Secondary use of ABS co-polymer recyclates for the manufacture of structural elements using the FFF technology

Piotr Czyżewski, Marek Bieliński and Dariusz Sykutera
Department of Materials Enginnering and Polymer Processing, UTP University of Science and Technology, Bydgoszcz, Poland, and
Marcin Jurek, Marcin Gronowski, Łukasz Ryl and Hubert Hoppe
UTP University of Science and Technology, Bydgoszcz, Poland

Abstract

Purpose – The aim of this paper is presenting a new application of material obtained from the acrylonitrile butadiene styrene (ABS) recycling process from electronic equipment housings. Elements of computer monitors were used to prepare re-granulate, which in turn was used to manufacture a filament for fused filament fabrication (FFF) additive manufacturing technology.

Design/methodology/approach – The geometry of test samples (i.e. dumbbell and bar) was obtained in accordance with the PN-EN standards. Samples made with the FFF technology were used to determine selected mechanical properties and to compare the results obtained with the properties of ABS re-granulate mould pieces made with the injection moulding technology. The GATE device manufactured by 3Novatica was used to make the prototypes with the FFF technology. Processing parameters were tested with the use of an Aflow extrusion plastometer manufactured by Zwick/Roell and other original testing facilities. Tests of mechanical properties were performed with a Z030 universal testing machine, a HIT 50P pendulum impact tester and a Z3106 hardness tester manufactured by Zwick/Roell.

Findings – The paper presents results of tests performed on a filament obtained from the ABS re-granulate and indicates characteristic processing properties of that material. The properties of the new secondary material were compared with the available original ABS materials that are commonly used in the additive technology of manufacturing geometrical objects. The study also presents selected results of tests of functional properties of ABS products made in the FFF technology.

Originality/value – The test results allowed authors to assess the possibility of a secondary application of used elements of electronic equipment housings in the FFF technology and to compare the strength properties of products obtained with similar products made with the standard injection moulding technology.

Keywords ABS recycling, Filament production, 3D printing, FFF technology, Processing properties, Mechanical properties

Paper type Case study

Introduction

Advantageous functional and processing properties of the acrylonitrile butadiene styrene (ABS) co-polymer and its combinations – for example with polyethylene terephthalate, Polycarbonate or Polymethylmethacrylate – make it a material used for manufacturing thin-wall moulds and housings of electronic equipment. Visible and continuous development of the electronic industry demonstrates itself mainly through the manufacture of new and innovative products. After a short period of use (up to 5 years), most of them lose their functional properties and constitute as a waste product. As a consequence, the amount of post-use waste in the E&E (Electrical and Electronic) industry, including used elements of electronic equipment housings, is rapidly rising. The management of waste made of ABS co-polymer is important because of the high cost of the primary materials used in the manufacture of electronic equipment (Plastics Europe, 2016).

Additive manufacturing technologies are currently one of the fastest developing methods of manufacturing prototypes and small batches of products connected with the processing of polymers (Bingheng *et al.*, 2015; Stucker, 2016; Stratasys, 2016; ASME, 2016). The fused filament fabrication (FFF) method, just as the fuse deposition modelling, uses extrudates (the so-called filament) as an input material, which is made of the ABS

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co-polymer or other polymers (3dprintingforbeginners, 2016; Feeney, 2016; SD3D team, 2016).

Considering the fact that the purchase cost of the filament is currently many times higher than the price of the ABS terpolymer as a granulate, the secondary use of the ABS recyclates from E&E equipment housings to produce the filament is economically justified. This activity is consistent with the most demanded tendency to use raw materials obtained from mechanical recycling to manufacture goods of high market value (up-cycling). It is also beneficial from the point of view of the reduction of CO_2 emissions and the higher importance of the mechanical recycling technology, which is preferred in the EU. The EU is planning to prohibit stockpiling of polymer waste by 2020. In some countries (i.e. Germany, Austria and Scandinavian countries), where the primary goal is to use the material potential of waste (mechanical recycling) by incineration, such legislation has already been made effective.

Additive technologies of manufacturing with the use of a polymer filament are studied in terms of, among others, determining mechanical properties, where the impact of process settings (Górski *et al.*, 2013; Sikorski *et al.*, 2013; Hossain *et al.*, 2013), printing orientation (Pepliński *et al.*, 2017; Górski *et al.*, 2014; Ahn *et al.*, 2002; Rodríguez *et al.*, 2001) and the impact of materials used (Makucha, *et al.*, 2016; Singh *et al.*, 2016; Tartakowski and Mydłowska, 2015) on the product quality are verified. Dimensional accuracy of products based on the standardised samples (Górecki, 2015; Siemiński and Rajch, 2014) and control geometries (Bieliński *et al.*, 2015; Czyżewski *et al.*, 2014; Bieliński *et al.*, 2013) is also subjected to verification.

The previously conducted studies on the secondary use of materials in additive technologies have been focused on the effective production of spatial objects out of the filament (Kreiger *et al.*, 2013; Peels, 2016; Horne, 2016a, 2016b; Długosz, 2016; Swiat druku 3D, 2016; Refil, 2016).

Various design solutions of printing heads of 3D printing machines that lean towards using granulate or re-granulate as an input material (Horne, 2016a, 2016b; Muñiz, 2016; Braanker *et al.*, 2016) are taken into consideration.

There are known attempts at managing polymer waste to obtain functional filament used in the RepRap devices (Baechler *et al.*, 2013). As opposed to the literature on the secondary use of ABS recyclates obtained by injection moulding, there is a lack of reports on the attempts at managing the ABS terpolymer waste to produce filament. The purpose of the present study is an attempt to use the ABS recyclate for direct manufacturing of the filament with expected and reproducible functional properties. It has been assumed that the quality of a filament from the secondary material is a derivative of mechanical recycling processes performed (especially, grinding and re-granulation moulding), and it also depends on the grain composition of the recyclate. The study aims at obtaining products with higher quality and dimensional reproducibility in the FFF technology.

Experiment

Research objective

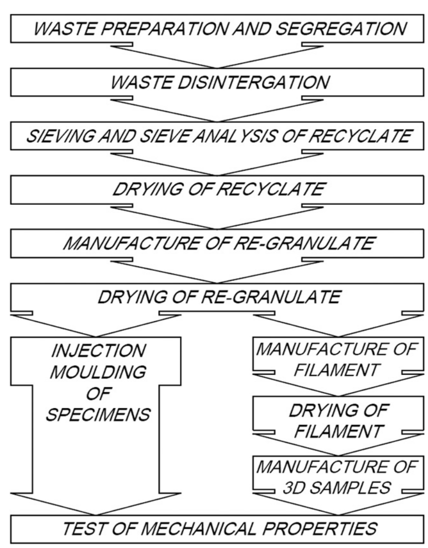
The study aimed at attempting the secondary use of used electronic equipment housings made of the ABS co-polymer. The moulded pieces subjected to mechanical recycling were selected in terms of external dimensions, wall thickness and a flow path length of a melted polymer in the primary processing (Figure 1).

Research methodology

The experiment proposed a procedure to allow the complete management of used electronic equipment housings through production of a filament used in the FFF additive technology (Figure 2). The housings were cleaned using air pressure without detergents and without damaging the continuity of the top layer of these components. Parts containing glue, labels and metal mouldings were separated. The prepared electronic equipment housings were initially cut with a hand-operated workshop guillotine to analyse the distribution of the wall thickness. Subsequently, the material was shredded using a Rapid 2a cutter shredder, where clearance between moving and static blades was set to 0.1 mm, and a sieve with openings of Ø8 mm in diameter was used. For the sieve analysis, sieves with the following opening sizes were used: Ø7, Ø5, Ø3.5 and Ø2 mm. Before being processed again, the recyclate had been dried at a temperature of 80°C for 4 h in the Binder FED-115 dryer (Germany). Then, using the granulate production line equipped with the W25-30D

Figure 1 An example of computer monitor housings made of ABS in the injection moulding technology and used in the research

Figure 2 Stages of the testing within secondary management of the ABS waste in the form of computer equipment housings



single-screw extruder manufactured by Metalchem (Poland) and with the cold re-granulation mill manufactured by Metalchem (Poland), the ABS co-polymer re-granulate was produced. An extrusion process was realised for the following temperature profiles of the extruder plasticising unit zone: I – 175°C; II – 210°C; III – 235°C; and head zone – 190°C. During the process(es), the extrusion screw worked at a rotation speed of 54 min^{-1}. During the re-granulate production, an extrusion head equipped with a nozzle of Ø3 mm in diameter was used. Before the next extrusion process, the re-granulate drying stage was repeated with same parameters as in the drying of the recyclate. The recyclate obtained was subjected to the sieve analysis to determine its grain distribution. Apart from using the ABS granulate for the manufacture of a filament, some part of the secondary material was used to manufacture test pieces in the injection moulding technology, with standard processing parameters applicable for that material (injection pressure: 80 MPa; injection time: 1.1 s; clamping pressure: 55 MPa; clamping time: 16 s; cooling time: 30 s; plasticising unit zone temperatures: IV – 235°C; III – 235°C; II – 225°C; I – 215°C; and dosage zone temperature: 40°C). The filament obtained was used to manufacture samples with the same geometrical features as the injection moulded piece with the FFF additive technology. For each level of inputs (variables), and for each of the compared technologies (the FFF, injection moulding), ten samples (ten repeats) were used. The dimensions were consistent with PN-EN ISO 527. The extrudate extrusion process efficiency was estimated at 5.62 $\mathrm{m{\cdot}min}^{-1}$ (mass efficiency: 0.85 kg · h^{-1}). Measurements to verify the diameter of the extrudate obtained were performed on-line with the use of a MarCal 16EWR digital calliper manufactured by Mahr (Germany) [Figure 3(a)]. The extrudate diameter measurement (filament for the FFF technology) was performed on its randomly chosen section (1,000 mm) every 10 mm [Figure 3(b)]. The test of the flow rate of different ABS filaments, including the one made of the ABS waste, was conducted using an Aflow plastometer manufactured by Zwick/Roell (Germany). For the tests of additive manufacturing, the GATE device, manufactured by 3Novatica (Poland), equipped with a working head with a nozzle of 0.5 mm in diameter was used [Figure 4(a)]. Process parameters of the FFF technology: head temperature: 235°C; table temperature: 80°C; print speed: 40 $\mathrm{mm{\cdot}s}^{-1}$; infill pattern: grid; infill percentage: 100 per cent; layer thickness: 0.2 mm; and printed time: 14 min. Transformation of models and generation of G code was done using Repetier-Host 1.0.6. 3Novatica (Poland).

Figure 3 Appearance of a filament obtained from the ABS re-granulate: a) on-line measurement of an extruded diameter during manufacture, b) finished product ready to be used in the FFF technology

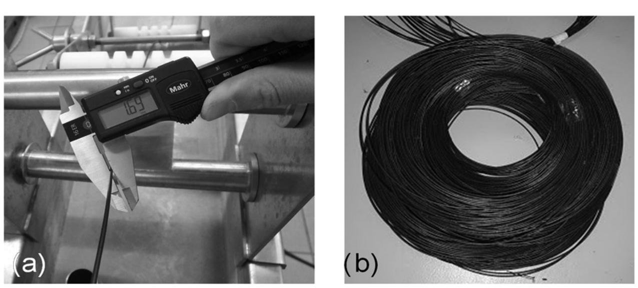
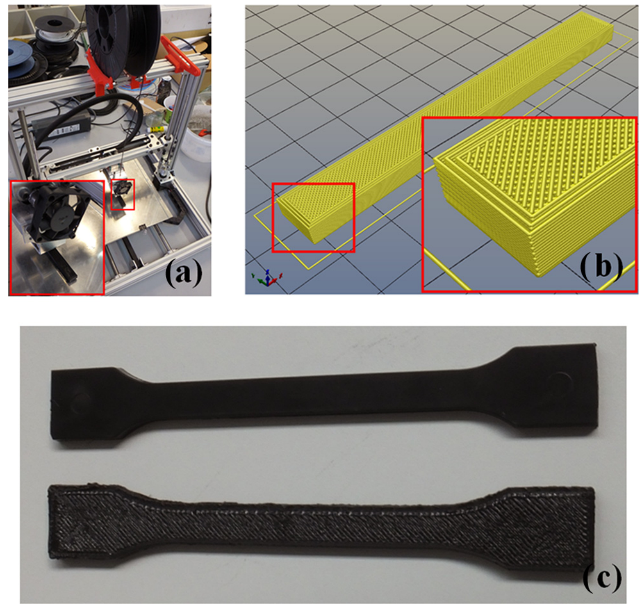


Figure 4 Additive manufacturing of a test sample in the FFF technology: a) Type G 3D printer manufactured by 3Novatica, b) Method of applying layers of material inside the sample, c) Example of samples intended for mechanical tests made in the injection moulding technology (upper) and the FFF technology (lower)

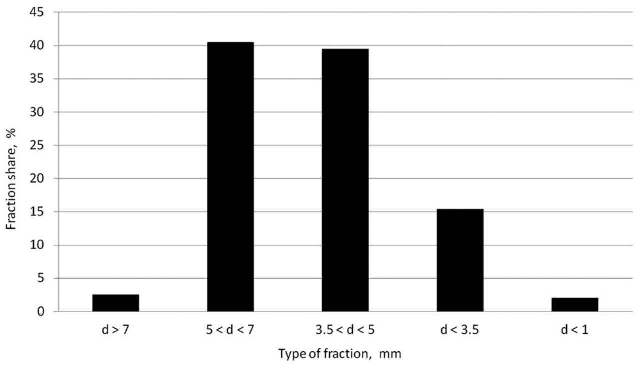


For the impact tests and the three-point static bend test, samples which were manufactured by the method of applying consecutive layers in accordance with Figure 4(b) were used. The filling method was chosen, whereby individual threads of a layer inside a sample were turned by 45° against the longest edge and by 90° against the previous layers. Figure 4(c) presents an example of the test samples made in the injection moulding technology and the FFF method and used in mechanical tests. To perform those tests, the Z030 universal testing machine, the HIT 50P pendulum impact tester and the Z3106 hardness tester – all manufactured by Zwick/Roell (Germany) – were used. Opta-Tech stereoscopic microscope (10× magnification) was used to evaluate the shape of the fractures. In addition, a detailed analysis of the surface topography of selected fragments was made using the Olympus Lext OLS 4000 con-focal microscope (Lens number 5, 1× magnification).

Results of the ABS recyclate properties tests

The results of the sieve analyses of recyclates were presented in Figure 5. It was found that the shredding of the housings in a cutter shredder provided recyclates with dominating grain sizes ranging from 3.5 to 7 mm, which constitutes 79.9 per cent of the total sample weight. Fractions with the grain size lower than 1 mm and higher than 7 mm constituted less than 5 per cent of the entire recyclate. The obtained distribution of particle sizes is similar to normal distribution, which is beneficial from the perspective of the secondary processing. The size of the dominating grains is similar to the size of granules of the original granulate. Such beneficial distribution of the recyclate grains arises from the appropriate parameters of the shredding process and the reproducible geometrical features of the initial feed (a housing with small dispersion of wall thickness).

Figure 5 Grain distribution of the ABS copolymer recyclate



Filament properties

The extrudate diameter varied within a range from 1.64 to 1.95 mm (extrudate average diameter D = 1.76 ± 0.06). Figure 6 demonstrates the distribution of the extrudate diameter for the eight measurement ranges adopted. In total, 85 per cent of all the diameter measurement results fall within the range from 1.68 to 1.83 mm, which indicates that the filament is suitable for using in the FFF technology. It was found that the extrudate was characterised by the local thickenings of diameters in the range from 1.84 to 1.95 mm, which constitutes 9 per cent of all the measurements. The reason for this geometrical instability is an apparent unstable viscosity of a melted ABS re-granulate and a gas entrapped inside, which is produced in the decomposition process of low-molecular substances, printing ink, etc., during the mixing process in the melting system of the extruder. Table I contains a comparison between the weight- and volume-related flow rate of the commercially available filaments, and the filament obtained from the elements of electronic equipment housings. Values of melt flow rate and melt volume rate obtained for the extrudate from the ABS recyclates clearly demonstrate that the secondary material was achieved which was characterised by its flowability, allowing it to be processed in the melting system of both an injection moulding machine and an extruder. Observations recorded during the plastometer test also allowed

Table I Comparison between the flow rate values for selected commercially available ABS filaments and the ABS filament obtained from the waste elements of computer monitor housings

Trade name of the filament	Melt flow rate (g/10min)	Melt volume rate (cm^3/10min)
3 Novatica ABS PREMIUM (blue)	22.8 ± 0.55	21.72 ± 0.53
Orbitech smartABS	9.07 ± 0.02	8.64 ± 0.02
3Novatica ABS PREMIUM (white)	19.12 ± 0.31	18.21 ± 0.3
Stratasys P430	34.51 ± 0.6	32.87 ± 0.58
Makerbot MP03886	7.54 ± 0.23	7.19 ± 0.22
Filament from recyclate	14.81 ± 0.25	14.11 ± 0.24

the appropriate selection of processing settings (i.e. board temperature, head temperature, material extrusion rate, board and head movement velocity) to be applied during the process of additive manufacturing.

Results of mechanical properties tests

The comparison between sample runs of the tensile test of injection moulded samples and 3D printed samples is demonstrated in Figure 7. Different shapes of the tensile curves are caused by different macro-structure of the sample made by the FFF technology (the displacement of applied layers and threads in layers), as compared to the moulded piece. Within the tension range from 0 to 8 MPa with elongation ca. 0.6 per cent – $\sigma(\varepsilon)$ curves for both samples are overlapping. It is beneficial because of the possibility to substitute moulded pieces with 3D printed pieces without losing their strength properties. Moulded pieces produced through injection moulding are characterised by higher – in comparison with printed pieces – tensile strength R_m and higher relative deformability, which is caused by the influence of high pressure on the material during the injection and clamping stages. This gives rise to a solid material, where macro-particles are considerably closer to one another, which leads to higher van der Waals forces. Such a different behaviour of the two samples is confirmed by a three-point bend flexural test. Products made in the FFF technology demonstrate lower strength in terms of elastic deformations (Figure 8). Table II

Figure 6 Distribution of diameters of extrudate cross section in the 1000 mm measurement test

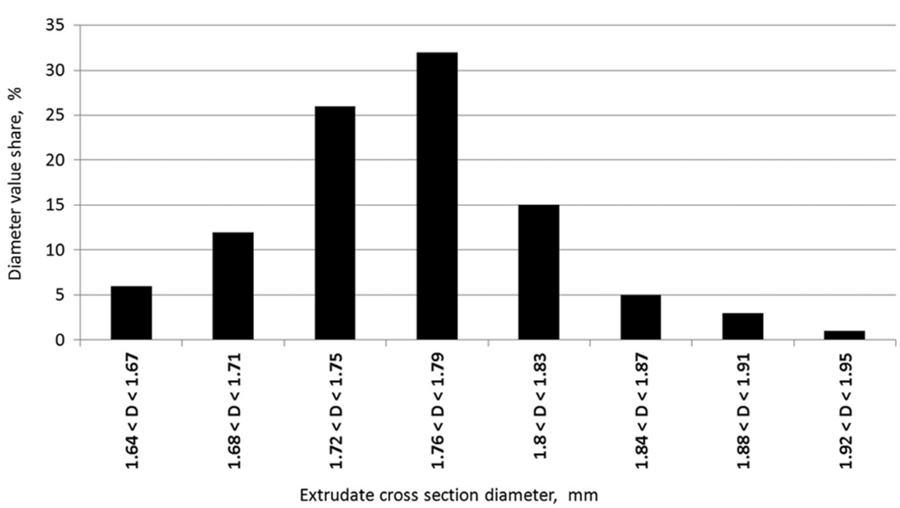


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Figure 7 Sample runs during the static tensile strength test for samples made in the injection moulding technology and samples made in the FFF technology

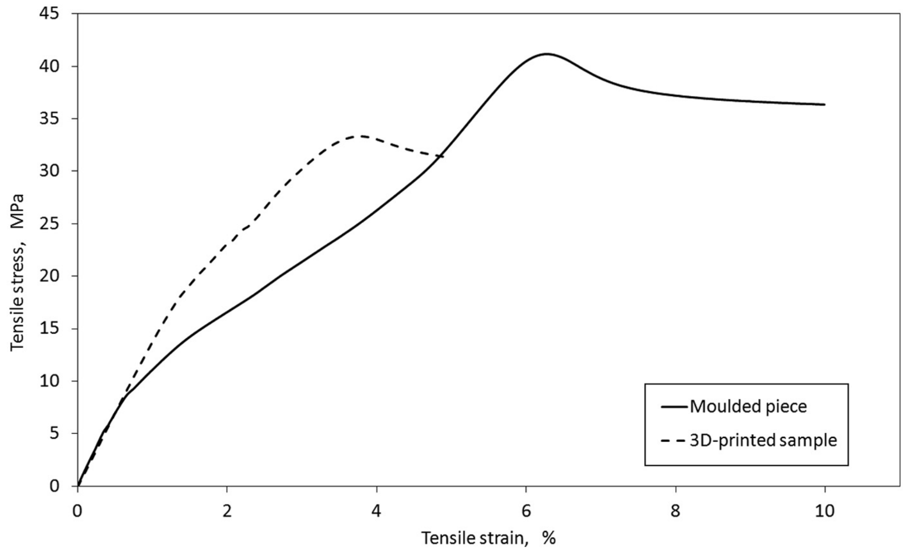


Figure 8 Sample runs during the bend test for samples made in the injection moulding technology and samples made in the FFF technology

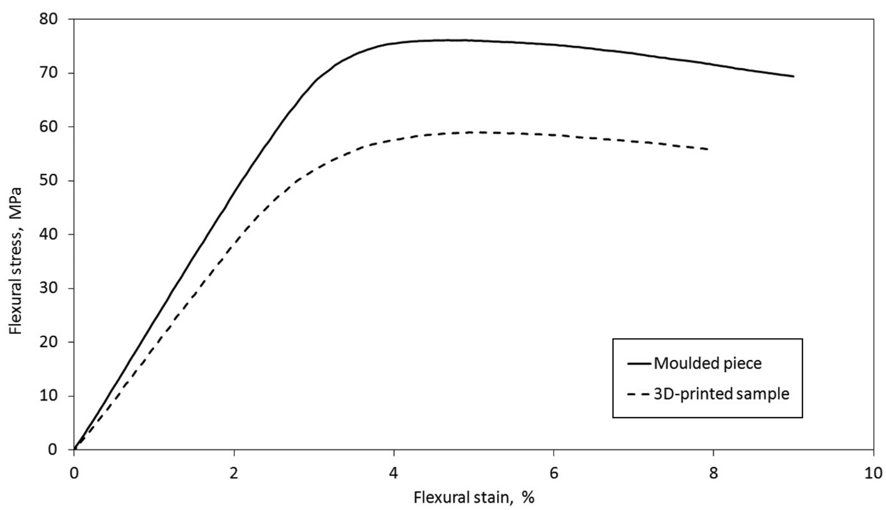
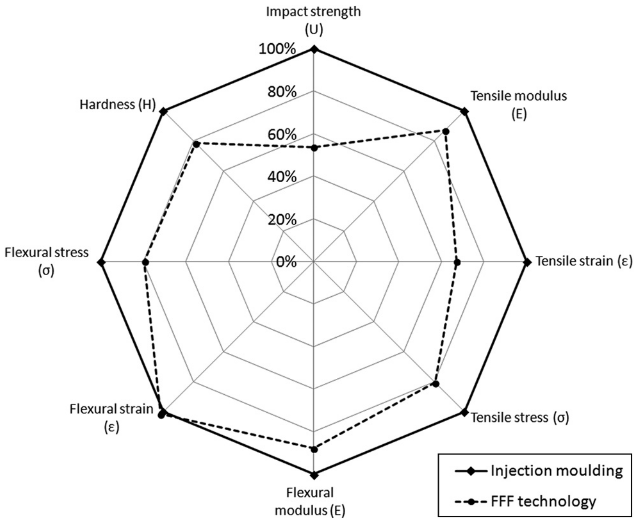


Table II Results of mechanical properties tests for samples made by injection moulding and samples made in the FFF technology

Sample type	Impact strength (kJ/m^2)	Tensile test Young's modulus (MPa)	Tensile test Stress (MPa)	Tensile test Strain (%)	Flexural test Young's modulus (MPa)	Flexural test Stress (MPa)	Flexural test Strain (%)	Hardness (MPa)
Injection moulding	51.59 ± 12.78	1,586 ± 61	41.75 ± 0.47	5.56 ± 1.17	2,169 ± 104	75.5 ± 0.45	4.73 ± 0.13	88.45 ± 7.66
FFF technology	27.82 ± 4.7	1,382 ± 46	33.6 ± 0.5	3.72 ± 0.1	1,906 ± 109	58.0 ± 2.26	4.8 ± 0.2	69.47 ± 2.42

juxtaposes the results of mechanical properties tests for the samples made by injection moulding and samples made in the FFF technology. To compare the results of the selected mechanical tests of samples made in the injection moulding technology and those made in the FFF technology, a radar chart has been prepared (Figure 9). The results for the injection moulded samples are reference values (a solid line) for the samples made in the FFF technology (broken line). The values of stress and Young's modulus (during the tensile strength test and the bend test) for samples obtained in the FFF technology exceed 80 per cent of the values of the mechanical properties of moulded pieces made by the injection of the ABS re-granulate.

Figure 9 Comparison between mechanical properties of samples made in the injection moulding technology and samples made in the FFF technology



Deformation of the printed sample during the tensile strength test is 67 per cent, while during the bend test, it is 101.5 per cent of the values obtained for injection moulded samples. Hardness and impact strength of the printed samples are considerably lower and ranges between 50 and 80 per cent in comparison with the solid ABS moulded pieces.

Figures 10 and 11 illustrate sample fractures obtained during impact tests. There were significant differences in the topography of the analysed fractures resulting from the manufacturing technologies applied (Figure 10). The injection moulding mould fracture is characterised by a flat surface, which indicates a brittle fracture in the impact load test [Figure 10(a)]. The noticeable colour change at the top of the fracture is because of the appearance of plastic

Figure 10 Exemplary fractures in the impact tests: a) injection moulding, b) FFF technology

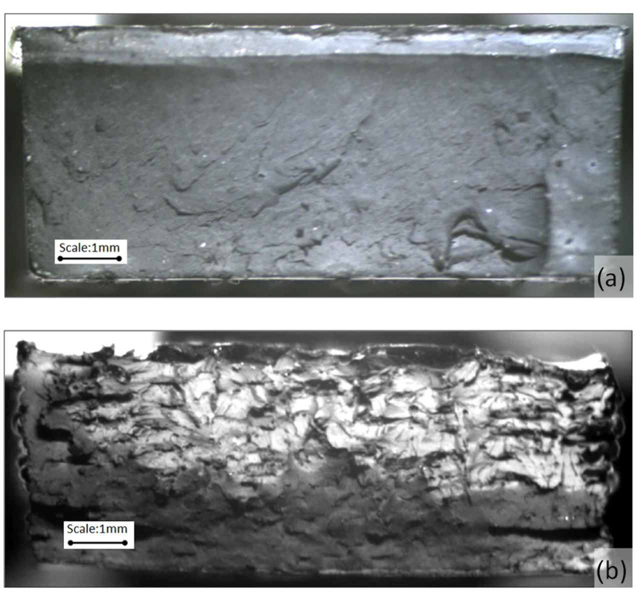
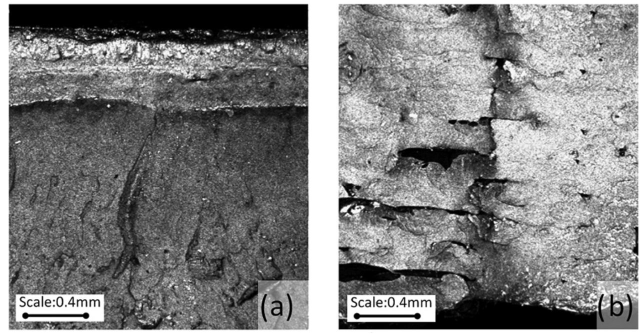


Figure 11 Difference in material structure of the samples (fractures) of the tested technologies: a) injection moulding, b) FFF technology



deformation in this zone, which is always accompanied by a change in hue. In the case of a fracture of a 3D-printed sample, a set of micro-plains is observed in an area of the fracture. There are discontinuity areas and plastically deformed sections observed between them [Figure 10(b)]. This is because of the process of incremental overlap of further layers of material and the filament diameters. Melted fragments are laid on successive layers, and after cooling off from the nozzle head, they are rapidly cooled down. Its melted fragments are laid on subsequent layers, and after leaving the nozzle head, they are quickly cooled down. For this reason, the adhesion between the sequentially laid tapes and between the tape and the layer below is limited, and this phenomenon increases the absence of process pressure (which is important in the injection process). The surface effects aforementioned can be closely seen with con-focal microscope. In Figure 11, in the case of the mouldings' images of breakthrough fragments observed at higher magnification are characterised by the domination of a flat surface with a deformed fragment [Figure 11(a)], whereas for the FFF samples, there are clearly visible areas of the structure discontinuity between the layers and tapes in the form of holes [Figure 11(b)]. Empty spaces with irregular dimensions (often multiplied in a cross section) are responsible for initiating material separation between the layers and in the single layer (between single strips). Particularly, large areas of discontinuity (macro-pores) in the volume of the FFF sample constitute the notches in which the stresses accumulate and which are the sources of crack initiation.

Conclusions

The tests confirm the possibility of secondary processing of used electronic equipment housings into an extrudate used as a filament in the FFF additive manufacturing technology. The mechanical properties of testing structural elements depend on manufacturing technology and process parameters, mainly temperature and pressure. Quality of the obtained FFF samples depends on repeatability and control of the properties of the feed material (ABS waste), recycling method and used parameters of grinding and extrusion processes and FFF parameters. The main cause of the significant deterioration in mechanical properties is the occurrence of discontinuity areas in the volume of the FFF samples, which results from the very essence of this manufacturing technology. However, the dimensions of these "openings" are probably a function of the FFF process parameters (especially temperature), but they, indirectly, also depend on the prefabricated mechanical recycling processes and a continuous filament production. The next step will be a detailed study of the distribution and size of discontinuity areas in the volume of the FFF samples using computer tomography and image analysis. The pursuit of a solid structure is a condition for the improvement of mechanical properties similar to that of the products obtained by injection moulding. The further research works on the use of ABS re-granulates for the filament of manufacture will focus on reducing the number of finished products' preparation stages. In the industry, they will have a positive impact on the possible applications of re-using ABS waste like a structural secondary material to apply in additive manufacturing.

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Corresponding author

Piotr Czyżewski can be contacted at: p.czyzewski@utp.edu.pl