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# Composites based on recycled polystyrene waste with tuff microparticles

# Kompozyty na osnowie odpadowego polistyrenu z mikrocząsteczkami tufu

**Abstract**

Nowadays electric and electronic equipments are produced on a large scale, for example, personal computers. Therefore, more and more waste is recycled. To increase the strength properties of recycled materials, they are reinforced with various types of fillers such as natural. Over the last few years, fillers from tuffs of volcanic origin have been gaining in popularity. It is characterized by a low mass, high hardness, high matrix adhesion, which allow for uniform distribution of particles in the volume of the material. In this paper, composites based on recycled polystyrene waste with tuff microparticles in mass weight of 10% and 20% were produced by injection moulding. The basic mechanical properties at room temperature (+24°C) -24 and +80°C were evaluated. The aim of the study was to demonstrate the possibility of using the mineral filler particles as compatibilizer for recycled polystyrene waste of poor quality.

**Keywords:** mechanical properties, natural fillers, ecology, recycling

**Streszczenie**

Obecnie urządzenia elektryczne i elektroniczne są produkowane na dużą skalę, np. komputery osobiste. Dlatego coraz częściej odpady te są poddawane recyklingowi. Aby zwiększyć właściwości wytrzymałościowe materiałów z recyklingu, wzmacnia się je różnego rodzaju napełniaczami, np. naturalnymi. Tuf charakteryzuję się niską masą, wysoką twardością i wysoką adhezją matrycową, które pozwalają na równomierny rozkład cząstek w objętości materiału. W artykule opisano kompozyty na osnowie odpadowego polistyrenu z cząsteczkami tufu w ilości masowej 10% i 20% wytworzone w procesie formowania wtryskowego. Wykonano podstawowe badania właściwości mechanicznych w temperaturze pokojowej (+24°C) oraz w skrajnych temperaturach eksploatacji -24 i + 80°C. Celem badań była ocena możliwości wykorzystania cząstek napełniacza mineralnego jako kompatibilizatora dla recyklatu polistyrenu o niskiej jakości.

**Słowa kluczowe:** właściwości mechaniczne, naturalne napełniacze, ekologia, recykling

## 1. Introduction

One of the major challenges that have emerged in this century is the increasing amount of commercial waste. This is especially true for developed countries, due to the rapid rise in solid precipitation resulting from swift population growth, urbanization, industrialization and economic development [1]. The majority of the waste is generated from plastic waste, due to the widespread use of it in everyday life. In the last two decades increasing interest has been observed in technological equipment which generates growth in waste from electrical and electronic equipment (WEEE) [2]. Nowadays electric and electronic equipments are produced on a large scale, for example, personal computers present a medium lifetime at around 5 years and cellular phones at 2 years [3]. Thus, industry is increasingly seeking to recycle and reuse plastics for economic and environmental reasons [4]. Many publications on plastics recycling have been written [5–7]. Their authors were mainly concerned with the use of polymers compostable for everyday use.

The most commonly used WEEE materials are thermoplastic copolymers like ABS, HIPS and PC, among others. HIPS has a wide range of uses; for instance, it is used for the WEEE, production of packaging materials, automotive components, toys and medical applications [8]. It is an aromatic polymer, which accounts for approximately 18 million (6%) tonnes of the world plastics market. It is a multiphase copolymer system which is formed by polybutadiene rubber particles dispersed in a matrix structure of polystyrene. Its main advantages include good impact resistance, ease of moulding and processing, stability and low cost.

In the literature, there are not many studies on recycled HIPS, mainly reported are the results of creating HIPS and ABS blend with appropriate compatibilizers [9, 10]. However, Garcia-Ivars et al. used commercial and recycled HIPS for preparation of flat-sheet membranes. Their research showed that recycled HIPS membranes have higher permeation of flux and a more porous structure. Additionally, compared to HIPS membranes recycled membranes have better antifouling capabilities and higher humic acid rejection [11].

One of the methods of reinforcing thermoplastic copolymers is the addition of various fillers such as glass or carbon fibres, but also natural fillers such as minerals, wood and many others. The use of fillers in polymeric materials is not only dictated by lower production costs, but also by improved physical and mechanical properties, dimensional stability and heat resistance. The most popular fillers used for thermoplastic compounds added to balance stiffness: calcium carbonate, talc, wollastonite and kaolin. Calcium carbonate is the cheapest and most commonly used mineral filler. In addition to economic issues, it is also used to reduce shrinkage and achieve better surface finish. Talc addition improves the stiffness of thermoplastic compounds and provides better dimensional stability. Wollastonite, either alone or with fibreglass, helps to improve the surface finish of the products. Kaolin provides good impact modifications. The future of filler development is related to pioneering materials that are meant to replace many traditional materials.

Over the last few years, fillers from tuffs of volcanic origin have been gaining in popularity. A tuff is a kind of light, compact porous sedimentary rock belonging to the crust rocks. It consists mainly of organic grain materials bound by silica or clay. It is characterized by a low mass, high hardness and high matrix adhesion, which allow for uniform distribution of particles in the volume of the material. Thanks to their use as a filler in polymer composites, the stiffness and hardness of the final material can be increased, as confirmed by studies [12–14]. In addition, it increases the temperature at which thermoplastic polymers can work and reduce their shrinkage.

Zmudka et al. studied the effect of volcanic tuff as a filler of thermoplastic polymers. They produced composites based on polyethylene, polyamide and polypropylene containing 10, 15, 20 wt % tuff filler. The mechanical properties, Vicat softening temperature and melt flow index were determined. They observed higher flexural modulus, higher temperature resistance, and shrinkage reduction. The composites obtained were characterized by a high flow index, which allows the injection of products with a complex shape [14].

Another important feature of polymer composites filled with tuff is a high flow index, which translates into an ease of forming products from this material using injection moulding. Yet another advantage is their low price and high availability, so that they can replace other more expensive additives such as antipyretic, pigments etc. in order to reduce production costs. Composites with tuff reinforcement can be used in electronics as friction materials and components that require increased creep and hardness. Properly ground tuffs result in polymer nanocomposites which have a very high temperature resistance and are also characterized by good mechanical properties (especially elastic range) as well as resistance to chemical and atmospheric agents.

As research shows, it is possible to use tuff as a plastic compatibilizer. Kuciel et al. confirmed that the addition of 5% mineral filler to PEHD recyclate improves the miscibility of waste polyolefins, increasing more than twice the strain at break, with almost unchanged strength. The obtained test results make natural mineral fillers like tuff an interesting alternative to expensive additives [12].

In this study, composites based on recycled polystyrene waste with 10 wt % and 20 wt % tuff microparticles were produced. The research part of the study was aimed at determining the basic mechanical properties of composites at room temperature (24°C) as well as extreme operating temperatures of -24 and +80°C. The values of lowered and elevated temperatures reflected the lowest and the highest temperatures at which recycled polystyrene waste composites can be used. Additionally, an analysis of the microstructure of the tuff particles and the adhesion to matrix was performed using a scanning electron microscope (SEM). Also the influence of tuff as an additive for improving miscibility was evaluated.

## 2. Experimental

The recycled polystyrene waste from technological waste of high-impact polystyrene (HIPS) from the KLGS plant in Myślenice (Poland) was used as a matrix. Recycled HIPS was obtained from disassembly of obsolete monitors and keyboards previously used as parts of personal computers.

As a filler tuff supplied from a mine in Filipowice (Poland) was used. Tuff is a solid pyroclastic rock, consists mostly of minerals such as sanidine, kaolinite, illite, biotite and quartz. The particle size in a range of 5-20 μm were obtained by grinding on a Retsch ZM 200 mill, which after grinding was rinsed in 1 molar hydrochloric acid and then calcined at 800°C.

Standard dumbbell samples were manufactured on the Engel ES 200/40 HSL injection moulding machine in accordance with PN-EN ISO 3167 standard with no previous process of extrusion and regranulation. The parameters of the injection process were as follows:

- zone temperature: 230 [°C], 235 [°C], 240 [°C], 245 [°C],
- a speed of screw rotation: 50 [rpm],
- mould temperature: 50 [°C],
- injection pressure: 80 [MPa],
- injection time: 2 [s],
- a holding pressure time: 5 [s],
- cycle time: 45 [s],
- cooling time: 25 [s].

The mechanical properties were obtained by using a universal MTS Criterion 43 (30 kN force capacity) testing machine with an MTS axial extensometer at room temperatures (24°C), -24 and +80°C by putting samples in a thermal chamber (Instron) for 30 minutes. The values were obtained from an average at least of 5 specimens. The tensile test (tensile strength ($\sigma_M$), modulus of elasticity ($E_t$) and strain at break ($\varepsilon_B$)) was carried out according to the ISO 527 standard with a constant cross-head speed of 5mm/min. The flexural three-point bending test (flexural modulus ($E_f$) and flexural stress at 3.5% strain ($\sigma_s$)) was performed with a constant cross-head speed of 5 mm/min (ISO178).

A Charpy impact test (ISO 179-1) was carried out using a Zwick/Roell HIT5.5P testing machine on unnotched samples. The microstructure of the obtained composites was observed with a JEOL JSN5510LV scanning electron microscope on gold-sputtered fracture surfaces specimens.

## 3. Results and Discussion

The symbols and mechanical properties determined in the tensile test for samples under standard condition are shown in Table 1. It can be observed that the elastic modulus increased proportionally to the increasing of tuff content. The addition of 20 wt % tuff led to an approx. 30% increase in stiffness, which is the observed effect of adding mineral fillers. The tensile strength slightly decreases by approximately 5% remaining constant with increasing amounts of tuff microparticles in the composite. There were no significant changes for deformability.

Figures 1, 2 and 3 show the comparison between the basic strength properties determined by the static tensile test of recycled polystyrene waste and its composites with 10 wt % and wt 20% tuff particles at room temperature (24°C) as well as extreme operating temperatures of -24 and +80°C. Addition of tuff particles had no significant influence on the tensile strength of composites. However, for all the tested materials the tensile strength was higher at the lower temperature of -24°C but at the higher temperature +80°C was the lowest. Addition of tuff

Table 1. Symbol of composite and mechanical properties of the tested materials

| Sample | Composition [wt%] | $\sigma_M$ [MPa] | $E_t$ [MPa] | $\varepsilon_B$ [%] |
|---|---|---|---|---|
| PS | Recycled polystyrene waste | 40.1±0.87 | 3460±49 | 1.5±0.06 |
| PS/10T | Polystyrene +10% tuff particles | 37.3±1.80 | 3960±205 | 1.6±0.05 |
| PS/20T | Polystyrene +20% tuff particles | 38.2±0.16 | 4400±199 | 1.4±0.25 |

particles increased the modulus of elasticity for composites proportionally to the increasing of tuff content at all temperatures and the higher is at -24°C. The highest over double increase in deformation compared to PS was observed at + 80°C, which is a favourable phenomenon and relatively rarely observed. Probably the developed surface of the hard filler microparticles increases the friction forces between them and the polymer matrix during the tensile test.

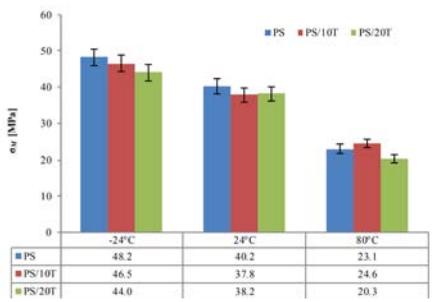


Fig. 1. Tensile strength for PS, PS/10T, PS/20T at room temperature (24°C), -24 and +80°C

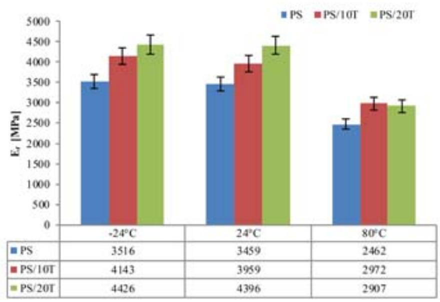


Fig. 2. Modulus of elasticity for PS, PS/10T, PS/20T at room temperature (24°C), -24 and +80°C

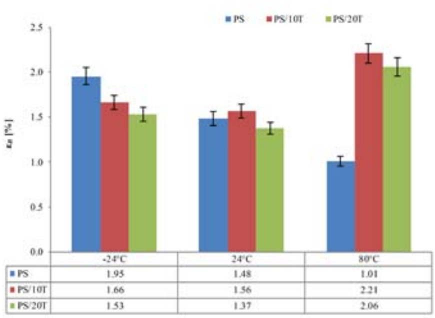


Fig. 3. Strain at break for PS, PS/10T, PS/20T at room temperature (24°C), -24 and +80°C

Figures 4 and 5 compare the static bending test results obtained with PS, PS/10T and PS/20T strength tests at room temperature (24°C), -24, +80°C. The value of the elastic modulus increased in proportion to the increase in tuff content in the composite over the entire range of temperatures tested. Bending strength increased especially at elevated temperatures +80°C, and slightly decreased at low temperatures.

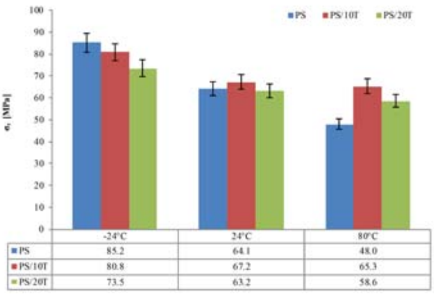


Fig. 4. Flexural stress at 3.5% strain for PS, PS/10T, PS/20T at room temperature (24°C), -24 and +80°C

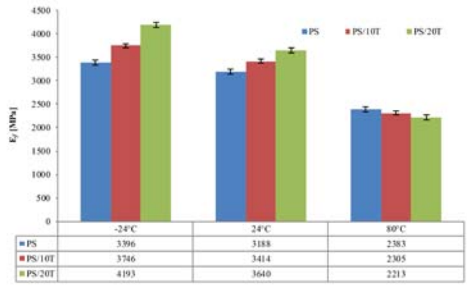


Fig. 5. Flexural modulus for PS, PS/10T, PS/20T at room temperature (24°C), -24 and +80°C

Figure 6 shows the results obtained in the impact test for polystyrene and its composites with 10 wt % and 20 wt % tuff particles. At room temperature, the addition of tuff particles did not significantly change the impact test result. A significant threefold increase in impact strength for samples PS/10T at an elevated temperature of + 80°C was observed. This demonstrates the improvement of homogeneity of polystyrene recyclate and improvement in the impact strength, which increases the possibility of its various applications for technical products. At lower temperatures, there was a 30% decrease in impact strength for composites, and its values for PS/10T and PS/20T composites were similar.

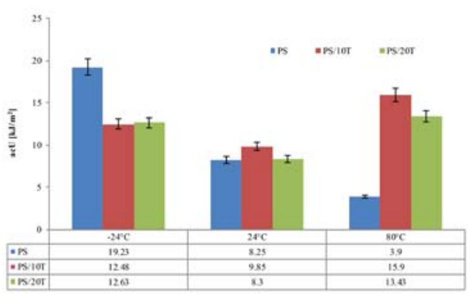


Fig. 6. Charpy impact strength for PS, PS/10T, PS/20T at room temperature (24°C), -24 and +80°C

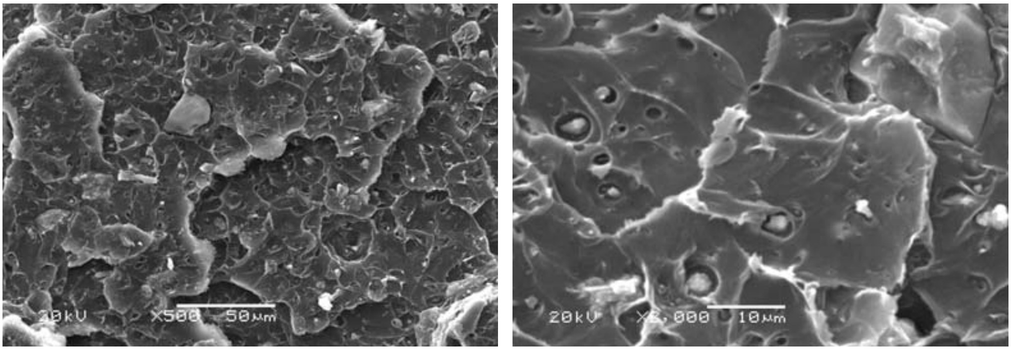

Fig. 7. SEM images of PS/20T tensile test fracture surfaces

Figure 7 show SEM images acquired on tensile fracture specimens of PS with 20% of tuff microparticles. Good homogenization of tuff particles uniformly deposited on the surfaces of the crystalline polystyrene can be observed. The surface of a brittle fracture is related to the supermolecular structure of polystyrene and the method of its crystallization and microparticles are loosely embedded in the polymer matrix.

## 4. Conclusion

The volcanic tuff microparticles proved to be an interesting modifier for recycled polystyrene waste leading to a significant 30% increase in the modulus of elasticity, maintaining the level of strain at break and only a slight decrease of a few percent in tensile strength.

The unchanging value of deformation at break is an extremely beneficial and rarely observed phenomenon; it is caused by the developed surface of tuff microparticles and confirms the compatibility of their effect on polymeric mixtures [15]. The consequence of these phenomena is a favourable increase in impact strength, especially at elevated temperatures.

The results of the study indicate that it is possible to produce composites based on recycled polystyrene waste matrix modified by rocks of volcanic porous origin with increased stiffness and impact resistance. This is of particular importance for the management of poor quality polystyrene waste from various sources, which is difficult to manage because of the significant deterioration in properties due to fluctuations in properties due to aging and compositional homogeneity. Tuff microparticles act as a physical compiler, which promotes miscibility by resolving micro-regions of polymers with different properties, which increases the deformability of the composites produced and their impact resistance. Various applications of polystyrene waste of poor quality for technical items and products with low aesthetic requirements can be indicated, which, thanks to the addition of a new mineral compatibilizer, will have better endurance properties.

Figure 8 shows one example of the possible disposal of recycled polystyrene waste extruded profiles with the addition of wood fibres. These profiles are more durable and less flammable than wood. It works well under wet conditions and at low temperatures. It has a high level of UV resistance and is resistant to fading. The addition of tuff to such a composition will improve its miscibility with wood fibre, and increase its stiffness and impact resistance.

Fig. 8. Example of fence made of waste polystyrene [16]

Summarizing, poor quality recycled polystyrene waste with the addition of a cheap mineral compatibilizer, tuff of volcanic origin, can become a sought after waste that can be used in combination with fibres or alone for long life products. This increases the possibilities of recyclable material of such waste and enables better use of natural resources.

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