6.

Table 2.6. Test parameters of the rubber wheel

Parameter	**Value**
Rubber wheel: original diameter, mm width, mm	Rubber on steel wheel 45 229 12
Abrasive: grain size, mm mass, kg	Quartz sand 0.01-0.30 0.5
Normal force, N	140
Rotation speed	60

3. REPROCESSING OF PLASTIC AND MULTI-MATERIAL SCRAP

3.1. Reprocessing of the composite plastic PMMA+GFP

The composite plastic plates of PMMA+GFP preliminarily cut into pieces with the dimensions of length 100 mm, width 100 mm and thickness 5 mm, were reprocessed by the mechanical method of milling by collision.

The reprocessing technology of the composite plastics in disintegrators consisted of three stages:

- preliminary milling of the composite plastic PMMA+GFP by the DSA-158 disintegrator in the conditions of direct milling (sieving was used to separate the glass fibre from the milled material);
- intermediate milling for the size reduction in the DSA-2 disintegrator in the conditions of multi-stage milling (powder samples for sieve analyses were taken and percentage of the separated glass fibre was determined);
- final milling to remove the glass fibre from the milled material was performed by the DSL-115 disintegrator system applying the direct or/and separative milling conditions

3.1.1. The grindability study of composite plastic

The preliminary size reduction of the composite plastic PMMA+GFP and the separation of the components in the disintegrator mills were separative operations. In our case the scheme of milling was the following: PMMA in the form of fine particles and coarse GF particles were separated by sieving. The particle size of the output in the DSA-158 disintegrator was approximately 13-25 mm. The material preliminarily crushed was suitable for direct milling in the DSA-2 disintegrator. To estimate grindability, the specific energy of treatment was used. The distribution of the particle size was described by the modified Rosin-Rammler distribution function. The results of the grindability study are given in Fig. 3.1.

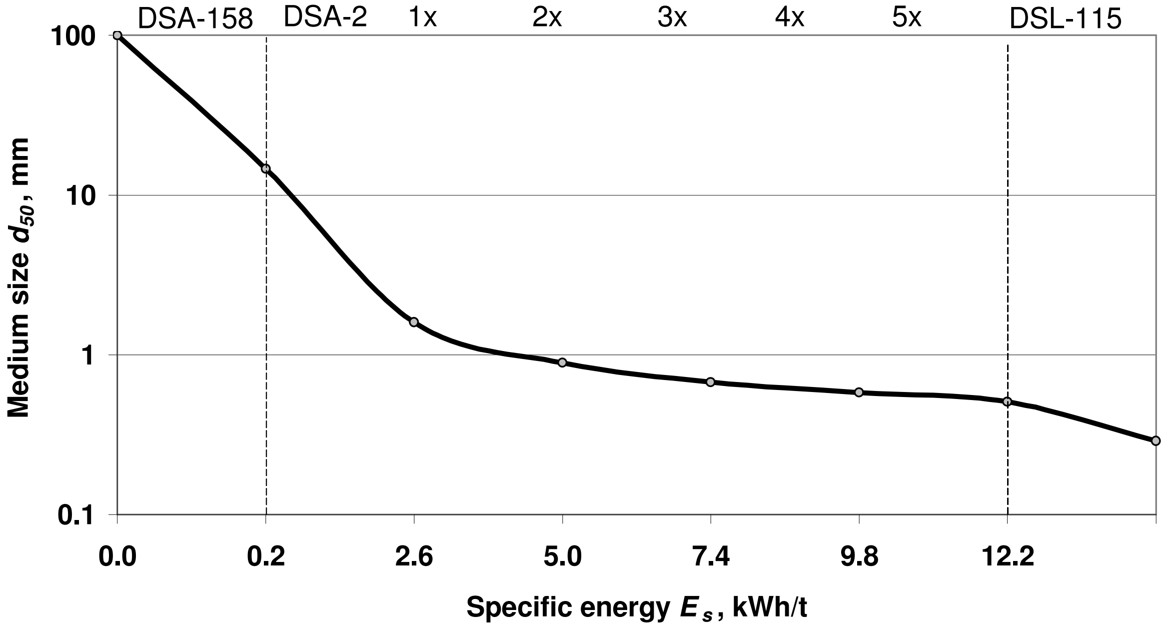


Figure. 3. 1. Dependence of the particles size of the milled composite plastic PMMA+GF on the specific energy of treatment.

As it follows from Figs 3.1 and 3.2, an intensive size reduction (from 100 mm to 13 mm) takes place by the preliminary milling in the DSA-158 disintegrator and by milling in the DSA-2 disintegrator (after the first two stages, a substantial size reduction takes place from 13 to 1 mm). The final milling in the DSL-115 disintegrator in the direct milling conditions at the double collision velocity reduced the size by 50 percent (from 0.71 mm to 0.35 mm) (Fig. 3.3).

Figure 3.2. The composite product PMMA+GF, pre-crushed in DSA-158

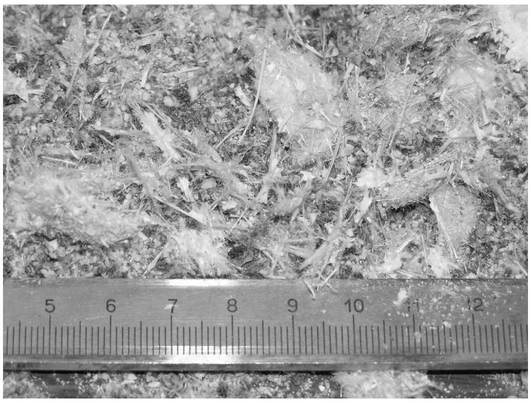


Figure 3.3. The product PMMA+GF milled in DSL-115

To compare the results of milling in the disintegrator devices with the existing advanced technologies for the size reduction, the machines of the company Weima are chosen. Their horizontal shredders with one rotor give the screen size of 15-100 mm, the shredders with four rotors give the screen size of 20-40 mm and the granulators give the screen size of 3-12 mm. The simultaneous separation of the composite components is not possible with shredders or granulators.

3.1.2. Separation of the components of composite plastic

The results of the separation of glass fibre and PMMA acrylic plastic are presented in the following charts (Figs 3.4 and 3.5).

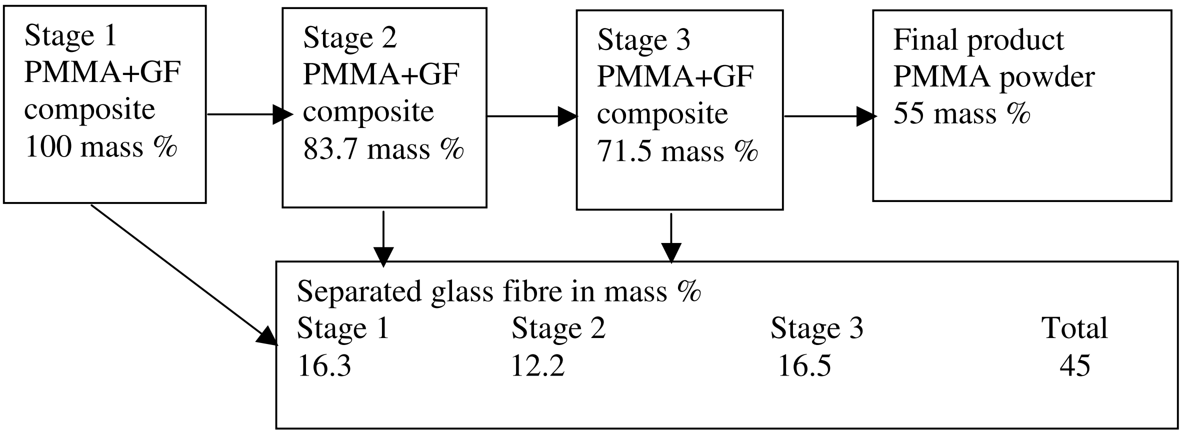


Figure 3. 4. The results of separation in a three-stage milling: stage 1 – the equipment used is DSL-158, stage 2 – the equipment used is DSA-2, stage 3 – the equipment used is DSL-115.

Before separation, the curve of the mixed plastics (PMMA+GFP) had two modes. After separation, acrylic plastic had two main fractions: particles of 2.1 mm – 47 mass % and 0.53 mm – 25 mass %; the main fraction of glass fibre was 0.27 mm in diameter and 10-15 mm in length (85 mass %).

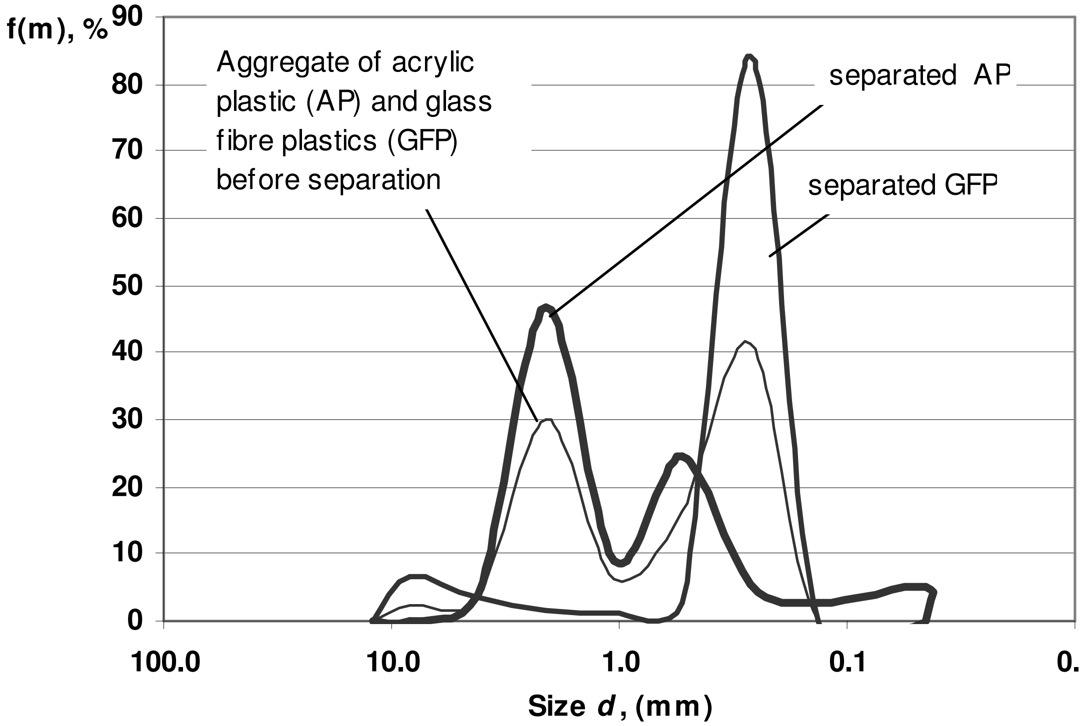


Figure 3.5. Size distribution of the particles size of the separated acrylic plastic powder (AP) and glass fibre (GF)

3.1.3. The granularity and morphology study

The study of the particle size and shape of the PMMA powder particles was performed by the sieving analysis (SA) and image analysis (IA). The data necessary in the particle size study by the image analysis were obtained using the image processing system, which consisted of the Nikon *Microphot-FX* optical microscope and a video transferring system. The measurements were performed in the transmission regime of OM, because it gave more accurate measurements of the particles size than the reflected regime. The size and shape parameters were studied by the image analysis system *Image-Pro Plus* 3.0.

AS the particles size of the PMMA powder varied on a large scale, the powder was classified by sieving to 7 fractions: ≤ 0.315; 0.315-0.63; 0.63-1.25; 1.25-2.5; 2.5-5.0; 5.0-11.2 and >11.2 mm and these fractions were analyzed separately. The results of IA (mean particle size by volume, d_m^v) are given in Figs 3.6–3.12 and Table 3.1

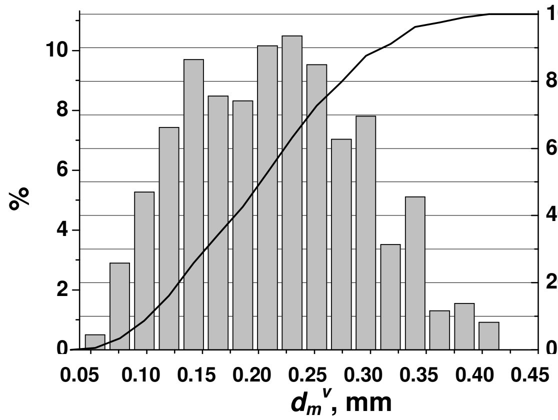


Figure 3.6. Particles size distribution of fraction less than 0.315 mm

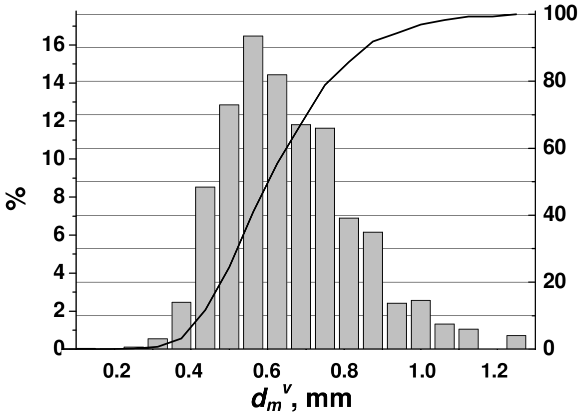


Figure 3.7. Particles size distribution of fraction 0.315 mm to 0.630 mm

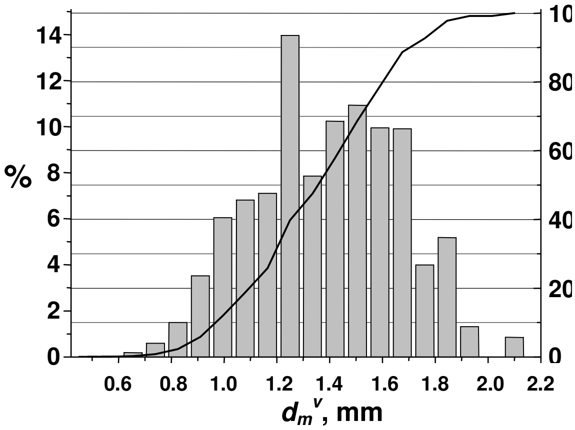


Figure 3.8. Particles size distribution of fraction 0.63 mm to 1.25 mm

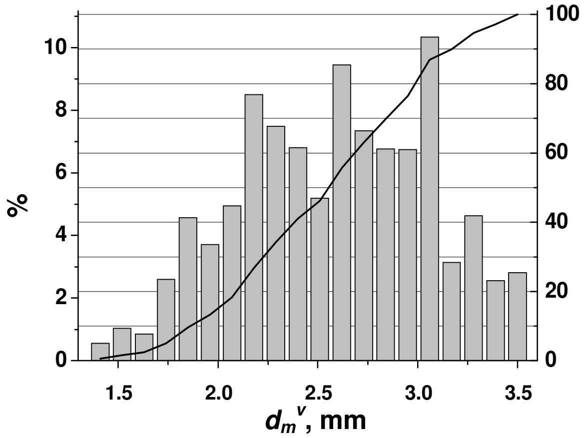


Figure 3.9. Particles size distribution of fraction 1.25 mm to 2.50 mm

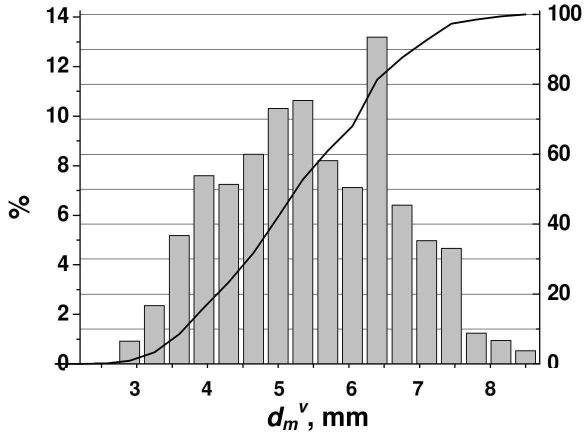


Figure 3.10. Particles size distribution of fraction 2.5 mm to 5.0 mm

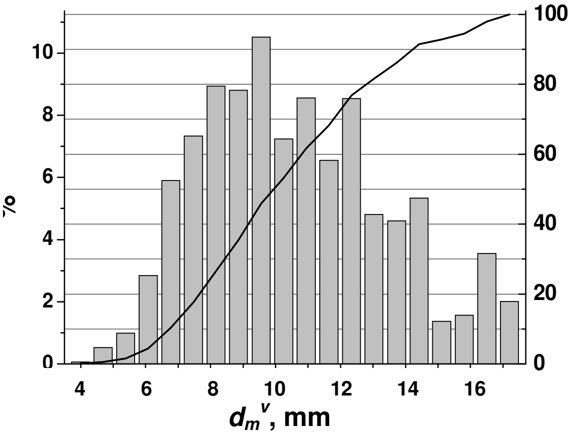


Figure 3.11. Particles size distribution of fraction 5.0 mm to 11.2 mm

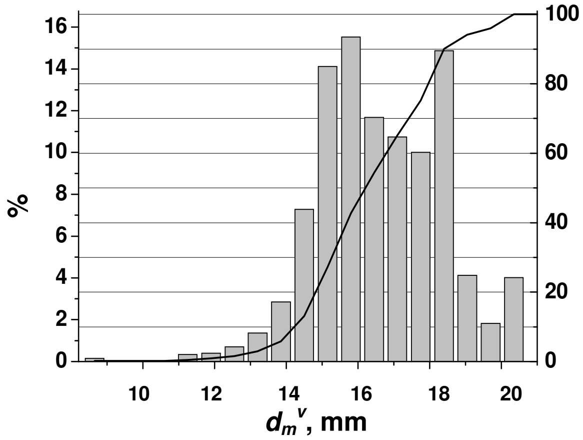


Figure 3.12. Particles size distribution of fraction more than 11.2 mm

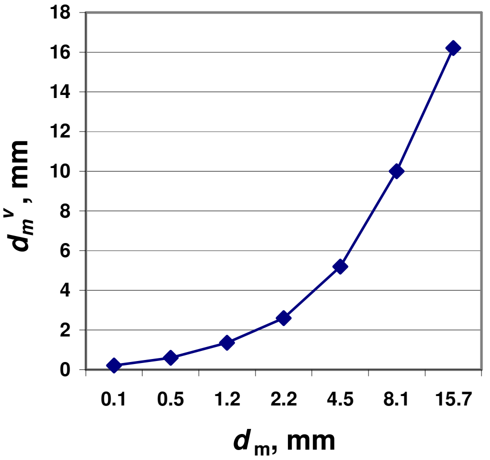


Figure 3.13. Mean particles size by volume d_m^v, mm of fractions 0 to 11.2 mm

The acrylic plastic powder particles of the medium size, less than 5 mm, are mainly equiaxed and their mean aspect and roundness are measured only flatwise. The plastic powder particles whose size is more than 5 mm are lamellar in shape, their aspect and roundness are calculated as an arithmetical average of the two images (flatwise and sidewise). For example, the particles of the medium size between 5-11.2 mm have the aspect flatwise 1.54 and the aspect sidewise 3.25, which make the average aspect 2.40 (see Figs 3.14 and 3.15).

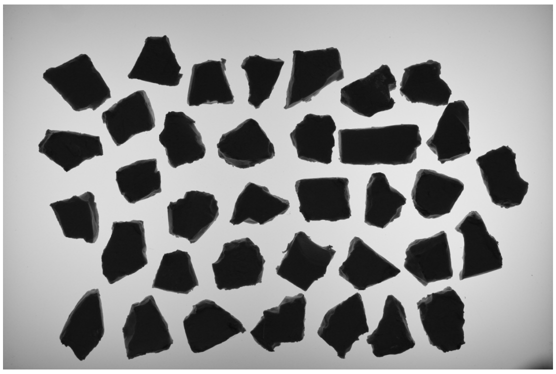


Figure 3.14. Image of the particles

Figure 3.15. Image of the particles

Table 3.1. Diameter, aspect and roundness of the acrylic plastic powder fractions

Fraction and share	**< 0.315 mm 3 %**	**+ 0.315 – 0.63 mm 7 %**	**+ 0.63 – 1.25 mm 14 %**	**+ 1.25 – 2.5 mm 27 %**	**+ 2.5 – 5.0 mm 35 %**	**+ 5.0 – 11.2 mm 10 %**	**> 11.2 mm 4 %**
d_{50}, mm	0.16	0.47	0.94	1.88	3.75	8.10	12.10
d_m, mm	0.11	0.49	1.16	2.20	4.45	8.08	15.70
d_m^v, mm	0.21	0.60	1.36	2.60	5.20	10.00	16.20
Aspect, *AS*	2.07	1.91	1.71	1.61	1.55	2.40	3.01
Roundness, *RN*	1.63	1.69	1.79	1.71	1.44	1.77	2.00

As it follows from Table 3.1, the mean particle sizes obtained by IA and SA methods differ more for coarse fractions (1.25-13 mm) than fine (0-1.25 mm) fractions. The main reason for this is the shape of the particles. The flat-shape particles of coarse fraction are stacking on the sieve mesh, even though it is bigger than the area of the particles cross-section. It can be explained by the higher values of the two shape parameters, the aspect *AS* (1.6-3.0) and the roundness *RN* (1.7-2). For fine fractions there is a possibility of passing the sieve mesh, which is smaller than their length.

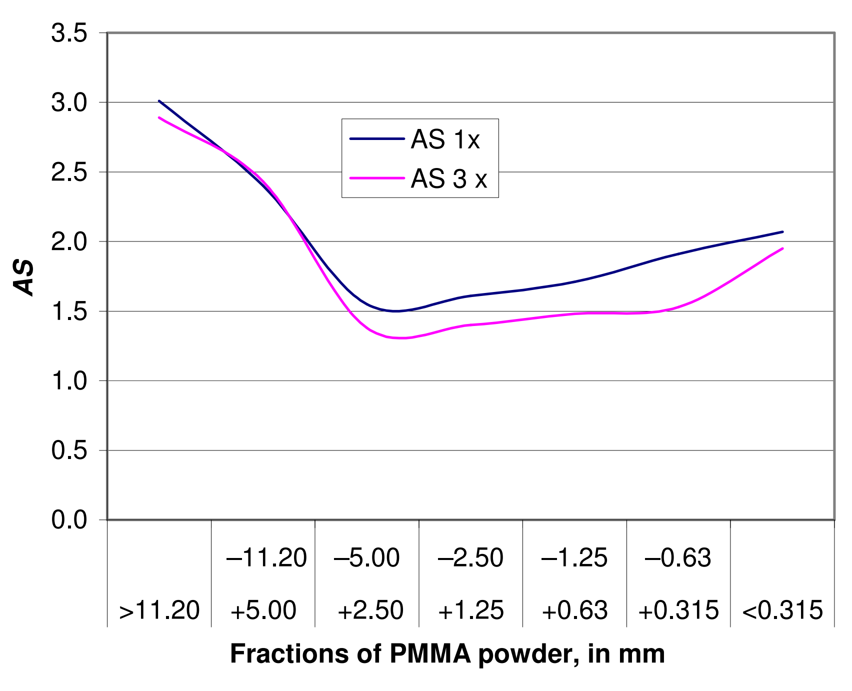


Figure 3.14. Dependence of the aspect *AS* on the PMMA powder particle in multiplicity of milling in DSA-2 (1x and 3x).

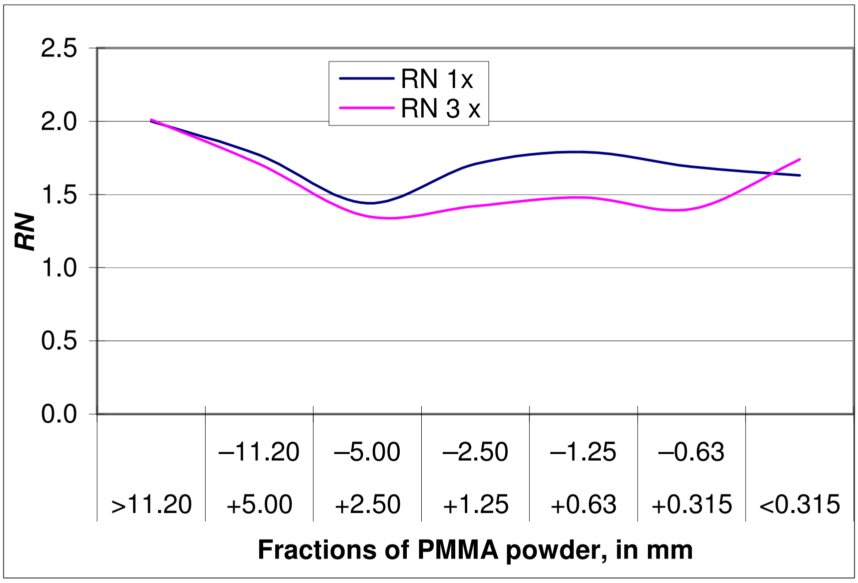


Figure 3.15. Dependence of the roundness *RN* on the PMMA powder particle in multiplicity of milling in DSA-2 (1x and 3x).

The morphology parameters of the milled PMMA powder were analysed again after five times of milling in the DSA-2 type of disintegrator and the final separative milling in the DSL-115 type of disintegrator. After SA it was clear that the particles size of the PMMA powder varied within a smaller range (0-2.5 mm) and the IA of the powder can be made without classifying it by sieving to separate fractions.

The particle morphology of the acrylic plastic powder showed that the roundness parameter is in the range of 1.31-1.32 (see Fig. 3.19 and Table 3.2) and the specific energy of treatment is 12.2 kWh/t for the powders with the mean particle diameter of about 0.55 mm; with the mean particle diameter of 0.3 mm, the specific energy of treatment is 62.2 kWh/t.

The mechanism of the fracture of the PMMA material was the same in direct milling and selective milling, because the aspect and the roundness parameters of the particle changed a little.

Table 3.2. Granularity and morphology parameters of the milled PMMA powder particles

Parameters	**Specific energy of treatment E_s, kWh/t**	
	12.2	**62.2**
Granularity:		
- main fraction, mm (70%)	+0.35 – 0.7	+0.18 – 0.35
- d_m, mm	0.55	0.30
- d_m^v, mm	0.82	0.39
Morphology:		
- Aspect *AS*	1.39	1.37
- Roundness *RN*	1.31	1.32

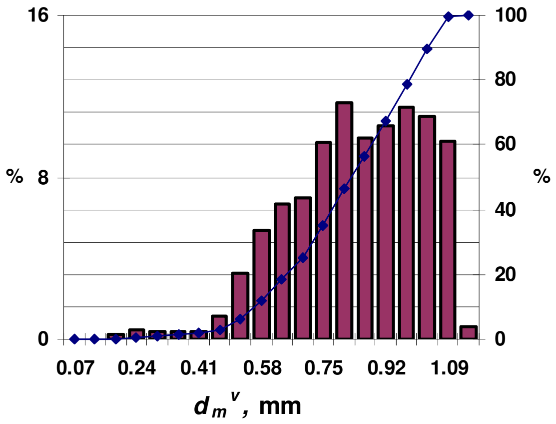


Figure 3.16. Particle size distribution after 5x milling in DSA-2

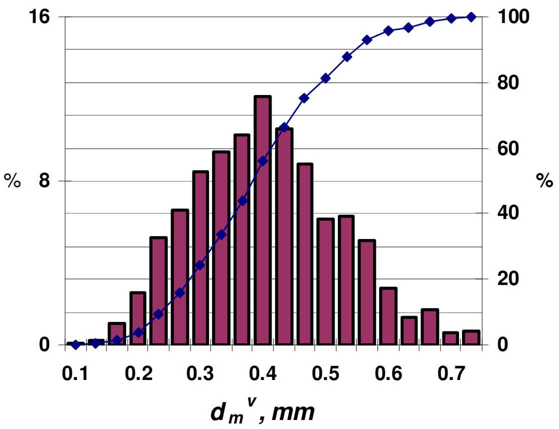


Figure 3.17. Particle size distribution after final selective milling in DSL-115

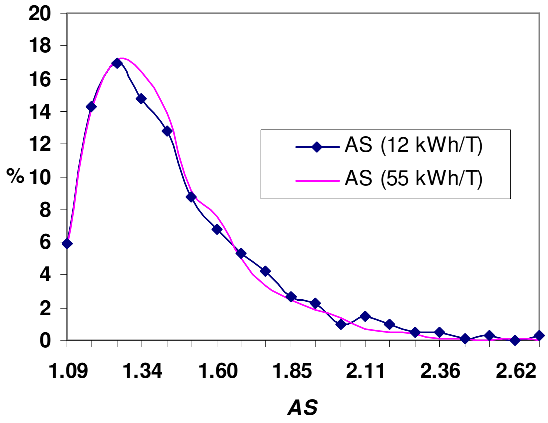


Figure 3.18 Percentage distribution of the aspect *AS* of the milled material at a different specific energy of milling

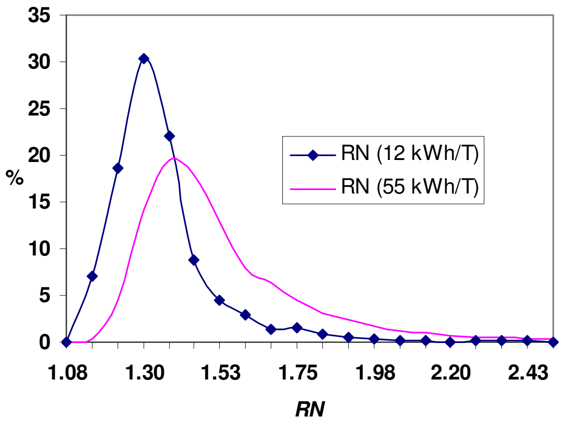


Figure 3.19 Percentage distribution of the roundness *RN* of the milled material at a different specific energy of milling

The comparison of the particle size distribution after five times of milling in the DSA-2 disintegrator and after the final milling in DSL-115 shows (Figs 3.16 and 3.17) that the mean diameter of the particle by volume $d_m{}^v$ has diminished two times (from 0.82 to 0.39 mm). Comparing the percentage distribution curves of the PMMA particles morphology characteristics *AS* and *RN* (Figs 3.18 and 3.19), only slight changes in the average value can be observed (see Table 3.2).

The percentage distribution curves of the PMMA particle aspect *AS* are overlapping. The percentage distribution of the roundness *RN* after the final milling in DSL-115 has smoothed away and the median value is more different (*RN* 1.31-38) than that calculated by IA analyzing program (RN 1.31-1.32).

3.1.4. Technological properties of the produced plastic powder

Density

From the technological point of view, the data about the density and flowability of the plastic powder are important, so the apparent density, tap density and flowability of the milled product –the plastic powder – were determined. The data about the tap and apparent density are necessary for the further design of a new composite material. The PMMA powder as a filler material with a higher apparent density in mixture with the binder agent saves the cost of the product by reducing the content of the binder agent, and thus affecting the properties of the composite. The results of the density measurements of the PMMA powder are given in Fig. 3.20.

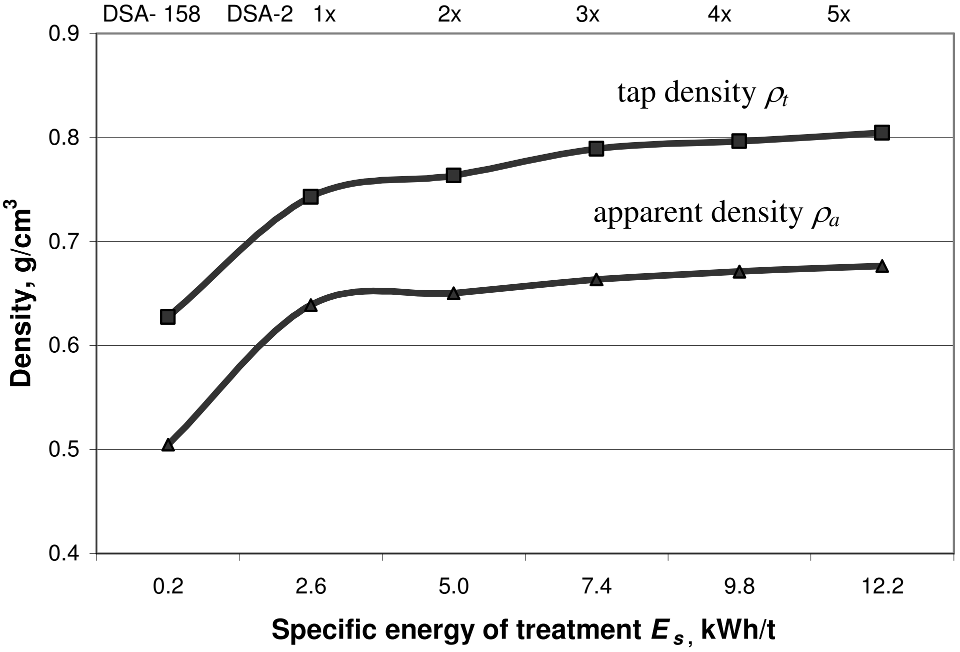


Figure 3.20. Dependence of the density of the PMMA milled powder on the specific energy of treatment.

As it follows from Fig. 3.20, the density of PMMA powder increases remarkably after preliminary milling in the DSA-158 disintegrator and by milling in the DSA-2 disintegrator (after the first three stages) and changes only little after final milling in DSA-2. This is similar to the medium particle size distribution (Fig. 3.1). The tap density of the milled PMMA powder is about 20 percent higher than the apparent density.

Relative flowability

To measure the relative flowability of the milled powders after the multi-stage milling in DSA-2, the powder was classified by sieving to 4 fractions mm: ≤ 0.315; 0.315-0.63; 0.63-1.25; 1.25-2.5 depending on the used funnel (with a 5.8 mm diameter of a hole).

The relative flow-rates of the plastic powder fractions were the following: 23 seconds for ≤ 0.315; 29 seconds for 0.315-0.63; 25 seconds to 32 seconds for 0.63-1.25; the flow rate for the 1.25-2.5 could not be measured.

The flow-rate of the PMMA powder after the five-stage milling in DSA-2 was 42 seconds and the flow-rate of the finally milled powder in the DSL-115 was 21 seconds.

3.2. Reprocessing of compounded plastic scrap of personal computers

The compounded plastics of the WEEE- personal computers whose share in monitor housings is 21 mass %, in processors 4 mass %, in keyboards 79 mass % (see Figs 3.25-3.27) were under the study. The compounded plastics (PC-ABS) of processors, monitors and keyboards from personal computers were cut into pieces and then preliminarily milled for the feed of DSA-158. The multi-stage milling (up to 16x) of the compounded plastics was performed in the DSA-2 disintegrator. After each step the samples for the sieve analyses were taken.

The results of the preliminary and final milling of the compounded plastics from different parts of computers are given in Table 3.3. The photos of different pre-crushed and milled compounded plastics are given in Figs 3.23 and 3.24.

Table 3.3. The medium particles size of the compound plastic powder

Type of disintegrator and milling step	**Multiplicity of milling**	**Energy of treatment kWh/T**	**PC+ABS monitors**	**ABS processors**	**ABS+HIPS keyboards**	**PS+ABS+PPE monitors**
			Medium size d_{50}, mm			
primary material	-	0	100	100	100	100
DSA-158 precrushing	2	1.4	12	9.9	15	9.7
DSA-2 preliminary milling	1	3.8	7.5	-	-	-
	2	6.2	6.2	-	-	-
	4	11.0	5.3	-	-	-
	8	20.6	4.1	4.1	6	4
	12	30.2	-	3.9	4.3	3.8
	16	39.8	3.6	3.8	3.6	3.6
DSL-115 final milling with separation	1	46.6	3.1	3.3	3	3
	2	53.4	2.6	2.8	2.8	2.8
	4	67.0	2.2	2.3	1.7	2.4

As for 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 2.1). After the fracture, the elongation of the PC-ABS plastics is exceeding five times that of the ABS plastics. These PC-ABS plastics were easy to cut with the guillotine shears for feeding the DSA-158 (100-150 mm). After the preliminary milling in the DSA-158, it was obvious that this material has a ductile fracture mechanism. The preliminary milled material was appropriate for the feed of the DSA-2. The multi-stage milling of the compounded material was performed by the DSA-2 disintegrator in the direct milling condition. The results of the preliminary size reduction of the compounded plastic PC-ABS in the disintegrator mill are shown in Fig. 3.21. As it follows from Fig. 3.21., the best results of the size reduction were achieved after the two times of preliminary milling in the DSA-158 when the mean particle size was about 11 mm, and after the third stage of milling in the DSA-2 when the particle size was 5 mm.

During the next stages of milling (from stage 4 to 8), the size reduction is less effective (from 5 mm to 4 mm). During the intermediate milling in the DSA-2 disintegrator, the size reduction of the material was less effective because of the plastic deformation of the particles after collision. Thus, for fracturing the particles of the ductile material, the number of collisions increases more than 10 times.

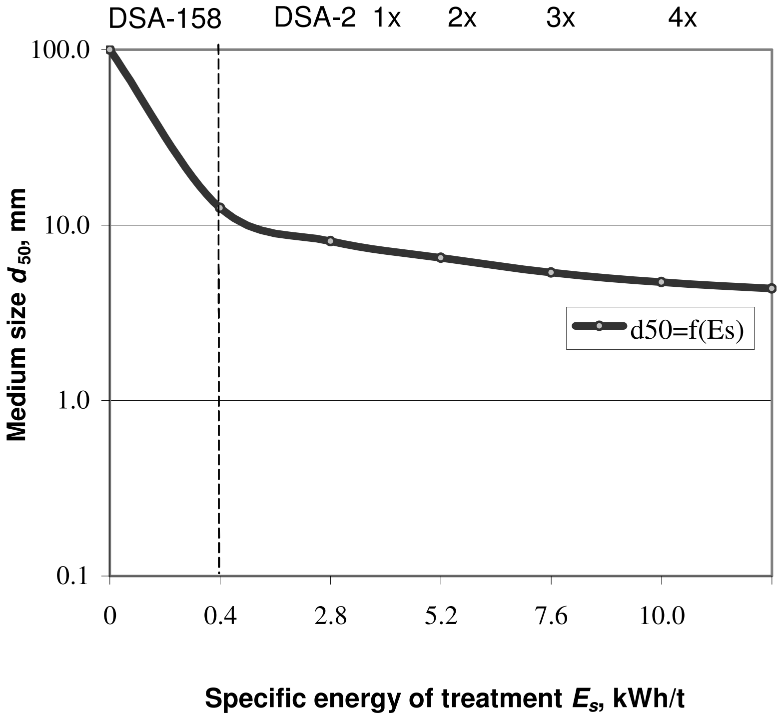


Figure 3. 21. The dependence of the medium particle size of the milled compound plastic PC-ABS on the specific energy of treatment.

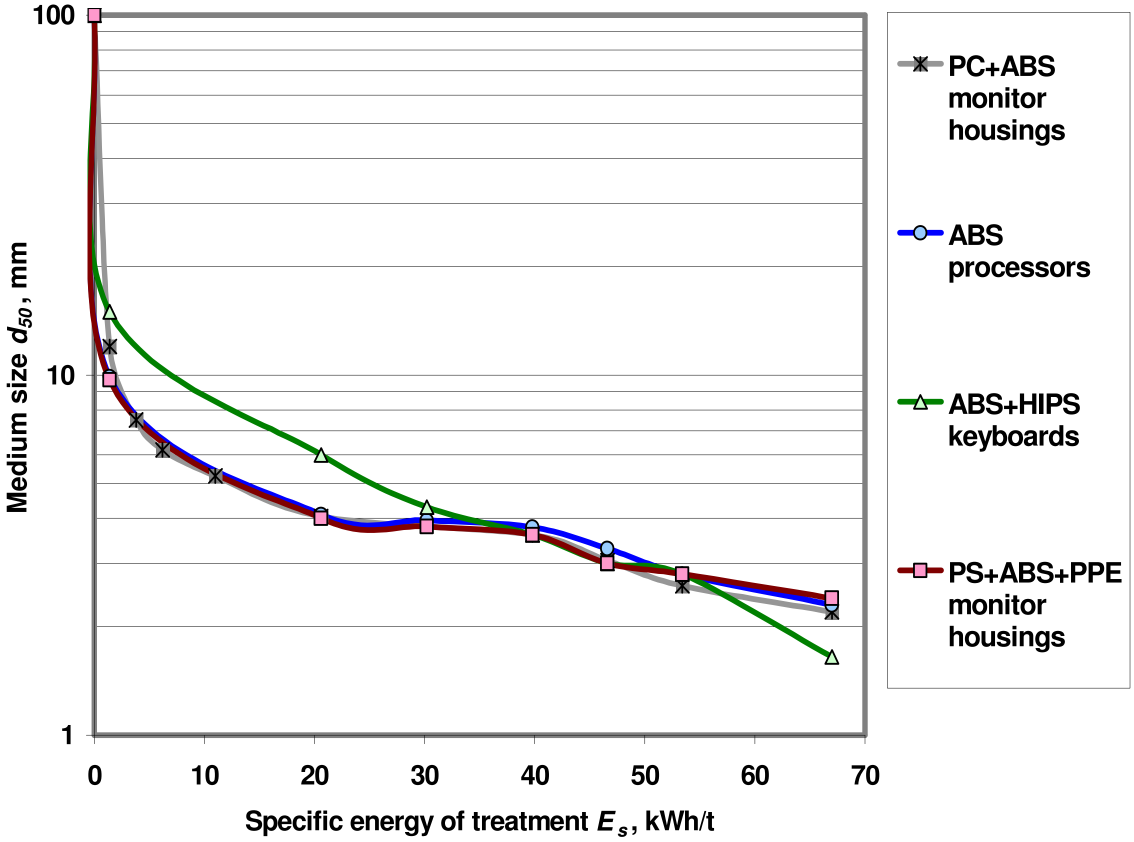


Figure 3.22. Dependence of the medium particles size on the specific energy of treatment of the compounded plastics.

As it follows from Table 3.3 and Figs 3.21-3.22, the best results of the size reduction were achieved after the two times of preliminary crushing in the DSA-158, when the mean particle size was about 9.7-15 mm (Figs 3.23); after the eight stages of preliminary milling in the DSA-2, the particle size was in the range of 4-6 mm (Fig. 3.24). The final milling in the DSL-115 disintegrator in the direct milling conditions reduced the size by 50 percent (1.7 to 2.4 mm).

Figure 3.23 A photo of ABS+PC precrushed plastic of monitors

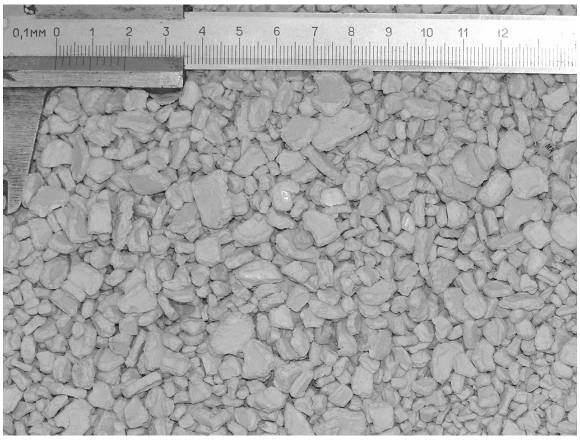


Figure 3.24. A photo of ABS+PC milled plastic of monitors

Table 3.4. The diameter, aspect and roundness of the milled PC-ABS powder fractions.

Fraction **Parameter**	**<0.315 mm**	**+ 0.315 – 0.63 mm**	**+ 0.63 –1.25 mm**	**+ 1.25 – 2.5 mm**
d_{50}, mm	0.16	0.47	0.94	1.88
d_m, mm	0.37	0.69	1.40	2.63
$d_m^{\,v}$, mm	0.46	0.74	1.70	3.60
Aspect, AS	1.67	1.62	1.55	1.52
Roundness, RN	1.57	1.53	1.43	1.32

As it follows from the table the roundness and aspect parameters of milled PC-ABC compounded plastics are increasing a little when size of particles is decreasing.

3.3. Reprocessing of the multi-material scrap of personal computers

The printed circuit boards (PCB) of personal computers were studied. As it follows from the diagrams of the sorted materials of dismantled personal computers (Figs 3.25-3.27), the printed circuit boards are forming 11.0 mass % of processors, 4.9 of monitors and 2.4 of keyboards. Printed circuit boards are mainly produced from thermosetting resin (epoxy or phenolic resin) and reinforced with fibres such as paper, wood, textile and glass (of high performance). FT-IR (Fourier Transform Infrared Spectrometer) spectra of the plastic pieces from the light fractions after the sink–float testing, show that PCB scrap mainly contains glass fibre reinforced by the plastics of epoxy resins with the density range of 1.5–2.0 g/cm^3 , the scrap of PCBs contains multi-elements: Al (2.8 mass %), Cu (10.0 mass %), Pb (1.2 mass %), Zn (1.6 mass %), Ni (0.85 mass %), Ag (280 ppm), Au (110 ppm) [79].

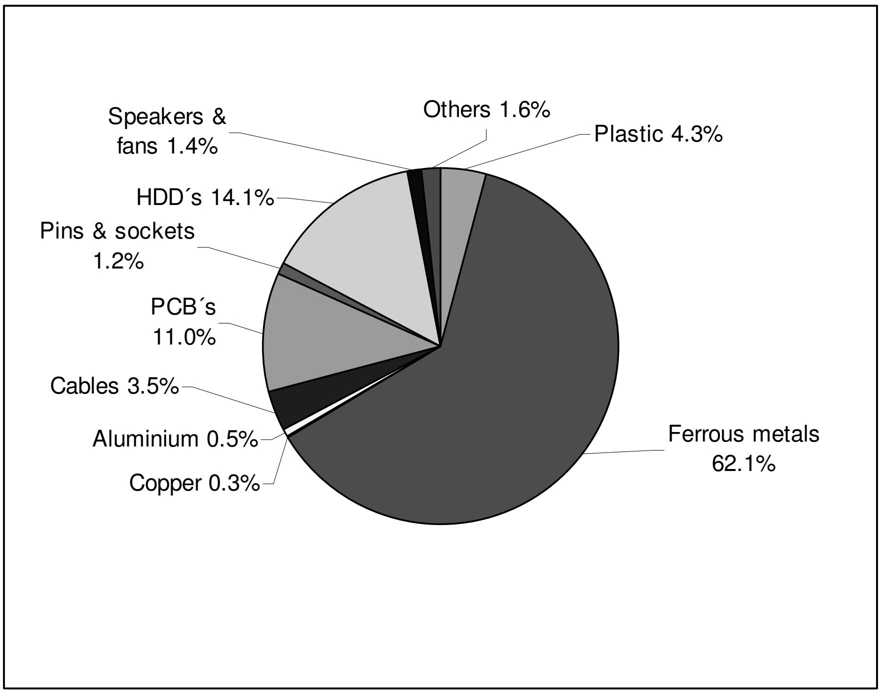


Figure 3.25. The sorted materials from the processors of personal computers (mass %)

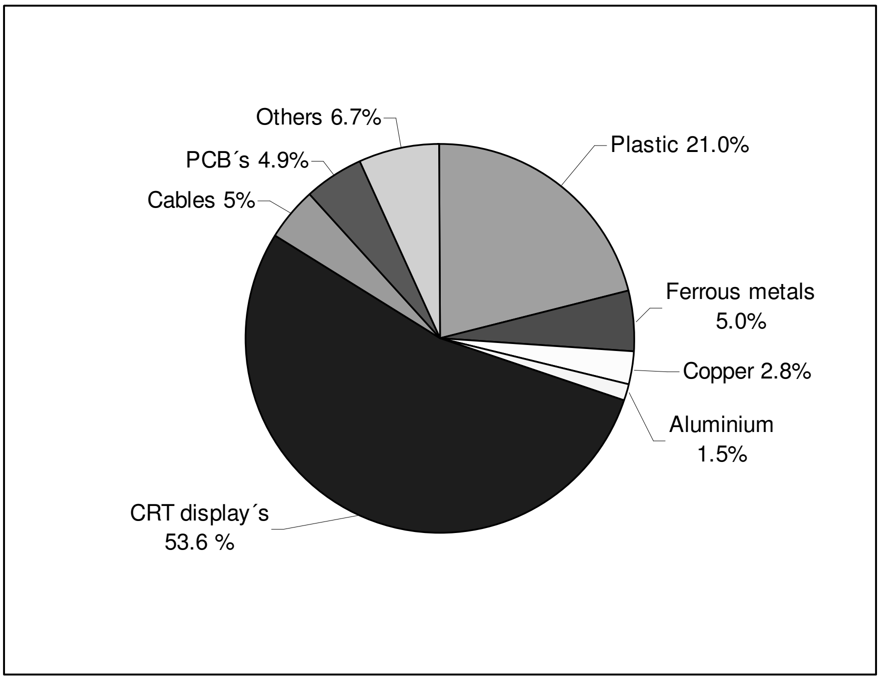


Figure 3.26. The sorted materials from the monitors of personal computers (mass %)

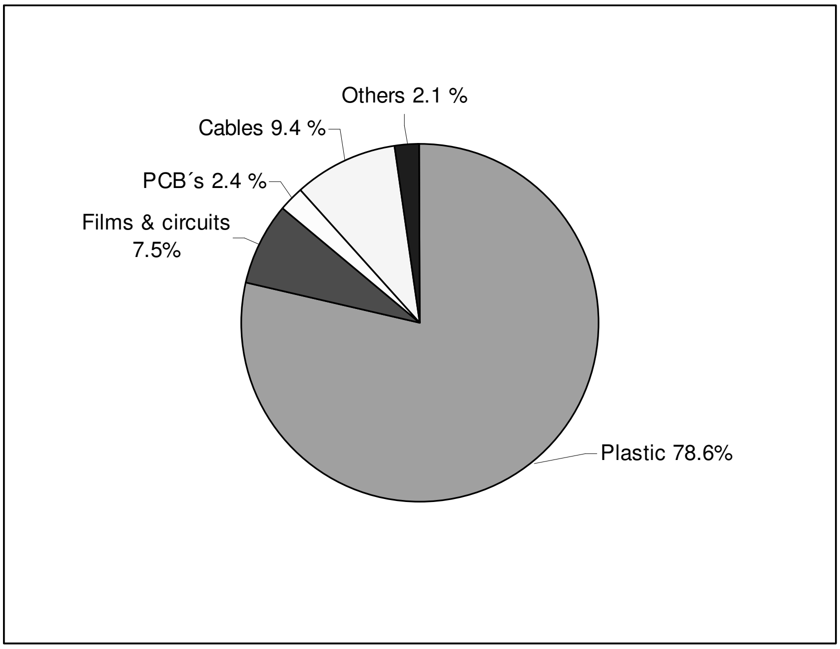


Figure 3.27. The sorted materials from the keyboards of personal computers (mass %)

The reprocessing technology of the printed circuit boards in disintegrators consisted of the following stages:

- the preliminary size reduction of the PCB plates by the experimental DSL-158 disintegrator (up to 2 times);
- the intermediate milling for the size reduction in the semi-industrial disintegrator DSA-2 (up to 6 times);
- the final milling by the DSL-115 disintegrator system in the selective milling conditions to separate the plastic and metallic components

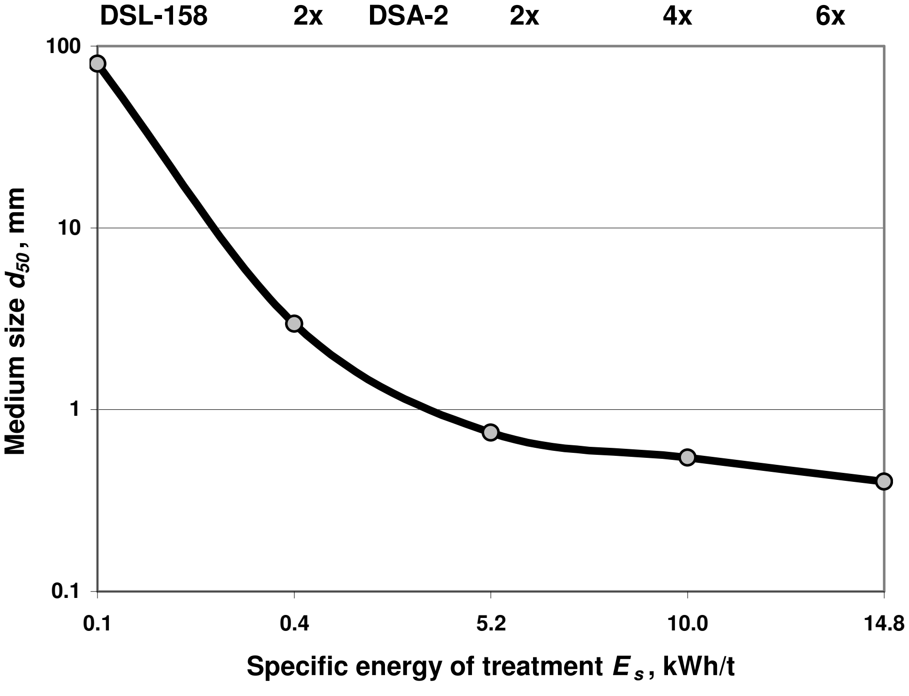


Figure 3.28. Dependence of the particles medium size of printed circuit boards on the specific energy of treatment

The results of the preliminary size reduction and intermediate milling are given in Figs 3.28 and 3.29. Similar to the reprocessing of the composite and compounded plastic scrap (see Figs 3.1 and 3.21), the particle size of the plastic component from PCB after a 2-stage milling is about 5-10 mm, after 1-2 times of milling in the disintegrator DSA-2 it is around 1 mm. The powder particles from PCB after the preliminary size reduction are mainly lamellar (Fig. 3.30) after preliminary milling and they stay lamellar after the milling in the disintegrator DSA-2.

Figure 3.29 The preliminarily milled PCBs from dismantled personal computers

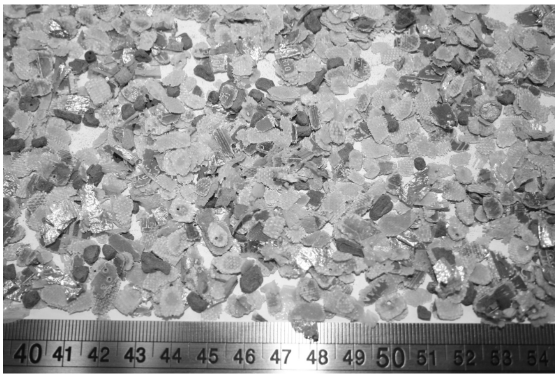


Figure 3.30. Separated composite plastic from the preliminarily milled PCBs

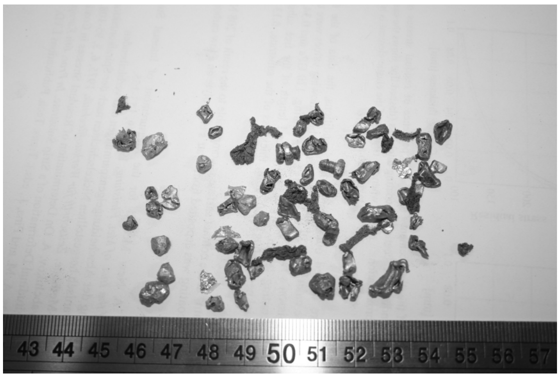


Figure 3.31. Separated metallic components from the milled PCBs

3.4. Conclusions

1. The prospective mechanical method – milling by collision – and the use of the disintegrator milling systems to obtain secondary raw material – plastic powder from the composite and compounded plastic scrap – is demonstrated. The reprocessing of the composite PMMA+GFP plastic scrap by using disintegrators in the separative and/or selective milling conditions will enable to produce the acrylic plastic powder with a determined granularity and technological properties (the apparent density and flowability).

2. The advantages of disintegrator milling in comparison with existing advanced technologies for the size reduction of the composite and compounded plastic scrap by using horizontal shredders and granulators are the following:
- simultaneous separation of the components of the composite (it is not possible with shredders and granulators)
- lower specific energy of treatment

3. From the reprocessing studies of the PMMA+GFP composite plastic scrap, the following conclusions can be drawn:
- as a result of the preliminary milling of the reinforced acrylic plastic wastes in DSA-158, about 1/3 of the glass fibre intact in the resin matrix is removable by sieves.
- as a result of the intermediate milling in DSA-2 under the multi-stage direct milling conditions the mean particle size was about 0.5-1 mm and about 25-30 mass % of glass fibre intact of the PMMA composite was removable by sieves;
- the one-stage final milling of the PMMA composite material by the disintegrator milling system DSL-115 in the selective milling conditions reduces the size of particles of the acrylic plastic powder about 2 times and leaves the content of the glass fibre as intact as the minimum.

4. The best results of reprocessing the compounded plastic scrap by disintegrators will enable a remarkable size reduction: after the two stages of preliminary crushing in DSA-158 the mean particle size was about 10-15 mm. During the intermediate milling in the DSA-2 disintegrator, the size reduction of the material was less effective because of the plastic deformation of the particles after collision. Thus, for the remarkable size reduction of the particles of the ductile compounded plastic, the number of collisions, i. e. the multiplicity of milling must be more than 10 times.

5. The reprocessing of the multi-material (plastic-metallic) scrap of personal computers, particularly, the printed circuit boards, is different at the first stage. Later on, the reprocessing does not differ essentially from the reprocessing of the composite and compounded plastic scrap. The separation of the plastic and metallic parts of the milled multi-material in the selective milling conditions needs an additional study to determine the optimum milling parameters and in the design of new classifiers accounting for the densities of plastic and metallic parts.

4. DEVELOPMENT OF NEW PRODUCTS ON THE BASIS OF PRODUCED PLASTIC POWDERS

4.1. Technological tests on the milled plastic powder used as the filler material in new composite plastic

For an ideal filler, the characteristics should include the following: a low cost, the availability, low oil absorption, a good wetting and bonding surface, a good chemical resistance, high strength (non-friable). In addition, if a complete manufacturing control were possible, the following properties would be desirable: colour, shape, density, modulus, size (and its distribution), thermal expansion and the electrical and thermal properties [72].

The most common method to describe the maximum packing systems of fillers is the continuous grading, which is the most common method to blend the fillers to obtain the minimum voids to be occupied by the binders. The system is actually based on the fact that the coarsest particle pack is to produce the gross volume of the system. As the finer material is added, it occupies the voids between the largest particles, so that the total volume is not expanded. Each additional finer material also has a void volume, which in its turn may be occupied by still a finer material. The resulting geometric progression then depends on the least voids left in the system and a very wide range of the particle sizes to achieve a high degree of packing [72].

4.1.1. Density of the mixed filler materials

The aim of the experiments was to maximise the density of the filler material. For this purpose six fractions (size in mm ≤ 0.315; 0.315-0.63; 0.63-1.25; 1.25-2.8; 2.8-5.6) of the PMMA powder were used to prepare the mixtures of the filler materials. At first, we measured the apparent density of each fraction. Then two different fractions were mixed at a time with the mixing ratio 50/50. By using only one fraction of the PMMA powder, the best apparent density 0.64 was measured for the fraction (5.6-2.8 mm). By mixing the two different fractions of the PMMA powder, the filler consisted of 50 mass % of coarse fractions (5.6-2.8 mm) and 50 mass % of fine fractions (≤ 0.315 mm) and thus, had the best apparent density 0.75 (see Fig. 4.1). The filler material consisted of 50 mass % of coarse fractions (1.25-2.8 mm) and 50 mass % of fine fractions (≤ 0.315 mm) had the best tap density.

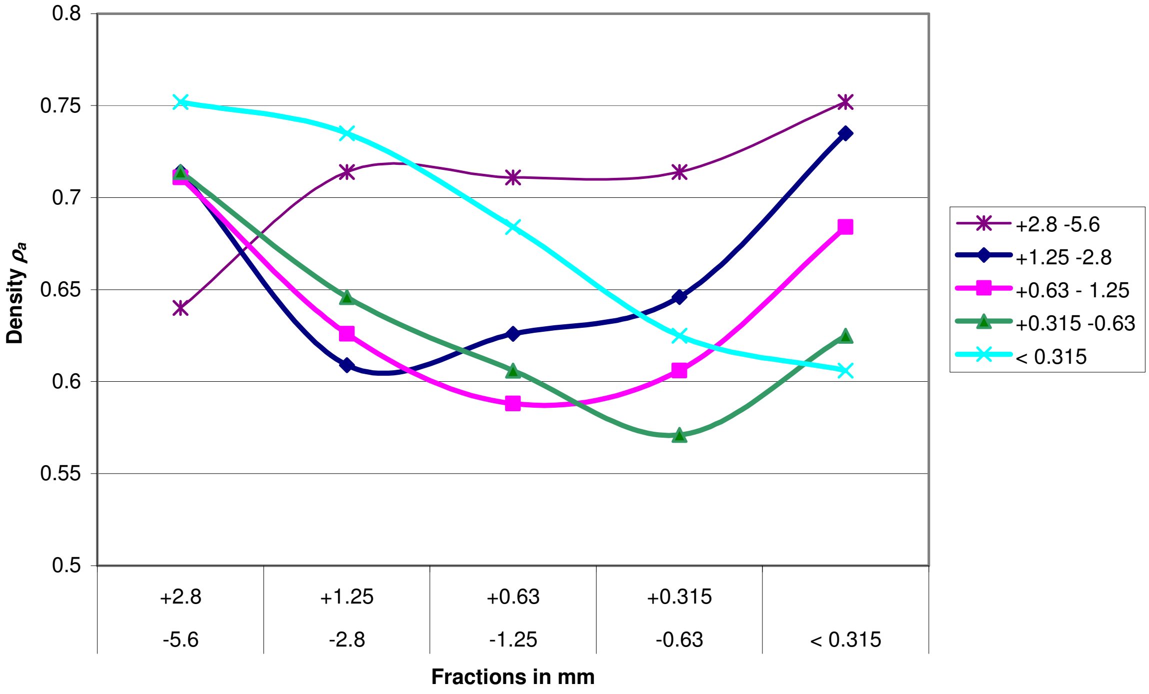


Figure 4.1. The apparent density of the mixed filler materials of different fractions

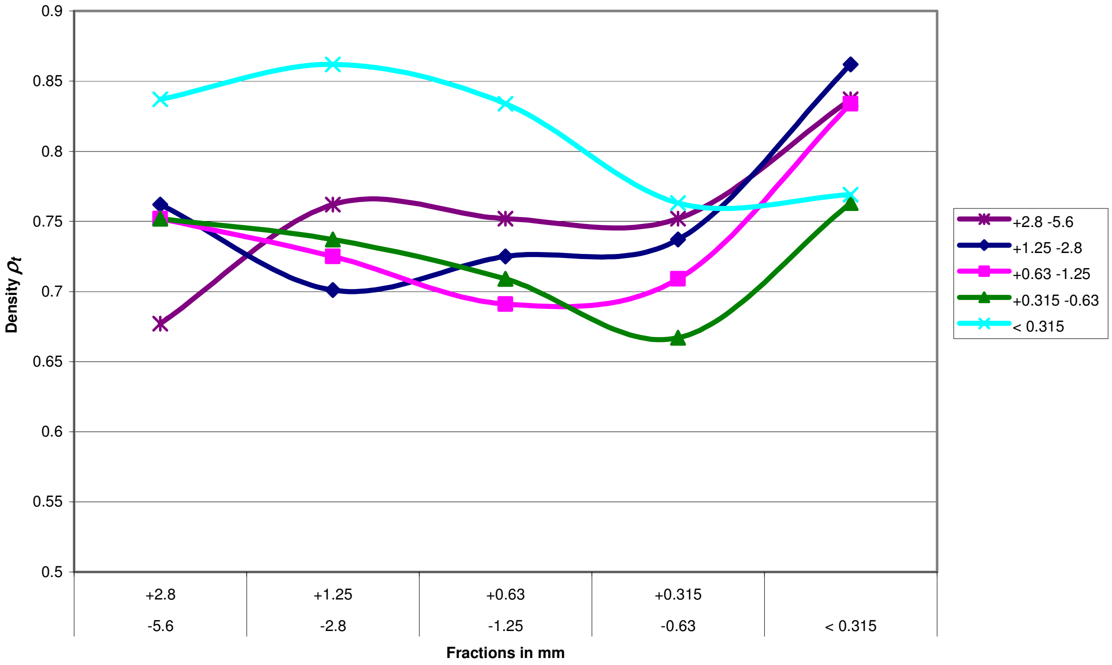


Figure 4.2. The tap density of the mixed filler materials of different fractions

4.1.2. Specific surface area of the plastic powder

The specific surface area of the fine fraction (< 0.315 mm) of plastic powder was determined by using the Blaine permeameter. The specific surface areas of the coarse fractions (0.315-0.63; 0.63-1.25; 1.25-2.5; 2.5-5.0; 5.0-11.2) were calculated on the basis of formula (1.7). The specific surface areas of sphere-shaped particles SSA_{spheric} and tabular particles $SSA_{tabular}$ were calculated on the basis of formula (1.7) – see Table 4.1. Two proportionality constants for the particle shape were used (6 for the spheres and the *AS* value for the tabular-shaped particles). The specific surface area obtained by the Blaine permeameter test for fractions less than 0.315 mm was within a range of 0.0600-0.0630 m^2/g.

Table 4.1. Diameter, aspect and specific surface area of the acrylic plastic powder fractions

Fraction and share	**< 0.315 mm 3 %**	**+ 0.315 – 0.63 mm 7 %**	**+ 0.63 – 1.25 mm 14 %**	**+ 1.25 – 2.5 mm 27 %**	**+ 2.5 – 5.0 mm 35 %**	**+ 5.0 – 11.2 mm 10 %**	**> 11.2 mm 4 %**
d_{50}, mm	0.16	0.47	0.94	1.88	3.75	8.10	12.10
Aspect, *AS*	2.07	1.91	1.71	1.61	1.55	2.40	3.01
SSA_{spheric}, m^2/g	0.0315	0.0107	0.0054	0.0027	0.0013	0.0006	0.0004
$SSA_{tabular}$, m^2/g	0.0652	0.0205	0.0092	0.0043	0.0021	0.0015	0.0013

A comparison between the results of the specific surface area of the fine fraction (< 0.315 mm) obtained by the Blaine permeameter test and the calculations presented in Table 4.1 show that the results are within a similar range. The aspect value is an important parameter for calculating the specific surface area of the particles of the tabular shape.

On balance, the particle size and the surface area provide only the means for classifying the particles and their effects on the composites.

4.1.3. Production of the new composite material using the solid surface technology

Technological tests with the new filler material – acrylic powder – were made by using the *Solid Surface* casting technology. Most of the sinks for bathrooms are produced by applying this technology. Composites were designed with the different mixing ratios of the binder (unsaturated polyester resin) and the filler (acrylic powder). The volume of the filler varied from 50 to 65 mass %. The mixing ratio of the filler was 50 mass % of coarse fractions (+1.25-2.5 mm) and 50 mass % of fine fractions (<0.315 mm) of the acrylic powder material. 1 mass% of the peroxide catalyst was added to accelerate the polymerization of transformation from the liquid state to the solid state with the maximum physical properties including hardness. The liquid mixture of the composite was cast into the mould of plate shape (500 x 500 mm) with a layer thickness of 15 mm. We assumed that by increasing the acrylic filler content in mixture with polyester resin gives the hardness and good wear resistant properties of the working surface of the sink. The curing time of the composite was 4 hours. The best flow characteristics in the mould were in the mixture with 50 mass % of acrylic filler and 50 mass % of resin, but after polishing the best surface quality appeared in

the mixture with 65 mass % of acrylic filler and 35 mass % of resin. The flow characteristics of the mixture 65/35 could be improved by using the binder agent with a lower viscosity.
Commonly the sinks are made of the composite material consisting of the binder agent (an unsaturated polyester resin), the filler material (the dolomite powder) and the catalyst agent added to the resin to make it cure more rapidly. The mixing ratios of the binder agent and the filler material are 25/75 mass %. The hardness and wear resistance of the working surface of bathroom sinks is achieved by applying a thick 0.5-1.0 mm gel-coat (resin) layer, which is either clear or coloured. That is the main reasons why these sinks are intended for the use only in bathrooms because it is easy to scratch or chip off the decorated surface in the kitchen and thus, the bacteria could live there. The traditional filler material used in the casting technology is a white dolomite filler with the chemical composition of calcium and magnesium carbonate $Ca\,Mg\,(CO_3)_2$ with the density of 2850 kg/m^3. The traditional filler is a concrete mix of coarse fractions (+0.20 –0.60 mm), (+0.10 –0.30 mm) and fine fractions (< 0.80 mm) with the apparent density of 2000 kg/m^3.

4.1.4. Porosity of the composite

We assumed that the air entrapment inside the composites will cause porosity of the material and this would influence the mechanical properties. For analyzing porosity, the micro-polishes of cross-sections were made and the images by OM were obtained (Figs 4.3 and 4.4). The images were analysed with *Image-Pro Plus* 3.0. Firstly, the surface areas of the matrix and the pores were calculated. The total area of the pores was 6.5 %. The pore size data obtained by the image analysis method were primarily described through the arithmetical mean diameter d_m of the measured values (Fig. 4.5). The mean diameter of the pores was 97 μm (Fig. 4.4). As it follows from Fig. 4.3, the mean diameter of the PMMA particle was 105 μm; at the same time, the mean roundness parameter RN of particles was 1.56 and the mean aspect AS was 1.67.

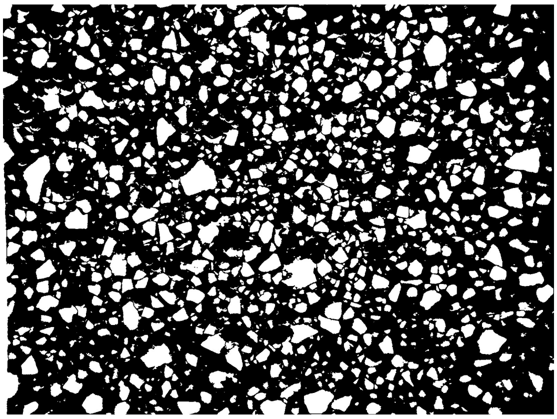


Figure 4.3 The cross-section of the PMMA + the polyester resin composite

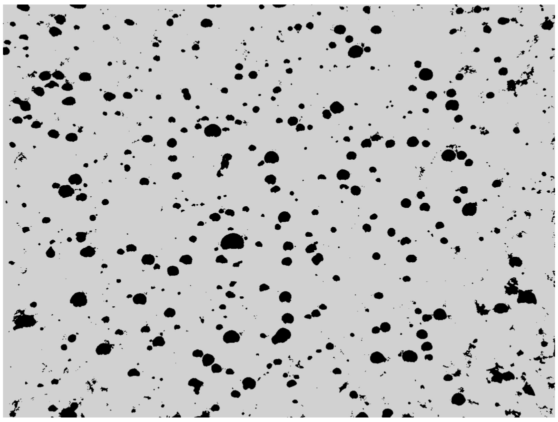


Figure 4.4 The pores in the composite

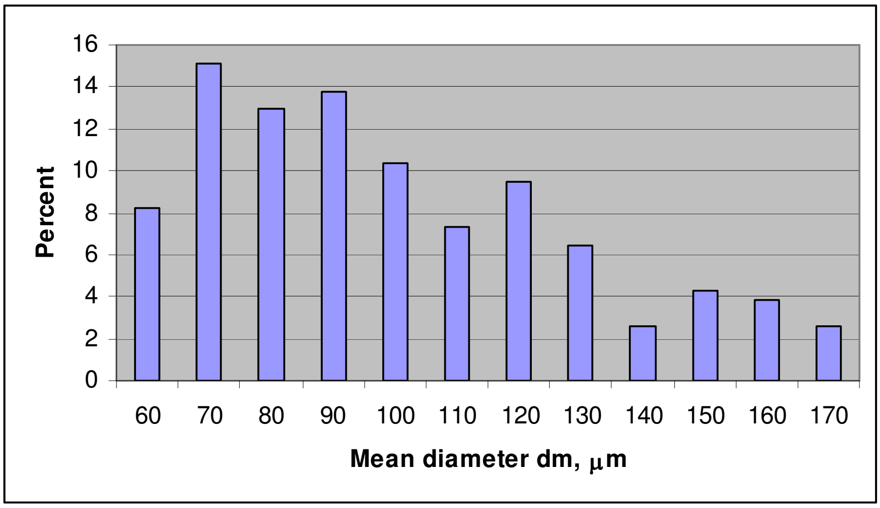


Figure 4.5 The mean diameter of the pores

4.2. Mechanical tests on new composite plastic materials

4.2.1. Tensile strength of the composite materials

The tensile tests were performed on the composites mixed differently (the ratio between the resin and the filler material was different). The test pieces for the tensile test were prepared according to ISO 527-2/1A/50 type1B (Fig. 2.12). The test pieces were cut out from the material of plastic composite plate and polished. The applied measuring device was the caliper and the measurement accuracy was 0.1mm. To compare the test results specimens of PMMA sheet material were made. The results of tensile tests are given in Table 4.2.

Table 4.2. Results of performed tensile test

No. of test piece	**Elongation after breakage ε_B, %**	**Tensile strength R_m, N/mm^2**
35/65-composite plastic (filler PMMA 65 mass % and binder 35 mass %)	0.60	10.5
34/66-composite plastic (filler PMMA 25 mass %, $CaCO_3$ $MgCO_3$ 41 mass % and binder 34 mass %)	0.36	13.2
40/60-composite plastic (filler PMMA 60 mass % and binder 40 mass %)	0.87	17.7
45/55-composite plastic (filler PMMA 55 mass % and binder 45 mass %)	1.28	15.9
50/50-composite plastic (filler PMMA 50 mass % and binder 50 mass %)	0.95	18.2
38/62-composite plastic (filler PMMA 62 mass % and binder 38 mass %)	1.30	20.7
0909- PMMA sheet material	2.02	41.6

Our aim was to compare the tensile strength of the new composite materials to PMMA sheet material (etalon material). The best tensile strength 20.7 MPA belonged to the mixed composite consisting of 38 mass % of resin and 62 mass % of. The tensile strength of the PMMA sheet material exceeded it two times. As it was mentioned before our assumption that the pores inside the material influence the tensile strength of new composite material was true. Pores in the specimen cross-section acted like stress concentrators, so it was difficult to estimate the influence of the resin and filler adhesion on composite material tensile strength.

4.2.2. Surface hardness of the composite

As it was mentioned above, an increase in the acrylic filler content in the mixed polyester resin ensures the strength and hardness of the material, good wear resistance properties for the surface of the materials. Therefore it is important to determine the hardness of the composite. The following hardness methods were used:

- the Brinell hardness HBS (a 1 mm diameter steel ball and a load of 49 N for testing materials 1-3 and 5, the load 98 N for material 4), because the load 49 N did not make indentation on the measured surface observable, and thus the load 98 N was used. The results of the hardness tests are given in Tables 4.3. , 4.4.
- the Rockwell hardness HRE (a 6.35 mm diameter steel ball and a load of 980 N)

Table 4.3. The Brinell hardness of composite materials

No.	**Type of material and composition**	**Load *F*, N**	***HBS***
1.	50/50-composite plastic (filler PMMA 50 mass % and binder 50 mass %)	49	21.5
2.	35/65-composite plastic (filler PMMA 65 mass % and binder 35 mass %)	49	13.3
3.	34/66-composite plastic (filler PMMA 25 mass %, $CaCO_3$ $MgCO_3$ 41 mass % and binder 34 mass %)	98	47.4
4.	45/55-composite plastic (filler PMMA 55 mass % and binder 45 mass %)	49	15.2
5.	40/60-composite plastic (filler PMMA 60 mass % and binder 40 mass %)	49	21.1
6.	38/62-composite plastic (filler PMMA 62 mass % and binder 38 mass %)	49	13.4
7.	0909- PMMA sheet material	49	26.1

The results of the *Brinell* hardness test showed that the best surface hardness belonged to the composite material 34/66, but the applied load was two times bigger than for other composites. To compare the hardnesses of the composite materials the Rockwell hardness test was performed.

Table 4.4. The surface and core hardness of the studied materials

Type of material	**Composition**	**Hardness**		
		HBS	*HRE*	
			surface	**core**
0909 PMMA sheet	Acrylic plastic without any technological additive	21.5	99.6	99.2
Composite - 34/66	Composite plastic (filler PMMA 25 mass %, $CaCO_3$ $MgCO_3$ 41 mass % and binder 34 mass %)	47.4	78.5	73.7
Composite - 40/60	Composite plastic (filler PMMA 60 mass % and binder 40 mass %)	21.1	81.9	80.3

There was a noticeable difference between the surface and core hardness of the composite material 34/66. The PMMA sheet material had the best surface hardness, which is mainly used for the production of bathtub shells. The best composite material for producing the washbasin is the composite 40/60, because it has the similar values of the surface and core hardness after the wear test.

4.2.3. The abrasive wear test

The abrasive wear resistance of plastic composites was determined using the method of the rubber wheel test (v = 10 m/s, F = 140 N) and abrasive quartz sand (+0.1 – 0.3 mm) with the amount of 0.5 kg. The wear test results are given in Table 4.5. Wear surface profiles were measured by the profilometer. The obtained results are presented in Figs 4.6-4.7.

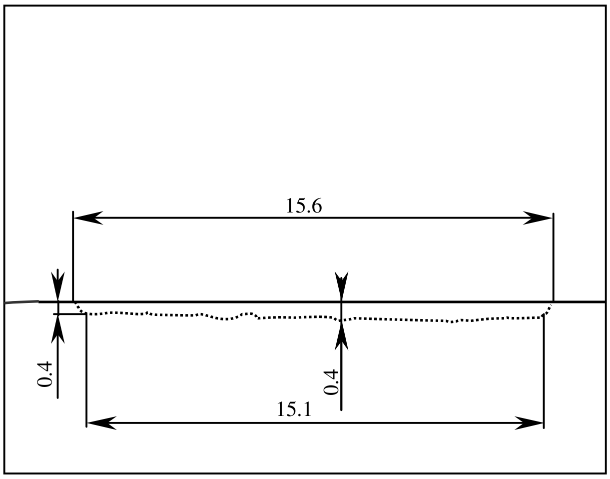


Figure 4.6 Wear surface profile of the 40/60 composite material

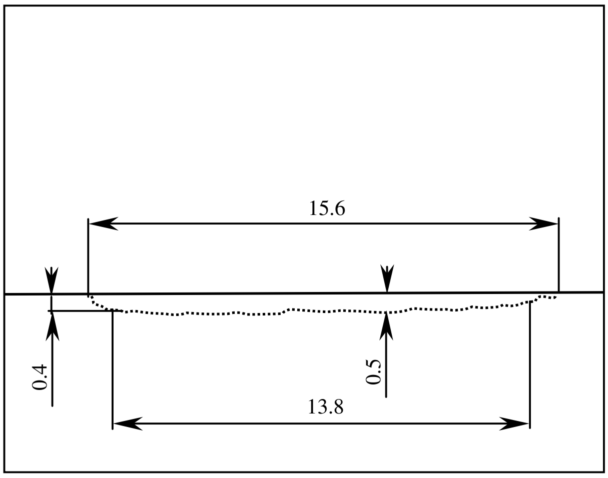


Figure 4.7 Wear surface profile of the 34/66 composite material

Table 4.5. The abrasive wear resistance of the plastic composite materials

Type of material	Density	Wear rate				Relative wear resistance ε_v	
		surface		core		surface	core
	g/cm^3	in mg	in mm^3/Nm 10^{-5}	in mg	in mm^3/Nm 10^{-5}		
0909-PMMA sheet	1.19	113.0	858.6	110.6	840.3	1.0	1.0
34/66-composite	1.57	241.2	1389.1	226.4	1303.7	0.62	0.64
40/60-composite	1.08	128.8	1078.6	106.6	892.10	0.80	0.94

4.3. Particles packing

Calculations for maximum particles packing were made on the basis of the cubic crystal systems of metallic atoms. The maximizing of the particles packing is discussed in literature [72, 94]

Particles packing with one fraction

Particle packing of one fraction in the cubic crystal structure:

- packing by the simple cubic crystal structure (Fig. 4.8). Compactness of the structure $\eta = 0.52$. This particle packing is improbable because of the random packing of the particles.

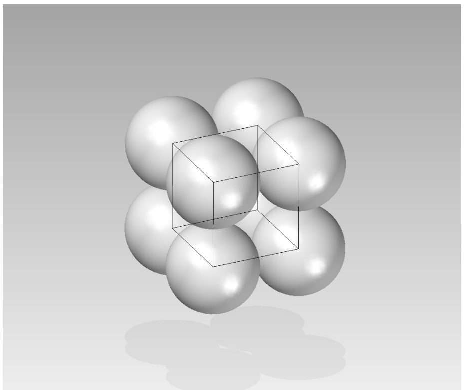


Figure 4.8. One fraction of the particles placed in the corners (8 pcs)

- packing by the body-centred cubic crystal structure – (Fig. 4.9). Compactness of the structure $\eta = 0.68$. This particle packing occurs more frequently in the mono-particle packing system.

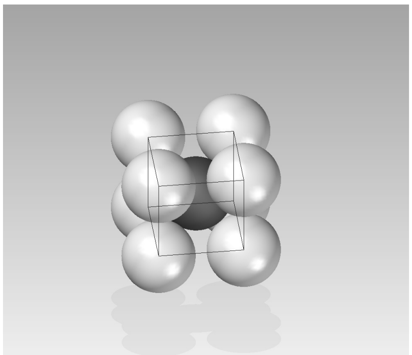


Figure 4.9. One fraction of the particles placed in the corners (8 pcs) and in the centre (1 pc)

Particles packing with two fractions

Particle packing of two fractions in the cubic crystal structure:

- packing by the simple cubic crystal structure (Fig. 4.8). The second smaller fraction is in the centre with the diameter $d_2 = 0.73 d_1$. Compactness of the structure $\eta = 0.82$. This particle packing is probable.

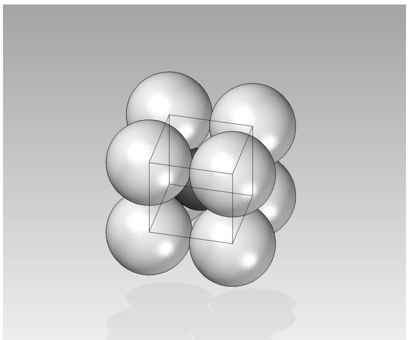


Figure 4.10. Two fractions of the particles - larger placed in the corners (8 pcs) and smaller in the centre (1 pc)

- packing by body-centred cubic crystal structure (Fig. 4.9). The second smaller fraction with the diameter $d_2 = 0.63 d_1$ is in face-surfaces. Compactness of the structure is $\eta = 0.87$. This particle packing is probable.

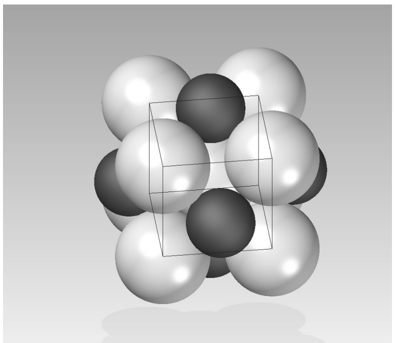


Figure 4.11. Two fractions of the particles packed in the body-centred crystal structure - larger particles in the corners and centre (9 pcs) and smaller ones in the centre of faces (6 pcs)

Particles packing with three fractions

Particle packing of three fractions in cubic crystal structure:

- packing by a simple cubic crystal structure (Fig. 4.8). The second fraction with the diameter $d_2 = 0.63 d_1$ is in the centre, the third smallest $d_3 = 0.155 d_1$ is on the edges of the cube. Compactness of the structure $\eta = 0.89$.

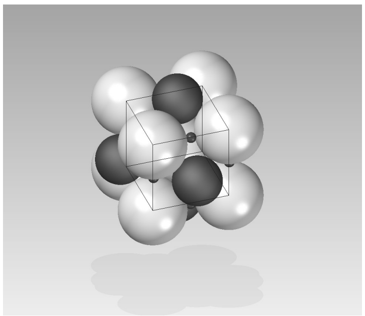
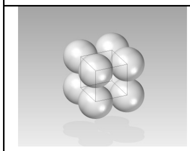
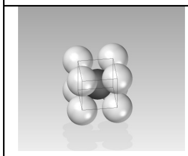
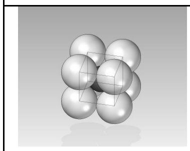
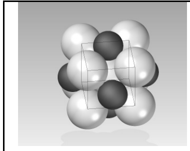
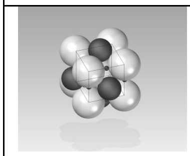


Figure 4.12. Three fractions of the particles packed in a body-centred crystal structure the larger packed in the corners and centre (9 pcs), the smaller in the centre of faces (6 pcs) and the smallest in the centre of edges (12 pcs).

The types of the particle packing are presented in Table 4.6.

Table 4.6. Particle diameters and compactness of the structure

Type of structure	Powder particle diameter	Compactness of the structure
 $R = \frac{a}{2}$	One fraction: $d_1 = 2R$	$\eta = 0.52$
 $R = \frac{a\sqrt{3}}{4}$	One fraction: $d_1 = 2R$	$\eta = 0.68$
 $R = \frac{a}{2}$	Two fractions: $d_1 = 2R$ $d_2 = 0.73d_1$	$\eta = 0.82$
 $R = \frac{a\sqrt{3}}{4}$	Two fractions: $d_1 = 2R$ $d_2 = 0.63d_1$	$\eta = 0.87$
 $R = \frac{a\sqrt{3}}{4}$	Three fractions: $d_1 = 2R$ $d_2 = 0.63d_1$ $d_3 = 0.155d_1$	$\eta = 0.89$

4.4. The numerical modelling

As it is pointed out above, one of the aims of the current study is to maximize the density of the filler material. The relation between the density of the filler material and the fractions of the PMMA powder are modelled on the basis of the experimental data (discussed in section 4.1.1). Two different numerical techniques are employed for the design of response surface (cubic B-splines and *Neural Networks*) [95-100]. When proceeding from the model of the obtained response surface, the search for the maximum values of the filler material density is performed. The minimum values of the filler material density are also of much interest as the most undesired (critical) cases.

4.4.1. An approximation based on the use of cubic B-splines

The *Bezier* and B-splines are widely used for the computer aided design. The surfaces constructed by the use of these two new splines do not normally contain the given points. In this respect some similarity with the least-squares method can be followed.
However, unlike a natural cubic spline and the least-square method, the Bezier and B-splines have the local control. This means that when modifying one control point, it only affects the part of the surface near that control point. This is very useful when using the B-splines for designing surfaces [95, 99]. The complexity of the approximation and the computation cost are reduced significantly. The interpolating B-spline surface patch depends on 16 points [95], as shown in Fig. 4.13.

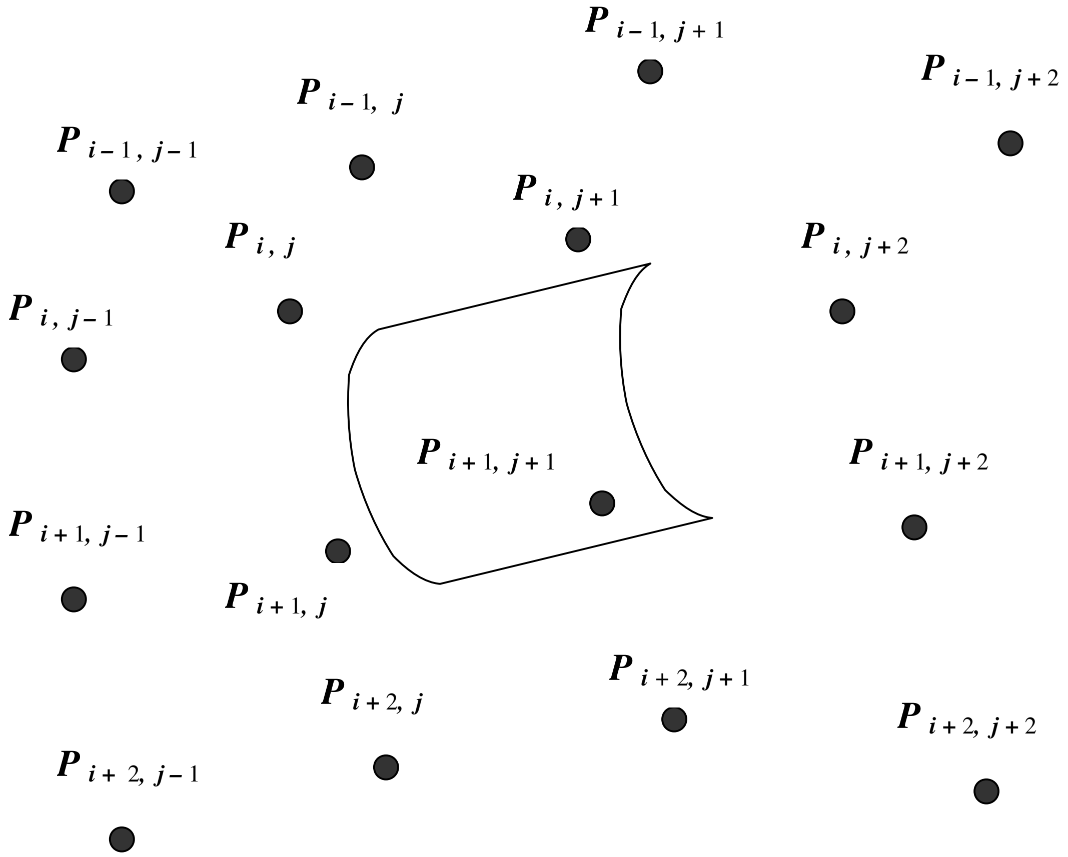


Figure 4.13. The example of B-spline surface patch [95]

Let us consider that a set of points $p_k = \{(\bar{x}_k, \bar{y}_k, \bar{z}_k), k = 0,...,n\}$ is determined from the experimental data and we wish to fit a surface to those points. The coordinates of any point on the surface can be given in the parametric form as

$$\begin{aligned} x_i &= x_i(u,v), \\ y_i &= y_i(u,v), \\ z_i &= z_i(u,v) \end{aligned} \tag{4.1}$$

where *u,v* are the independent variables that range over a given set of values ($0 \le u \le 1$ and $0 \le v \le 1$);

Proceeding from the matrix formulation for cubic B-splines given in [68] the coordinates $x_{i,j}(u,v)$ can be computed as

$$x_{i,j}(u,v) = (1/36)\left[u^3,u^2,u,1\right]M_b X_{i,j} M_b^T \left[v^3,v^2,v,1\right]^T \tag{4.2}$$

where $\overline{X}_{i,j}$ is the 4 x 4 matrix

$$\overline{X}_{i,j} = \begin{pmatrix} \overline{x}_{i-1,j-1} & \overline{x}_{i-1,j} & \overline{x}_{i-1,j+1} & \overline{x}_{i-1,j+2} \\ \overline{x}_{i,j-1} & \overline{x}_{i,j} & \overline{x}_{i,j+1} & \overline{x}_{i,j+2} \\ \overline{x}_{i+1,j-1} & \overline{x}_{i+1,j} & \overline{x}_{i+1,j+1} & \overline{x}_{i+1,j+2} \\ \overline{x}_{i+2,j-1} & \overline{x}_{i+2,j} & \overline{x}_{i+2,j+1} & \overline{x}_{i+2,j+2} \end{pmatrix} \tag{4.3}$$

containing the *x*-coordinates of the 16 given points (see Fig. 4.6) and the matrix M_b is given by

$$M_B = \begin{pmatrix} -1 & 3 & -3 & 1 \\ 3 & -6 & 3 & 0 \\ -3 & 0 & 3 & 0 \\ 1 & 4 & 1 & 0 \end{pmatrix} \tag{4.4}$$

similar bicubic equations (cubic in u and v) hold for y and z coordinates

$$\begin{aligned} y_{i,j}(u,v) &= (1/36)\left[u^3,u^2,u,1\right]M_b Y_{i,j} M_b^T \left[v^3,v^2,v,1\right]^T, \\ z_{i,j}(u,v) &= (1/36)\left[u^3,u^2,u,1\right]M_b Z_{i,j} M_b^T \left[v^3,v^2,v,1\right]^T \end{aligned} \tag{4.5}$$

In the current application, the given surface points are located regularly with respect to x and y coordinates, since the values of the x and y coordinates are considered as the average values of the particle sizes of each fraction of the PMMA powder. Furthermore, the same values of the particle sizes are used in both x and y axes. As result, the expressions for x and y coordinates reduce from bi-cubic equation to cubic equations (no coupling). Thus, these equations can be solved with respect to u (or v). The latter fact allows the simplified application of the parametric relations (4.2), (4.5). Namely, first, the parameters u and v can be computed for fixed values of x and y, next, the value of the z coordinate can be determined from the second equation of (4.5). The same procedure can be applied to a general case, where the given surface points are not located regularly, however, in the latter case the complexity of the numerical solution increases significantly.

The computation algorithm is implemented in Maple 10 (symbolic and numeric calculation) and Matlab 6.5 (numeric calculation, graphics) codes. The data exchange between the Maple and Matlab software packages is performed through text files. The surface models corresponding to the apparent and tap densities of the materials of the PMMA powder are depicted in Figs 4.14 and 4.15, respectively.

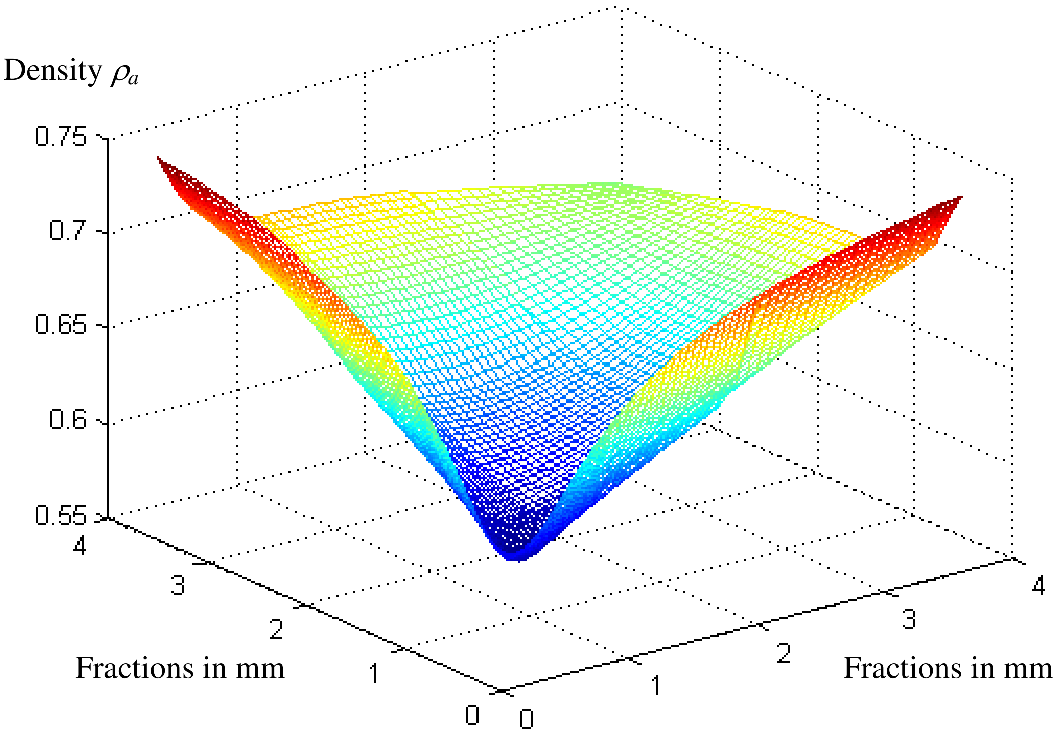


Figure 4.14. A surface model of the apparent density of the acrylic powder (cubic B-splines).

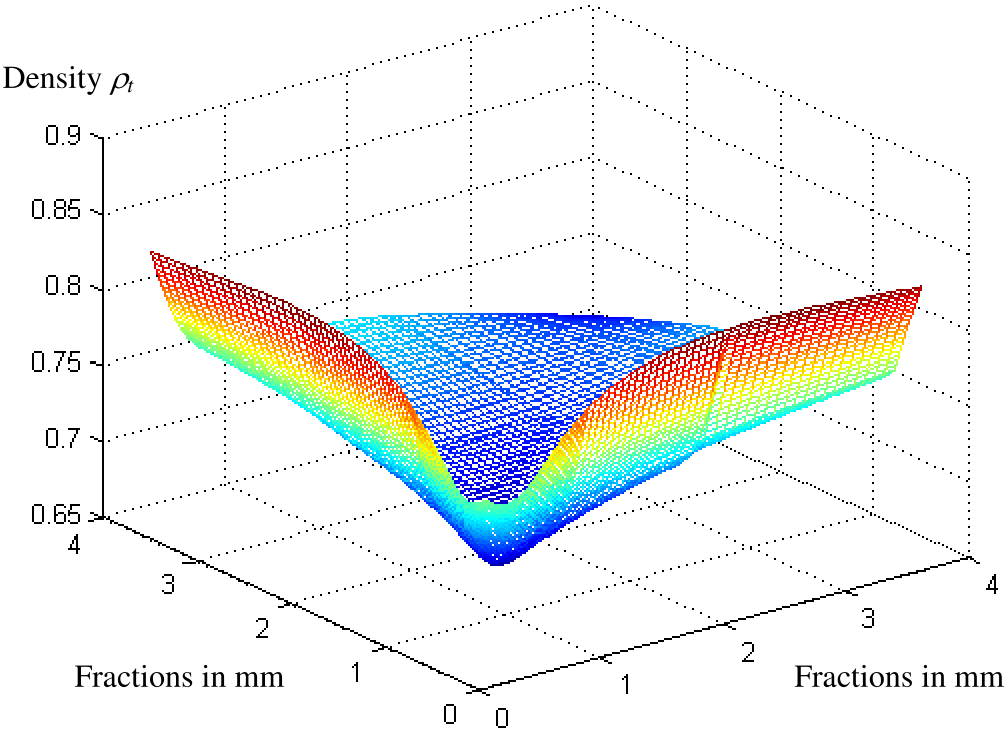


Figure 4.15. A surface model of the tap density of the acrylic powder (cubic B-splines).

The proposed algorithm enables to determine the densities of the filler material at any point of the constructed surface. How to determine the extremal values of densities of the filler material is discussed in section 4.2.3.

4.4.2. An approximation based on the use of *Neural Networks* (NN)

Neural networks (NN) were started about 50 years ago. Their early abilities were exaggerated, casting doubt on the field as a whole. An increasing interest in this field can be observed with the appearance of new techniques (recursive neural network, the Kohonen's feature map, etc.) and a better theoretical understanding of their capabilities.

The problems of neural networks can generally be categorized as one of four types: classification, function approximation, prediction and clustering. The problems studied by the use of neural networks belong to the different areas, such as marketing, the investment analysis, signature analysis, process control, speech generation, statistical analysis and others.

In the current study the neural networks are applied to the approximation of the density function (surface fitting). An approach suggested is based on the use of the MATLAB neural network toolbox. The model is constructed by applying the generalized regression of the neural network function newgrnn (I,O,spread). Three input parameters can be described as

I – n × m matrix of m input vectors,
O – n × m matrix of m target class vectors,
spread – the spread of radial basis functions, default = 1.0.

The function newgrnn creates a two-layer network. The first layer has radbas neurons and the second layer has purelin neurons. A larger spread corresponds to a smoother function approximation. To fit the data very closely, a smaller spread than the typical distance between input vectors should be used. To fit the data more smoothly, a larger spread should be used.

In order to calculate the outputs for a concurrent set of input vectors, a network simulation function sim (net, P) is used. The first input parameter is a network object generated by the function newgrnn and the second parameter stands for the input matrix. This is the batch mode form of simulation, in which all of the input vectors are placed in one matrix P. Finally the outputs of the function sim (net, P) can be used as inputs (Z) for function mesh (X,Y,Z), which is used for the surface drawing.

The obtained surface models corresponding to the apparent and tap densities of the PMMA powder materials are depicted in Figs 4.16 and 4.17, respectively.

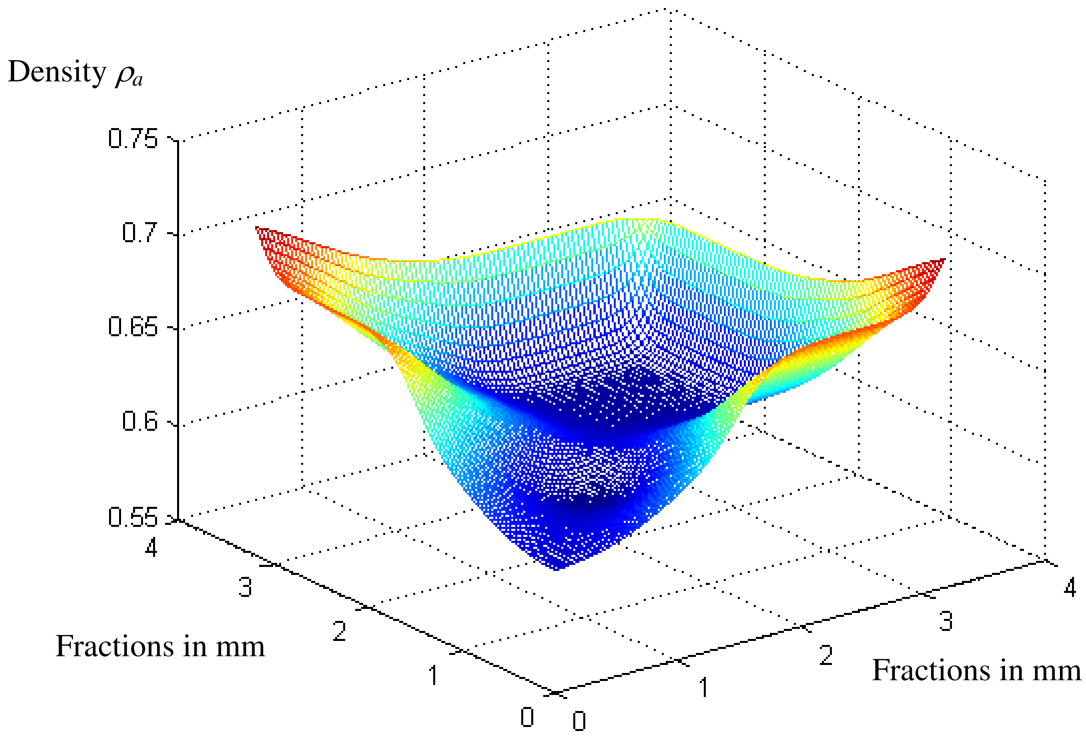


Figure 4.16. A surface model of the apparent density of the acrylic powder (*neural networks*).

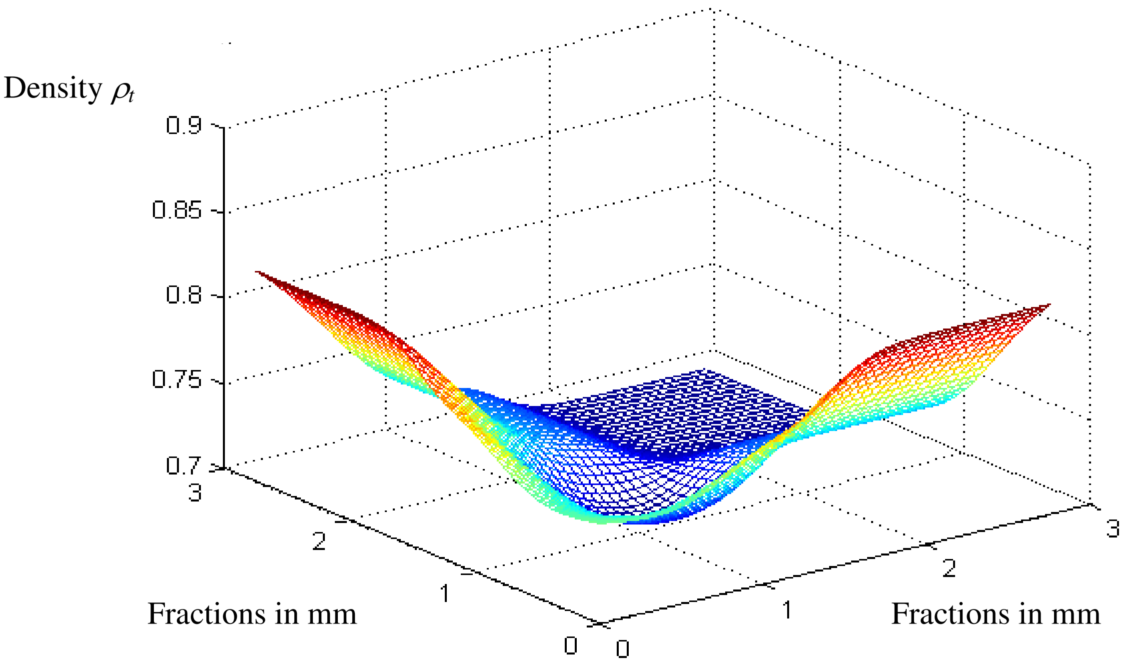


Figure 4.17. A surface model of the tap density of the acrylic powder (*neural networks*).

In Figs 4.8 and 4.9, relatively small values of the spread parameter (0.5) are used in order to fit data very closely. As a result, the data are not fitted very smoothly and the corresponding surfaces have several local and global extremals. The results obtained by both approximations are reasonable. However, due to the absence of the closed-form analytical solution to the considered problem, here is no possibility to compute and compare the absolute errors corresponding to cubic B-splines and neural network based models. An approximation based on the use of neural networks seems more flexible due to the presence of the spread parameter in a model. It should also be mentioned that the implementation of the neural network based model was much simpler, since most of the work can be done by built-in functions of the MATLAB and its toolboxes.

4.4.3. The maximum and minimum densities of the filler material

Let us proceed from the surface modelled by the use of neural networks or cubic B-splines. In order to determine the extremal values of the density of the filler material, a search for the global maximum and minimum should be performed. The MATLAB functions fmincon (optimization toolbox) and patternsearch (genetic algorithm and direct search toolbox) has been utilized for this task. The extremal values of the density of the filler material depend on the value of the parameter spread (NN approximation). As it can be expected, the extremal values of the density of the filler material increase with the decreasing values of the modelling parameter spread, and vice versa (see Table 4.7).

Table 4.7. Dependence of the density of the filler material on parameter of the modelling spread (NN)

Spread	0.5	0.7	1.0	2.0
Min of the apparent density	0.607	0.620	0.631	0.637
Max of the apparent density	0.733	0.730	0.727	0.711
Min of the tap density	0.701	0.706	0.724	0.755
Max of the tap density	0.800	0.789	0.784	0.774

It is noted that the function (fmincon) converges to the nearest extremal. Thus the global extremal can be achieved by the selection of a suitable initial point. The initial max (min) point is chosen from the set of grid points (the surface is covered by a uniform grid).
The extremal values of the density of the filler material corresponding to cubic B-splines approximation are given in Table 4.58.

Table 4.8. The extremal values of the density of the filler material (cubic B-splines)

Min of the apparent density	0.695
Max of the apparent density	0.741
Min of the tap density	0.763
Max of the tap density	0.834

Table 4.5 is significantly reduced in comparison with Table 4.4, since in the case of cubic B-splines the approximation is not a modelling parameter.

An attempt was made to use the MATLAB Genetic Algorithm and Direct Search Toolbox. The genetic algorithm functions ga and gatool allow to determine the global extremal. Unfortunately, these functions do not support the solution to the constrained problems. There exists the possibility of applying limitations on the initial range of data. However, the subsequent generations can contain the points whose entries do not lie in the given initial range. The direct search algorithm – patternsearch – supports the linear constraints given in the form of inequality or equality, also setting the lower and upper bounds on the variables, but similarly to the optimization function fmincon, it converges to the local minimum (maximum). For that reason the initial grid covering the whole surface and the preliminary search for the optimum in the grid points is employed.

4.5. New products made of the recycled material

4.5.1. The concept of a sustainable product development

The term *sustainable development* was coined by the World Commission on Environment and Development in 1987 meaning a development satisfying the needs of the present generation by considering the needs for a sufficient supply for the future generations. Five years later at the conference in Rio, the members of more than 100 countries formulated an action programme along this basic principle and made it known worldwide. It is universally accepted, but meanwhile the sustainable development has to consider the social, economic, and ecological aspects because it is the development goal for the humankind on the globe [3].

Sustainability = Social welfare + Economy + Ecology

The environmental impact has been generated solely by the existence of human beings in the world – the social, even religious or technological aspects govern this. The environmental impact has certainly been caused by our standards of living and the use of technology to improve it.

Environmental impact = Population x Wealth/population x Environmental impact/ wealth

The environmental impact may mean the saving of resources, the minimization of emissions including the disposal, or the avoidance of hazardous substances. The saving of resources will obtain the equal importance in the future if the limited availability of fuel will get more public awareness again. The positioning of a product depends mainly on its efficient use of resources, on an environmental impact of its production, on the environmental savings by using it at the marketplace, and the amount of the final waste left at the end of its lifetime – in other words, on its ability to be recycled. Plastics fit into this framework because they make a positive contribution to most of these sectors.

4.5.2. A case study – the washbasin

With the evolution of new processes and products, the name *cultured marble* has come to be identified with gel coated, calcium carbonate filled, and cast polyester matrix. With onyx (solid or densified) and granite (polyester, epoxy and acrylic modified) products coming into the market place, the market trend is to identify all these products as cast polymers [101].

As the filler loading exceed 50% by mass, the resulting material begins to enter into “densified” or “solid surface” description. Solid surface products are formed without protective gel coats and with very high filler content. They employ high performance resin systems such as acrylic and unsaturated polyester. The parts are fabricated by casting the combination of resin and filler in the open moulds.

The typical steps followed by production of solid surface include [101, 102]:

- a mould of desired shape is prepared with a release agent.
- a mixture of the polyester resin and filler (matrix) is catalysed, coloured for the marble effect and poured or trowelled into the mould. The mould is vibrated to release the air and level the matrix.
- after curing and shrinkage, the part is removed from the mould, trimmed and polished if necessary. The parts are laid on a flat surface and allowed to cure if removed from the mould while green (flexible).

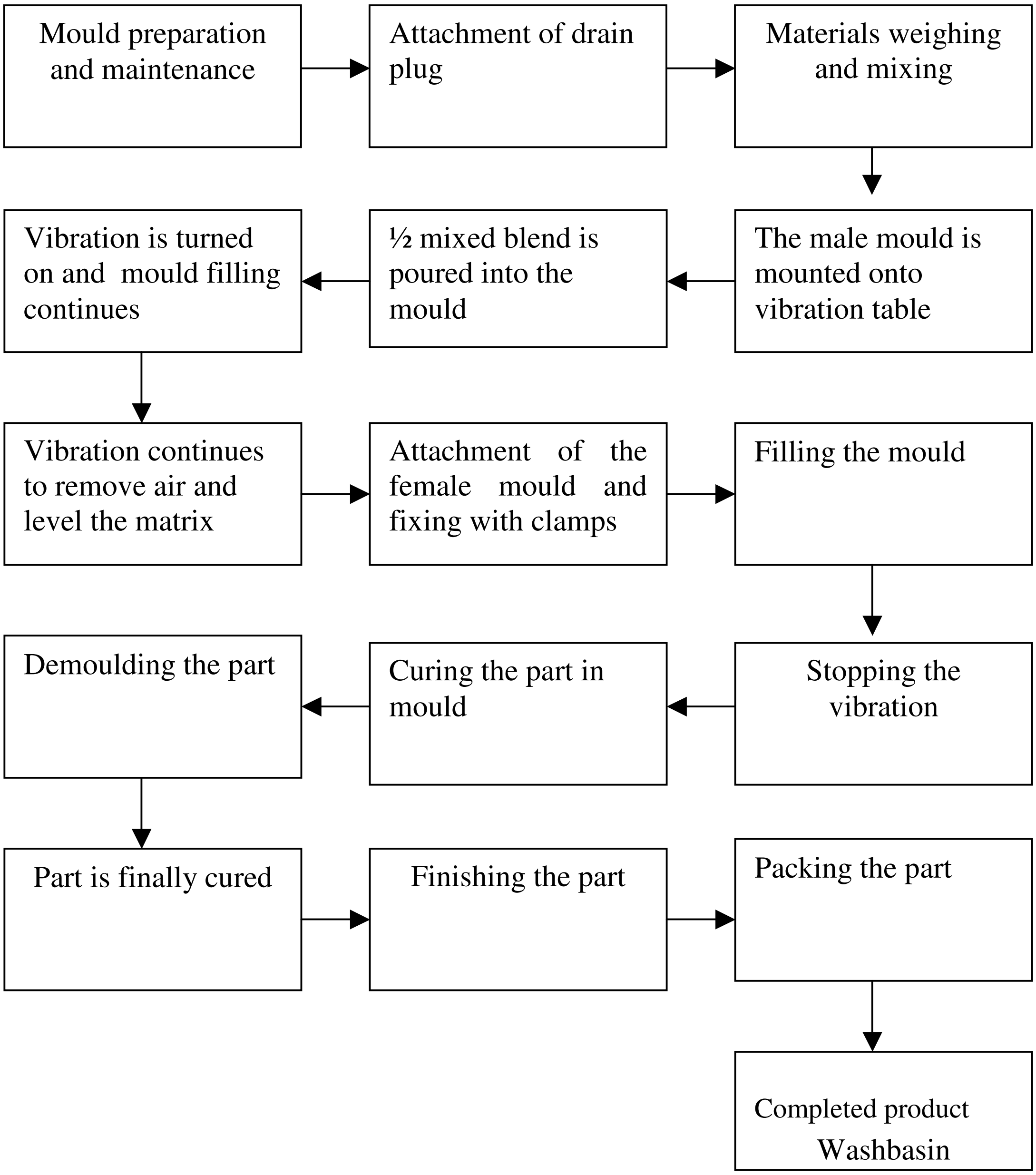


Figure 4.18 A technological scheme of producing the washbasin by using the solid surface technology

The solid surface products are formed without the protective gel-coats (coloured resin layer), with a very high content of the filler, and they have the following properties:

- the solid surface products employ a high-performance resin-systems such as acrylic and unsaturated polyester resin; the design benefit of the solid surface materials is their ability to be machined like wood;
- the solid surface products are homogeneous – that is, the colour or pattern must be absolutely consistent throughout every part;
- the solid surface products have a remarkable hardness, which is normally tested by the steel balls dropped from different heights;
- the solid surface products are non-porous;
- the solid surface products are stain- and chemical-resistant

Solid surface products great performance characteristics make it ideal for many situations in both the residual and commercial applications: bathroom panelling, bathtubs, floor tiling, handbasins, table surfaces etc.

Our attempt was to produce the washbasin by using the solid surface technology. We used the developed filler material, which consisted of 40 per cent of coarse fractions and 60 per cent of fine fractions. The used composite mixture was 60 mass % of acrylic filler and 40 mass % of resin.

The washbasin produced by the solid surface technology using the recovered plastic powder as the filler material had the mass almost two times smaller than that of the washbasin produced by using calcium carbonate as the filler material.

Figure 4.19 The washbasin produced by the solid surface technology of the newly developed composite material.

4.6. The scheme for reprocessing and recovery of the composite plastic scrap

The scheme for the reprocessing and material recovery of the composite material scrap consists of the collection of post-consumer electronic equipment containing composite plastics and/or multi-materials (see Fig. 4.20).

The post-consumer electronics equipment is dismantled manually and the composite plastic details are identified by the MIR or FTIR equipment [75-78]. The dismantled components require an effective separation technology to recover the main plastics.

One shaft shredder is used for the preliminary size reduction of large details (metals, plastics, composite plastics). Then the disintegrators will be applied for further size reduction enabling the separation of the components of the composite plastic – the PMMA plastic powder and glass fibre. A two-step, wet and dry separation technology is required, including an innovative sink-floating tank for the separation of the composite plastic and metallic content from the milled PCBs [34, 79].

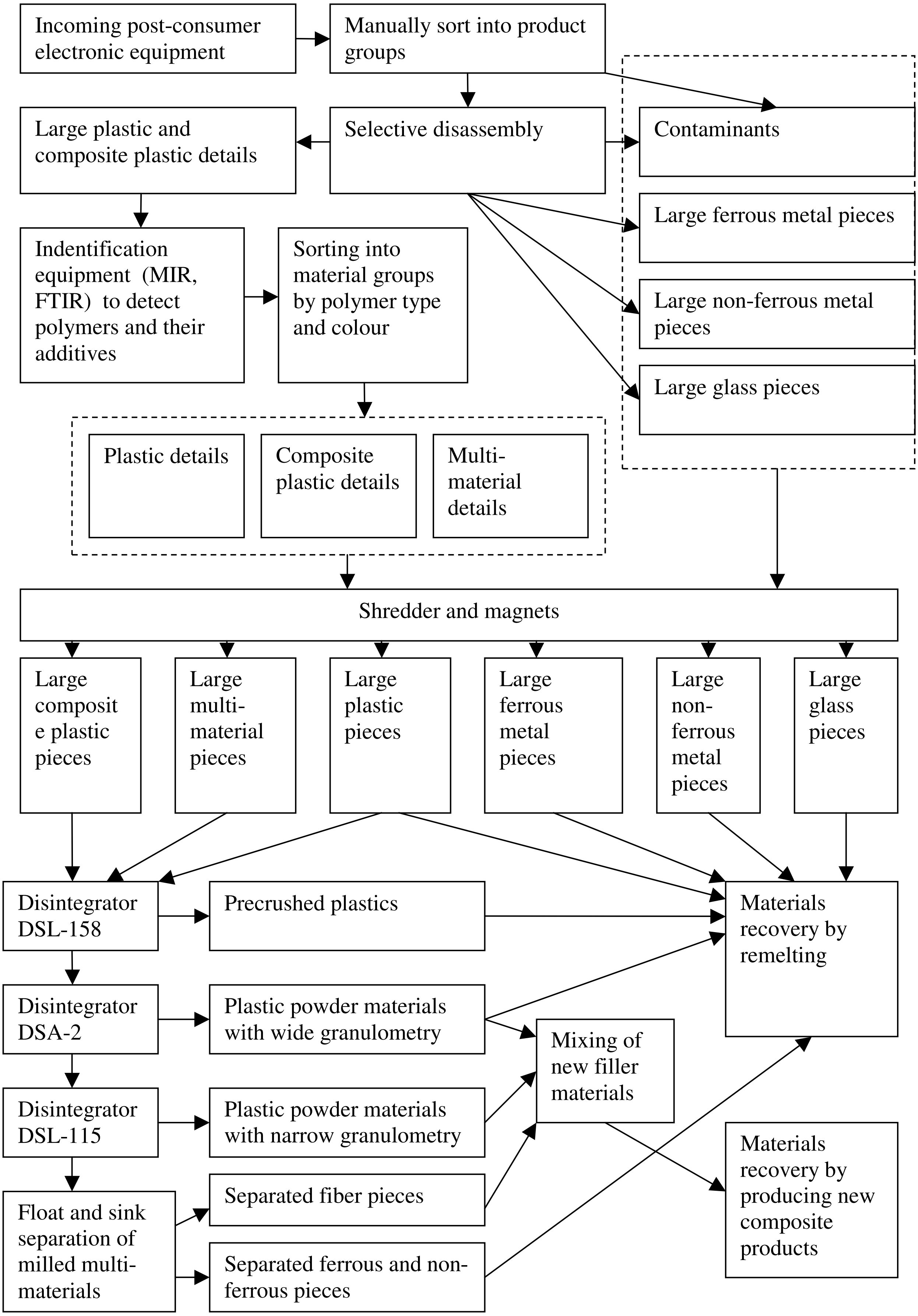


Figure 4.20. Reprocessing and material recovery scheme of composite plastic scrap

4.7. Conclusions

1. To develop the new filler material from the milled PMMA powder, the following technological properties were examined:

- the ratio of density of the filler material to the size of the fractions of the PMMA powder was determined. The most densified filler consisted of 1:1 weight ratio of coarse fractions and fine fractions (≤ 0.315 mm) and thus, had the best apparent density 0.74 and also the best tap density 0.86;
- the mixed filler had the best properties of flowability, it consisted of 50 mass % of coarse fractions (+0.63 – 1.25 mm) and 50 mass % of fine fractions (≤ 0.315 mm) and thus, had the best flowability (through 5.8 mm opening of the funnel) 21 seconds;
- the best ratio of a new mixed composite material contained 40 mass % of resin and 60 mass % of the PMMA powder filler.

2. The mechanical tests performed on the composites had the following results:

- the mixed composite with UP resin and PMMA powder ratio 38/62 had the best tensile strength. The tensile strength of the composite with PMMA filler 38/62 was 25% higher than that of composite with traditional mineral filler.
- the mixed composite 40/60 had the best surface hardness by the Rockwell test method.
- the composite 40/60 had the best relative wear resistance. The core wear resistance was better than that of surface.

3. The results obtained by the numerical modelling of the density of the PMMA powder are reasonable by both of the used approximations. However, due to the absence of the closed form, the analytical solution to the problem considered here has no possibility of being computed and compared to the absolute errors corresponding to cubic B-splines and the neural network- based models. An approximation based on the use of neural networks seems more flexible due to the presence of the spread parameter of the model. It should be also mentioned that the implementation of the neural network- based model was much simpler, since most of the work can be done by the built-in functions of the MATLAB and its toolboxes. The extreme values of the density of the filler material depend on the value of the parameter of the modelling spread. The maximum value of the tap density 0.8 corresponds to the spread value 0.5. By increasing the spread value up to 2.0, the maximum tap density decreases to 0.77.

3. As a result of this case study, a new technology for the production of the washbasin was developed, which consisted of the following steps:

- the new filler material for the washbasin production with the determined particle size and technological properties was developed
- the new composite material with good mechanical and technological properties was developed

5. A scheme for the reprocessing and material recovery of the composite plastic scrap was developed for recycling companies in Estonia. To achieve the targets of the material recovery provided by the European Union WEEE directive the content of the plastic and composite plastic should be separated.

GENERAL CONCLUSIONS

1. Mechanical reprocessing exists as the main possibility of recycling thermosetting composite plastics. The reprocessing of the composite PMMA+GFP plastic scrap by using disintegrators in the separative and/or selective milling conditions will enable to produce the acrylic plastic powder with a determined granularity and technological properties (the apparent density and flowability).

2. To compare the results of milling by the disintegrator devices with the existing advanced technologies for the size reduction of the composite and compounded plastic scrap (by using horizontal shredders and granulators), the advantages of disintegrator milling are demonstrated:
   a. simultaneous separation of the components of composite (impossible with shredders and granulators)
   b. lower specific energy of treatment

3. The new filler material developed from the milled PMMA powder had the following technological properties:
   - the most densified filler consisted of 50 mass % of coarse fractions and 50 mass % of fine fractions.
   - the best ratio for a new mixed composite material was 40 mass % of resin and 60 mass % of the PMMA powder filler.

4. The results obtained by the numerical modelling of the density of the PMMA powder are reasonable by both of the used approximations. The extremal values of the density of the filler material depend on the value of the parameter of the modelling spread. The maximum value of the tap density 0.8 corresponds to the spread value 0.5. By increasing the spread value up to 2.0, the maximum tap density decreases to 0.77.

5. The washbasin produced by the solid surface technology using the recovered PMMA plastic powder as the filler material had the mass almost two times smaller than that of the washbasin produced by using calcium carbonate as the traditional filler material.

6. The recycling system of composite plastic is proposed for Estonian recycling companies. To achieve the targets of the material recovery provided by the European Union WEEE directive the content of the plastic and composite plastic should be separated and recovered.

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II. **Kers, J.,** Kulu, P. and Goljandin, D., 2005, Recycling of reinforced acrylic plastic wastes by high-energy mills, Proc. 1st Int. Conference on Engineering for Waste Treatment, Albi, CD

III. **Kers, J.,** Kulu, P., Goljandin, D. and Mikli, V., 2006, Reuse of reinforced acrylic plastic waste in new composite material development. Proc. 5th Int. DAAAM Conference, Tallinn, Estonia, 2006, 267- 271

IV. **Kers, J.,** Kulu, P., Goljandin, D. and Mikli, V., Reprocessing technology of composite plastic scrap and properties of materials from recycled plastics, *Proc. Estonian Acad. Sci. Eng.,* 2007, 13, 1 (in press)

V. **Kers, J.,** Kulu, P. and Goljandin, D., 2006, Sustainable materials from composite plastic wastes, International Balaton Conference Reinforced Plastics 2006,

VI. **Kers, J.,** Küttis, T., Kaupmees, E., Kasutuselt kõrvaldatud personaalarvutites sisalduva plasti ringlusse võtt, *Keskonnatehnika,* 7, 2006, 18-21

Approbation
International conferences

1. 5th Int. Conference of DAAAM Baltic Industrial Engineering – Adding Innovation Capacity of Labour Force and Entrepreneur , Tallinn, Estonia, 20-22 April, 2006

2. Int. Balaton Conference Reinforced Plastics, Balatonvilagos, Hungary, 23-25 May, 2006

3. JEC Composites Forums, Paris, France, 28-30 March, 2006

4. European Congress on Advanced Materials and Processes EUROMAT 2005, Prague, Czech Republic, 5-8 September, 2005

5. 1st Int. Conference On Engineering for Waste Treatment, WASTEENG, Albi, France, 17-19 May, 2005

6. Global Symposium on Recycling, Waste Treatment and Clean Technology, REWAS 2004, Madrid, Spain, 26-29 September, 2004

ABSTRACT

The aim of this work was to study and develope prospective methods and technique for mechanical reprocessing of industrial composite plastic scrap, plastics and multi-materials of post–consumer electronics. Composite plastic scrap consisted of acrylic plastic with glass fiber reinforcement in polyester resin matrix was used as industrial scrap, compounded plastic and multi-materials (printed circuit boards) from personal computers were used as the plastic scrap from of electric and electronic equipment.

This study describes mechanical reprocessing of different type of plastic scrap in high-energy disintegrator mills in the direct and selective milling systems. The grindabilty and separation of components of composites – acrylic plastic and glass fiber were analysed. The granularity and morphology properties of the plastic powders are discussed. New filler material from acrylic plastic powder was developed for application on solid surface technology. Mechanical properties of new composite materials, produced by experimental manufacturing technology, were tested. The potential application areas of the new PMMA filler material are in production of washbasins, bathtubs, table surfaces etc. The scheme for recycling – reprocessing and material recovery of composite plastic scrap was presented.

Keywords: recycling, composite plastics, compounded plastics, mechanical reprocessing, disintegrator technology, recovery of plastics.

KOKKUVÕTE

KOMPOSIITPLASTIDE RINGLUS (TÖÖTLEMINE JA TAASKASUTUS)

Komposiitplastijäätmed, nii tööstus- kui ka olmejäätmed, on probleemiks kogu maailmas, sest neid ei ole võimalik töödelda ja taaskasutada nagu enamikku termoplaste. Eestis on komposiitplasttooteid valmistavatel ettevõtetel (sanitaartehnika, paatide, suusabokside jms. valmistajad) oma tootmisjäätmetega samuti probleeme – käesoleval ajal ladustatakse neid prügilasse, tulevikus on see aga keelatud. Tööstuslikud komposiitplastijäätmed koosnevad termoplastist (PMMA või ABS), millest valmistatakse vaakumvormimise teel näiteks vannikoorikuid, ja tugevdavast reaktoplastist, mille maatriksiks on polüestervaik ning armatuuriks klaaskiud – klaasplast (GFP).

Ülaltoodud plastijäätmetele lisanduvad olmejäätmed (elektroonika ja kodumajapidamisseadmed), mis tuleb peale kasutuselt kõrvaldamist vastavalt ELi elektroonikaromu direktiivile (1. grupp – suured valged kodumasinad) kokku koguda ja 80% nende massist, kas korduv- või taaskasutusse suunata. Seega tuleb töödelda ka nende toodete massist 1/3 moodustav komposiitplastist koorik.

Komposiitplastijäätmetele PMMA+GFP ei ole siiani maailmas ühtegi töötlemis- ega materjali taaskasutuse meetodit välja töötatud ja sellest tulenevalt on käesolevas väitekirjas võetud vaatluse alla komposiitplastist tootmisjäätmete ja elektroonikaromus sisalduvate komposiit- ja kompaundplastijäätmete ja multimaterjalide mehaaniline töötlemine desintegraatorjahvatuse teel.

Käesoleva töö eesmärgid olid:

1. Töötada välja probleemsete plastijäätmete töötlemistehnoloogia, kasutades mehaanilisi meetodeid, uurida tootmises tekkivate komposiitplastijäätmete, elektroonikaromus sisalduvate plastijäätmete ja multimaterjalide ringlusse võtuks põrkejahvatust ja saadud teisese plasti – plastipulbri – taaskasutamise võimalusi. Optimeerida desintegraator-jahvatuse tehnoloogiat saamaks etteantud omadustega (osiseline koostis, osakeste kuju ja tehnoloogilised omadused) plastipulbreid.

2. Teisese plasti alusel töötada välja uus täiteaine PMMA-plastipulber, millel on optimaalne tihedus ning head tehnoloogilised omadused, ja uued tooted taaskasutatava plastipulbri baasil, kasutades numbrilise modelleerimise, mehaanilise katsetamise ning eksperimentaaltootmise tehnoloogiat.

3. Kavandada elektroonikaromus sisalduvate plastijäätmete kogumise, sorteerimise, töötlemise ja taaskasutamise süsteem Eesti jäätmekäitlejatele, saavutamaks ELi elektroonikaromu direktiiviga kehtestatud eesmärgid materjali taaskasutamiseks.

Töö eksperimentaalosas uuriti komposiit- ja kompaundplastijäätmete ning multimaterjalide töödeldavust desintegraatortehnoloogiat kasutades. Selleks kasutati mitut tüüpi desintegraatorveskeid ja jahvatussüsteeme (laboratoorsed ja pooltööstuslikud; otse-, selektiiv- ja/või separatsioonjahvatus). Materjalide jahvatusprotsess koosnes reeglina kolmest etapist: eelpurustus, eeljahvatus ja lõppjahvatus. Uuriti materjalide jahvatatavust, osiselist koostist ja osakeste kuju sõel- ja kujutisanalüüsi meetodeid kasutades. Viidi läbi katsed PMMA-pulbri tehnoloogiliste omaduste määramiseks. Saadud katsebaasi põhjal modelleeriti kahe erineva arvutusmeetodiga (närvivõrgud ja kuup-B-splainid) puiste- ja rappetiheduse pinnamudelid ning leiti pinna globaalsed ekstreemumid (minimaalne ja maksimaalne tihedus). Uuriti saadud

täiteainete kasutatavust uute komposiitmaterjalide valmistamiseks *solid surface* tehnoloogiaga ning saadud komposiitplastide mehaanilisi omadusi (tõmbetugevus, kõvadus, kulumiskindlus). Katsetulemuste põhjal valiti välja parimate mehaaniliste ja tehnoloogiliste omadustega komposiitplast, mida kasutati uue toote – kraanikausi – valmistamiseks eksperimentaaltehnoloogiat kasutades.

Tulemused ja järeldused:

1. Reaktoplastide nagu ka komposiitplastide ringlusse võtuks ei ole välja töötatud tehnoloogilisi lahendusi. Nende töötlemiseks on keemiliste ja termiliste meetodite, nagu gaasistamine, algaineteks lagundamine ja energiakasutus, kõrval perspektiivsed ka mehaanilised meetodid. Mehaaniline töötlemistehnoloogia on üks peamisi võimalusi reaktoplastidest komposiitplastide töötlemiseks.

2. Põrkejahvatustehnoloogia ja desintegraatorjahvatussüsteemid on perspektiivsed sekundaarse toorme – plastipulbri – saamiseks komposiitplasti ja kompaunditud plasti jäätmetest. Komposiitplastijäätmete (PMMA+GFP) separatsioon- ja/või selektiivjahvatamine desintegraatorveskeis võimaldab saada PMMA-plastipulbrit etteantud osiselise koostise ja tehnoloogiliste omadustega (puistetihedus ja voolavus).
   Desintegraatorjahvatusel saadud tulemusi on võrreldud teiste levinud plastijäätmete töötlemise tehnoloogiatega (horisontaalpurustid ja granulaatorid) ning on välja toodud desintegraatortehnoloogia eelised:

- plastkomposiidi komponentide samaaegne separeerimine jahvatusprotsessis (ei ole võimalik purustite ja granulaatoritega),
- madalam töötlemise erienergia.

   Multimaterjalidest (komposiitplast + metalne materjal) koosnevate jäätmete (näiteks personaalarvutite trükiplaadid) töötlemine on võimalik ainult selektiivjahvatust kasutades: eraldatud komposiitplast on sobilik kasutamiseks täiteainena uute komposiitplastide valmistamisel, metalsed komponendid aga kasutamiseks metallurgias.

3. Desintegraatorjahvatusel teel saadud PMMA-plastipulbrist kavandati uus täiteaine järgmiste tehnoloogiliste omadustega:

- maksimaalne tihedus ja parimad voolavusomadused on täiteainel jämeda ja peene osise 1:1 massijaotuse korral;
- optimaalseks (tugevus, majanduslik külg) maatriksi ja täiteaine vahekorraks väljatöötatud komposiitmaterjalis on 40 massi% vaiku ja 60 massi% kahefraktsioonilist täiteainet.

4. PMMA-pulbri pakketiheduse numbrilisel modelleerimisel osutusid sobilikuks mõlemad uuritud lähendused (närvivõrgud ja kuup-B-splain). Täpse analüütilise lahenduse puudumise tõttu antud probleemile ei saa välja arvutada ja võrrelda kuup-B-splaini- ja närvivõrkude (NN) mudelite vastavaid absoluutseid vigasid. NN-meetodi eeliseks võib lugeda suuremat paindlikkust tingituna pinnasileduse parameetri sisaldumisest mudelis. Samuti võib välja tuua NN-mudeli rakendamise lihtsuse, sest mudel põhineb MATLABi sisseehitatud funktsioonidel ja töövahenditel. Täiteaine tiheduse ekstremaalsed väärtused sõltusid modelleerimisel kasutatud pinnasileduse parameetri väärtusest. Maksimaalne rappetihedus vastas pinnasiledusele 0,5;

pinnasileduse parameetri suurendamisel 2,0-ni kahanes maksimaalne rappetihedus 0,77 ni. Modelleerimise tulemustele toetudes võib välja tuua järgmist:

- teatud algandmete korral koondusid algoritmid *patternsearch* ja *fmincon* vastavalt globaalseks ja lokaalseks miinimumiks.
- funktsioon *fmincon* näitas paremaid tulemusi arvutusaja suhtes, töötades märkimisväärselt kiiremini kui funktsioon *patternsearch*.

5. Uut väljatöötatud PMMA-plastipulbrist täiteainet kasutati eksperimentaalse kraanikausi valmistamisel *solid surface* tehnoloogiaga. PMMA-plastipulbrist valmistatud kraanikausi mass on ligi kaks korda väiksem, võrreldes tavapärase kaltsiumkarbonaadist täiteainega kraanikausist.

6. Välja on pakutud komposiitplastide ringluse skeem Eesti jäätmekäitlejatele (kogumine, sorteerimine, töötlus, taaskasutus), mis sisaldab mehaanilise töötluse tehnoloogiat nende taaskasutamiseks.

ELULOOKIRJELDUS

1. Isikuandmed

Ees- ja perekonnanimi	Jaan KERS
Sünniaeg ja –koht	11.07.1978 Tallinn
Kodakondsus	Eesti
Perekonnaseis	Vabaabielus
Lapsed	Jaana (sünd. 2005)

2. Kontaktandmed

Aadress	Mehaanikateaduskond, Materjalitehnika instituut, Tallinna Tehnikaülikool (TTÜ), Ehitajate tee 5, 19086 Tallinn, Eesti
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3. Hariduskäik

Õppeasutus	Lõpetamise aeg	Haridus
Tallinna Tehnikaülikool	2002-k.a.,	Masina- ja aparaadiehitus/ doktoriõpe, juhendaja Priit Kulu
Tallinna Tehnikaülikool	2002	Tootmistehnika/ tehnikateaduste magister
Tallinna Tehnikaülikool	2000	Tootmistehnika/ tehnikateaduste bakalaureus
Tallinna Kunsti Gümnaasium	1996	Keskharidus

4. Keelteoskus

Inglise keel	kõrgtase
Saksa keel	kesktase
Vene keel	kesktase
Soome keel	kesktase
Eesti keel	emakeel

5. Täiendõpe

Õppimise aeg	Õppeasutuse või muu organisatsiooni nimetus
2006	TTÜ – inglise ärikeel
2004	TTÜ – projektijuhtimine

6. Teenistuskäik

Töötamise aeg	Ülikooli, teadusasutuse või muu organisatsiooni nimetus	Ametikoht
Alates 2006	TTÜ	assitent
2000-2006	AS Balteco	projektijuht

7. Juhendatud lõputööd

Kristo Kekkonen, bakalaureus, Plastkomposiitmaterjalide abrasioonkulumine, TTÜ, Tallinn, 2006

8. Teadustöö põhisuunad

Komposiit- ja kompaunplastjäätmete ning multimaterjalide töötlemis- ja taaskasutustehnoloogiate välja töötamine Eesti jäätmekäitlejatele.

9. Teised uurimisprojektid

INTERREG IIIA programmist finantseeritav projekt – RePlast FinEst plasti (sh. elektroonikaromus sisalduva plasti) taaskasutamine (15.03.2005-31.10. 2007) .

CURRICULUM VITAE (CV)

1. Personal data

First name and surname	Jaan KERS
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Citizenship	Estonian
Family status	Unregistered marriage
Children	Jaana (sünd. 2005)
Current position	Assistant

2. Contact

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3. Education

Educational institution	Graduated	Speciality
Tallinn University of Technology	2002-included	Mechanical engineering/ *Ph.D student*, supervisor Priit Kulu
Tallinn University of Technology	2002	Production Engineering/ *MSc*
Tallinn University of Technology	2000	Production Engineering/ *B.Sc*
Tallinn Secondary Art Gymnasium	1996	Secondary education

4. Language skills

English	Advanced
German	Intermediate
Russian	Intermediate
Finnish	Intermediate
Estonian	Native language

5. Further training

Apprenticeship	Educational or other organizations
2006	TUT – Business English course
2004	TUT – project management

6. Professional employment

Period	Organisation	Position
since 2006	TUT	Assistant
2000-2006	AS Balteco	Project manager

7. Supervised diploma work

Kristo Kekkonen, B.Sc, Abrasion wear of plastic composites, TUT, Tallinn, 2006

8. Main research interest

Development of reprocessing and recovery system of composite, compounded plastic scrap as well as multi-materials for recyclers in Estonia

9. Other research projects

RePlast FinEst project for recovery of plastics (included plastic fraction from WEEE). Project is financed from INTERREG IIIA program and its duration is 15.03.2005-31.10. 2007.