

## Characterization of virgin Poly(butylene Terephthalate) and recycled Poly(butylene Terephthalate) from toothbrush bristles

Nga Thi-Hong Pham<sup>1</sup>

<sup>1</sup>(Mechanical Engineering Faculty, HCMC University of Technology and Education, 1 Vo Van Ngan st., Thu Duc district, HCM City, 71307, Vietnam)

---

### **Abstract:**

**Background:** Recycling engineering plastics has been a hot topic that has received a lot of attention. Poly (butylene terephthalate) (PBT) is a brittle material that is generally not suitable for certain applications, while the source of toothbrush bristles recycling is enormous. Research on recycled materials not only has a positive impact on the environment but also can reduce material costs by using recycled materials.

**Materials and Methods:** In this research, blends of recycling PBT, which comes from PBT toothbrush bristles, and origin PBT (Pocan B4225 includes 20% glass fiber) were researched. Tensile strength, flexural strength, Charpy impact testing are studied according to ASTM D638, ASTM D790, and ASTM D256 standard. The samples then were measured through SHORE hardness measurement methods according to ASTM D2240 and microstructure is observed by using a high-resolution FESEM microscope.

**Results:** The virgin PBT and R-PBT are very brittle, the fracture mechanism of these samples is also brittle fracture. The average tensile strength of R-PBT samples was significantly higher than the PBT samples. However, the tensile elastic, flexural elastic, and impact strength of PBT are higher than that of R-PBT sample. About microstructure, virgin PBT sample and R-PBT sample show white particles. No crack initiation area was observed on the fracture surface and no crack propagation was investigated in an unstable way. PBT sample has a smoother organization than R-PBT sample.

**Conclusion:** Recycled PBT blend showed to have competitive mechanical properties compared to virgin PBT materials

**Key Word:** Poly (butylene terephthalate), Recycling Poly(butylene Terephthalate), Microstructure, Tensile strength, Flexural strength.

---

Date of Submission: 22-03-2021

Date of Acceptance: 06-04-2021

---

### I. Introduction

In recent decades, plastic recycling has been a hot topic that has received a lot of attention, especially recycling engineering plastics [1-3]. The environmental troubles caused by polymer waste have prompted actions to reuse and recycle those materials [4-6]. Some methods are applied in polymer recycling technology such as depolymerization, burning polymers for energy recovery, mechanical recycling... In which mechanical recycling has more advantages. However, the degradation of the material is one of the major problems with this polymer recycling method [7-9]. Author Miguel Jorge Reig [10] researches recycled ABS/recycled PC blend. The results showed that the inclusion of the PC in the blend caused the viscosity difference, which was greater as the higher the PC content. Viscosity correction with proposed correction is valid when compared with tested blends of recycled materials. Paul Miller [11] elaborated recycling HDPE with 8 different virgin polyolefins. The results showed that long-chain mixes, such as film blowing grade materials, showed significant variation in viscosity ratio compared to long-chain blends, such as injection molding grade materials. Li Sheng [12] analyzed PE special material for pipe, recycled PE, PE/recycled PE blends, and pipe products, found an easy strategy to define PE/recycled PE blends by assessing Fe and Ca content.

In addition, many scientists also add compatibilizers to the recycling mix. Gregor Radonjic [13] used EPR as a compatibilizer to blend recycled LDPE/PP 80/20 and recycled HDPE/PP 80/20 blends. With the addition of EPR, the notched impact strength and elongation at yield of the recycled LDPE/PP 80/20 blend were significantly improved and Young's modulus and elongation at yield improved slightly. Notched impact strength of recycling HDPE/PP 80/20 blend has been improved to some extent. Xiaodong Liu [14] studied four different plastics from scrapped Volvo cars. They are ABS and ABS/PC, PMMA and PA. Blending recycled ABS and PC/ABS (70/30) with a small amount of methyl methacrylate-butadiene-styrene core-shell impact modifiers has better properties than any of their individual ingredients. Approximately 10% PMMA from the rear light cover can conform to PC/ABS blends made. Property profile will be improved.

Studies of recycled polymers show that the chemical structure, melting temperature, and thermal properties of the whole primary mix change negligibly when recycled. In the study of Emel Kuram [15], PC/ABS and PA6/PC/ABS blends were recycled five times. The result showed that increasing the number of recycling processes slightly changes the flexural and tensile strength of the PC/ABS blend, but decreases MFI and impact strength. On the other hand, the modulus of elasticity and tensile strength of PA6/PC/ABS decreases with the number of recycling processes, while the MFI, flexural properties, and impact strength of the PA6/PC/ABS blend are improved.

Other researches have shown that recycled materials are also suitable for use as insulation in the construction industry or can be developed from a laboratory to an industrial scale. Author Khadija M. Zadeh [16] researched recycled LDPE, recycled HDPE, recycled PP with date palm leaf fiber (DPLF) and 1phr Polyethylene grafted maleic anhydride. It was found that the green synthetic material based on recycled polyolefin/DPLF is fabricated with low thermal conductivity, good strength, and high thermal stability. Such properties make it suitable and attractive for use as insulation materials in the construction industry. Áurea A. Matias [17] studied PP/PET 70/30 wt% with the aim of replacing virgin resins. This ratio developed with recycled PET is a prime example of the applicability of laboratory-developed work to an industrial scale. Sylvie Bertin [18] studied the characterization of virgin and recycled LDPE/PP blends. The result showed that the regenerated material has tensile properties similar to that of the virgin material. In LDPE/PP blend, PP acted as the reinforcement in the matrix and gave the mixture more rigidity.

Poly (butylene terephthalate) (PBT) is a brittle material that is generally not suitable for certain applications, especially for applications requiring high impact strength. Less and less recycled PBT is used, while the source of recycling is enormous, eg toothbrush bristles. Studies on recycled PBT are extremely rare. Elisabete [19] studied aging of PC/PBT blend. The results showed that when studying the recycling potential of PC/PBT blends, elongation is the most important characteristic. The elongation of the recycled PC/PBT is very good, Elongation at break which decreases with each aging step and rises again with each reprocessing step, recommends that the principal factor in the aging process is surface degradation. After re-treatment, polymer chains degraded in non-degraded material appear diluted. The tensile modulus and tensile strength of the recycled PC/PBT are not significantly affected by the processes in which the blend are conformed. The impact strength of the recycled PC/PBT has decreased after aging, which indicates a recovery of a few percent upon reprocessing. Melting flow index enhances during reprocessing, but does not affect molding process of recycled PC/PBT. The addition of pigment has no essential effect on the properties and appreciates the emergence of the recycled PC/PBT blend. Emel Kuram [20] researched on the recycling possibilities of PBT/PC/ABS (acrylonitrile-butadiene-styrene) ternary blend with glassfiber by using were recycled at five times injection molding process. FTIR results confirmed that the chemical nature of the PBT/PC/ABS blends did not alter upon repeated recycling, as evidenced by the fact that all TGA curves of the virgin and recycled blend were close to each other. The mechanical properties such as tensile tension, elastic modulus, and high impact strength of recycled PBT/PC/ABS are better than virgin PBT/PC/ABS blends. In contrast, recycled PBT/PC/ABS blends give lower tensile strength, yield strength, flexural strength, and flexural modulus than neat blends; however, these mechanical properties of PBT/PC/ABS blend are less affected by the recycling process. For melt flow index, PBT/PC/ABS blends showed different behaviors with an increasing amount of recycling. When adding GF, the recycled PBT/PC/ABS-GF blend has strain at break, impact strength, and MFI value higher than that of the primary material. The fifth recycled PBT/PC/ABS-GF blend can be used instead of the neat PBT/PC/ABS blend. However, recycled PBT/PC/ABS-GF material has lower tensile strength, yield strength, elastic modulus, flexural strength and flexural modulus than that of the PBT/PC/ABS-GF blend.

From the above results, it can be seen that recycled material has competitive mechanical properties compared to virgin polymer. Even the chemical nature of the mixtures does not change when repeatedly recycled. Thus, research on recycled materials not only has a positive impact on the environment but also can reduce material costs by using recycled materials. In this research, blends of recycling PBT, which comes from PBT toothbrush bristles and origin PBT were researched. The microstructure and mechanical properties of R-PBT and origin PBT blends have been investigated.

## II. Material And Methods



Recycled PBT from toothbrush bristles scraps → Injection molding → Molded specimens

**Figure 1.** Schematic illustration of the experimental procedure.

The experiment chose to use recycling PBT (R-PBT) which is taken from the toothbrush bristles scraps (Figure 1). PBT resins is Pocan B4225(includes 20% glass fiber) from Saudi Arabia.

**Tensile strength:** Use the TKC series vertical injection molding machine to inject tensile test samples, according to ASTM D638. The tensile test specimen dimension is 19 x 113 x 4 mm, was performed on Shimadzu Autograph AG-X Plus 20 kN (Japan). The constant tensile speed used is 50 mm/min, the maximum test power is 20 kN.

**Flexural strength:** The flexural strength specimen dimensions are 125 × 12 × 3.2 mm, according to ASTM D790. The flexural test device is an Instron 5566 Materials Testing Machine that has a load capacity of 10 kN (2.250 lbf).

**Charpy Impact Testing:** The impact test samples are tested according to ASTM D256 standard. Test performed on Tinius Olsen IT504 impact tester.

**Hardness:** The hardness of the sample was measured through SHORE hardness measurement methods according to ASTM D2240 method, using the tester SHORE D Durometer DESIK.

**Microstructure:** Microstructure is observed by using a high-resolution FESEM microscope HITACHI S-4800. The basic parameters: the resolution of secondary electronic image 1.0nm, 1.4 nm, accelerated voltage reducer.

### III. Result

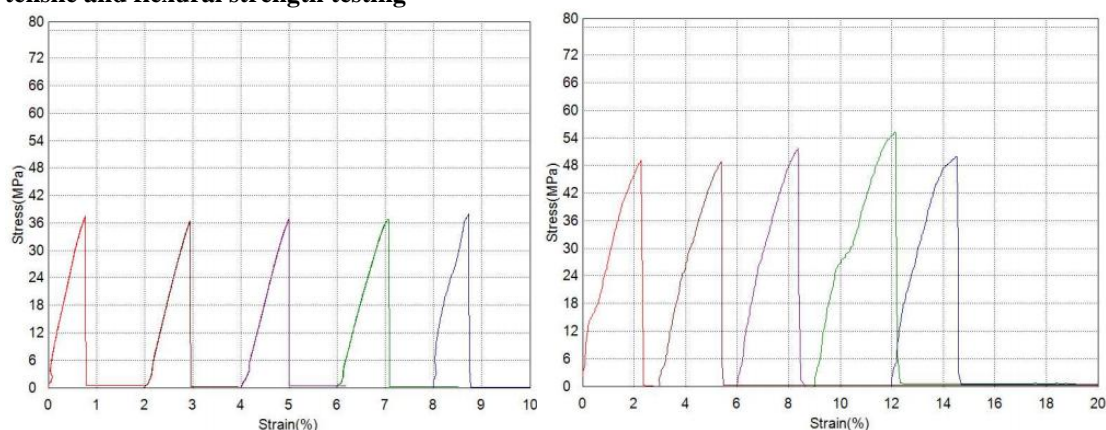
#### Surface morphology



Figure 2. Surface morphology of R-PBT sample.

Figure 2 shows the specimen after injection molding. Through the practice, it can be seen that it is very difficult to inject sample R-PBT, the mold constantly stuck. The blue color of the samples is the inherent blue color of the recycled brush bristles. According to Emel Kuram [20], the addition of colorants does not significantly affect the mechanical properties and improves the appearance of the recycled material.

#### The tensile and flexural strength testing



a) PBT sample b) R-PBT sample

Figure 3. The stress-strain curves of PBT and R-PBT samples

The stress-strain curves of the samples are presented in Figure 3. The pure PBT sample and R-PBT sample are very brittle with a very low value of elongation of about 2.5%. Generally, the fracture mechanism of these samples is also brittle fracture. It is worth noting that the average tensile strength of R-PBT samples (51.73 MPa) were significantly higher than the PBT samples (37.54 MPa). Table no1 demonstrates the tensile elastic of

PBT and R-PBT samples. It can be seen that the average tensile elastic of PBT is 4410.68 MPa, higher than that of R-PBT (2680.45 MPa). The flexural elastic of PBT and R-PBT samples are indicated in Table no2. Similar to tensile elastic, flexural elastic of PBT is also higher than R-PBT samples, at 3725.90 MPa and 2437.77 MPa for PBT and R-PBT samples, respectively.

**Table no1:** Tensile elastic of PBT and R-PBT samples.

Name Parameters	Tensile elastic of PBTsample Stress 12 - 20 MPa	Tensile elastic of R-PBTsample Stress 12 - 20 MPa
Unit	MPa	MPa
1	4673.31	2478.38
2	4373.73	2541.26
3	4180.94	2960.29
4	3883.10	2467.64
5	4942.30	2954.70
Average	4410.68	2680.45
Standard Deviation	413.546	254.469

**Table no2:** Flexural elastic of PBT and R-PBT samples.

Name Parameters	Flexural elastic of PBTsample Stress 10 - 50 N/mm <sup>2</sup>	Flexural elastic of R-PBTsample Stress 10 - 50 N/mm <sup>2</sup>
Unit	N/mm <sup>2</sup>	N/mm <sup>2</sup>
1	3715.65	2312.88
2	3749.10	2479.56
3	3746.31	2397.41
4	3753.47	2542.60
5	3664.99	2456.42
Average	3725.90	2437.77
Standard Deviation	37.1820	87.0489

### Charpy Impact Testing

The average impact strength of test samples was specified in Table no 3. The impact strength of PBT sample is 3.70kJ/m<sup>2</sup> higher than 2.67kJ/m<sup>2</sup> of the R-PBT sample. In Mengyao Shang's research, pure PBT exhibited a character of brittle fracture with the impact strength of only 2.2 kJ/m<sup>2</sup> [22]. Low impact strength is an inherent property of PBT, therefore, there are many studies aimed at improving this property of PBT, eg adding compatibilized, plasticizer, Bio-Content...

**Table no 3:** The average impact strength of test samples.

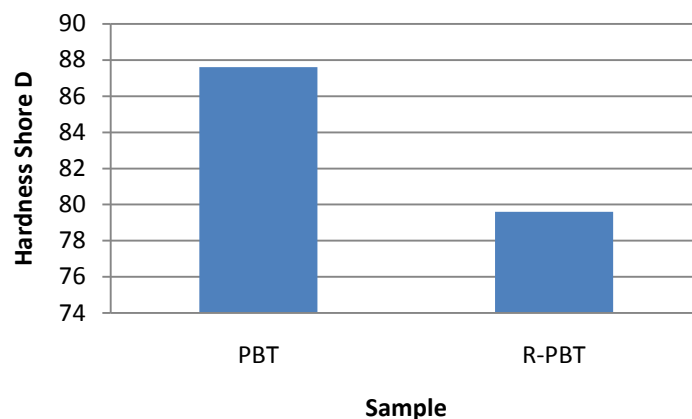
Name	Impact strength of PBT sample			Impact strength of R-PBTsample		
	J	kJ/m <sup>2</sup>	J/m	J	kJ/m <sup>2</sup>	J/m
1	0.1230	3.69	37.29	0.1730	2.55	26.22
2	0.1214	3.64	36.79	0.2210	3.35	34.53
3	0.1259	3.78	38.17	0.1983	3.03	31.48
4	0.121	3.63	36.67	0.1695	2.52	26.70
5	0.1247	3.74	37.78	0.1721	2.57	26.49
Average	0.12	3.70	37.34	0.18	2.67	27.30
Standard Deviation	0.00	0.06	0.60	0.03	0.47	4.74

### Hardness

The average hardness of test samples was shown in Figure 4. Hardness Shore D of R-PBT sample is 80, while R-PBT sample has 85.36 hardness Shore D.

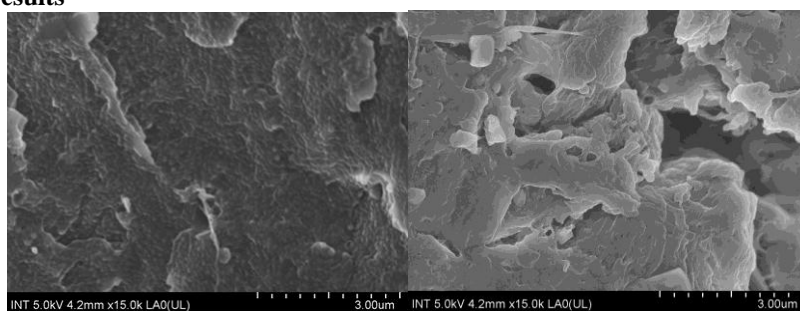
**Table no 4.** Hardness of test samples (Shore D)

Name	Hardness of PBT sample	Hardness of R-PBT sample
Unit	Shore D	Shore D
1	88	82
2	89	80
3	89	79
4	86	77
5	86	78
Average	87.6	79.6



**Figure 4.** The average hardness of test samples

#### Microstructure results



a) PBT sample      b) R-PBT sample

**Figure 5.** SEM micrographs of PBT and R-PBT samples.

Figure 5 presents the SEM micrographs of PBT and R-PBT samples. From the figure, it can be recognized that the origin PBT and R-PBT samples show white particles corresponding to crystals of  $Sb_2O_3$ [23]. The fracture surface of PBT and R-PBT specimens exhibited a typical brittle fracture. No crack initiation area was observed on the fracture surface and no crack propagation was investigated in an unstable way. It should be emphasized that the PBT sample has a smoother organization than the R-PBT sample.

#### IV. Discussion

From the above results, it can be seen that the tensile strength of the R-PBT sample is even higher than that of virgin PBT. The results of this study are similar to previous studies on recycled materials. According to Jan Rybnicek [21], recycled blends showed to have competitive mechanical properties compared to virgin materials. Recycling has only a small effect on the tensile properties and dynamic fracture strength. Yield stress and strain are virtually unchanged when compared to mixtures made from virgin materials. Recycling reduces tensile strain at break, but this is not important to safe engineering design. The toughness is highly dependent on the processing temperature. It may be suggested that the thermomechanical load of the molten material is kept as low as possible.

In addition, Gregor Radonjic [13] emphasizes that recycled thermoplastics often contain additives, grades, and degradation products that affect interoperability. Similar results were found in Xiaodong Liu' work [14], antioxidants and metal deactivators do not help recyclers exhibit better mechanical properties.

#### V. Conclusion

- The virgin PBT and R-PBT are very brittle, the fracture mechanism of these samples is also brittle fracture. The average tensile strength of R-PBT samples is significantly higher than the PBT samples. However, the average tensile elastic and flexural elastic of PBT are higher than that of R-PBT sample.
- The impact strength of PBT sample is  $3.70\text{kJ/m}^2$  higher than  $2.67\text{kJ/m}^2$  of the R-PBT sample.

- Virgin PBT and R-PBT sample show white particles no crack initiation area was observed on the fracture surface and no crack propagation was investigated in an unstable way. PBT sample has a smoother organization than the R-PBT sample.

### References

- [1]. Nouh S. A., Abou Elfadl A., Alhazime Ali A., Al-Harbi Abdulaziz M. Effect of proton irradiation on the physical properties of PC/PBT blends. *Radiation Effects and Defects in Solids*, 2018
- [2]. Nga Thi-Hong Pham, Van-Thuc Nguyen. Morphological and mechanical properties of Poly(butylene terephthalate)/ High-density polyethylene blends. *Advances in Materials Science and Engineering*. Vol. 2020, Article ID 8890551, 2020, <https://doi.org/10.1155/2020/8890551>
- [3]. Dechet Maximilian, Gómez Bonilla Juan, Lanzl Lydia, Drummer, Dietmar, Bück, Andreas, Schmidt, Jochen, Peukert, Wolfgang. Spherical Polybutylene Terephthalate (PBT)-Polycarbonate (PC) Blend Particles by Mechanical Alloying and Thermal Rounding. *Polymers*, 2018, Vol. 10, Iss. 12
- [4]. Wang Baolong, Wu Di, Zhu Lien, Jin Zheng, Zhao Kai. High-Density Polyethylene-Based Ternary Blends Toughened by PA6/PBT Core-Shell Particles. *Polymer-Plastics Technology and Engineering*, 2017
- [5]. Deng Bin, Lv Hanxiao, Song Shixin, Sun Shulin, Zhang Huixuan. Modification of the reactive core-shell particles properties to prepare PBT/PC blends with higher toughness and stiffness. *Journal of Polymer Research*, 2017, Vol. 24, Iss. 6
- [6]. Tang Lei, Wang Lulin, Chen Pingxu, Fu Jinfeng, Xiao Peng, Ye Nanbiao, Zhang Mingqiu. Toughness of ABS/PBT blends: The relationship between composition, morphology, and fracture behavior. *Journal of Applied Polymer Science*, 2017
- [7]. Ignaczak W., Sui X. M., Kellersztein I., Wagner H.D., El Fray M.. The effect of fibre sizing and compatibiliser of PP/PBT blends on the mechanical and interphase properties of basalt fibre reinforced composites. *Polymer International*, 2017
- [8]. Hong-Nga Thi Pham, Van-Thuc Nguyen. Effect of calcium carbonate on the mechanical properties of polyethylene terephthalate/polypropylene blends with styrene-ethylene/butylene-styrene. *Journal of Mechanical Science and Technology*, 2020, Vol. 34 Iss. 10, pp. 3925-3930.
- [9]. Hajibaba Armin, Masoomi Mahmood, Nazockdast Hossein. Compatibilization effectiveness of maleated polypropylene compared to organoclay in PBT/PP blends. *Iranian Polymer Journal*, 2016, Vol. 25, Iss. 2
- [10]. Miguel Jorge Reig, Vicente Jesus Segui, Jose Domingo Zamanillo. Rheological Behavior Modeling of Recycled ABS/PC Blends Applied to Injection Molding Process. *Journal of Polymer Engineering*, 2005, Vol. 25, Iss. 5, pp. 435-457
- [11]. Paul Miller, Igor Sbarski, Edward Kosior, Syed Masood, Pio Iovenitti. Correlation of Rheological and Mechanical Properties for Blends of Recycled HDPE and Virgin Polyolefins. *Journal of Applied Polymer Science*, 2001, Vol. 82, Iss. 14, pp. 3505-3512
- [12]. Li Sheng , Xiaoying Lu, Liang Chi , Chunyan Xu and Shuanhong Li. The Effects of the Recycled Polyethylene on the Properties of Special Material Blends for Pipe and Final Products. 2020 4th International Conference on Material Science and Technology, IOP Conf. Series: Materials Science and Engineering, 2020, Vol. 774, 012052
- [13]. Gregor Radonjic, Nenad Gubelj. The Use of Ethylene/Propylene Copolymers as Compatibilizers for Recycled Polyolefin Blends. *Macromolecular Materials and Engineering*, 2002, Vol. 287, Iss. 2, pp. 122-132
- [14]. Xiaodong Liu, Hans Bertilsson. Recycling of ABS and ABS/PC Blends. *Journal of Applied Polymer Science*, 1999, Vol. 74, pp. 510-515
- [15]. Emel Kuram, Babur Ozcelik, Faruk Yilmaz. The effects of recycling process on thermal, chemical, rheological, and mechanical properties of PC/ABS binary and PA6/PC/ABS ternary blends. *Journal of Elastomers and Plastics*, 2015
- [16]. Khadija M. Zadeh , I. M. Inuwa, Reza Arjmandi, Azman Hassan, M. Almaadeed, Zurina Mohamad, P. Noorunnisa Khanam. Effects of Date Palm Leaf Fiber on the Thermal and Tensile Properties of Recycled Ternary Polyolefin Blend Composites. *Fibers and Polymers*, 2017, Vol.18, No.7, pp. 1330-1335
- [17]. Áurea A. Matias, Mafalda S. Lima, João Pereira, Paula Pereira, Rodrigo Barros, Jorge F.J. Coelho, Arménio C. Serra. Use of recycled polypropylene/poly(ethylene terephthalate) blends to manufacture water pipes: An industrial scale study. 2020, Vol. 101, No.1, pp. 250-258
- [18]. Sylvie Bertin, Jean-Jacques Robin. Study and characterization of virgin and recycled LDPE/PP blends. *European Polymer Journal*, 2002, Vol. 38, pp. 2255-2264
- [19]. Elisabete Maria Saraiva Sanchez. Ageing of PC/PBT blend: Mechanical properties and recycling possibility. *Polymer Testing*, 2007, Vol. 26, pp. 378-387
- [20]. Emel Kuram, Babur Ozcelik, Faruk Yilmaz, Gokhan Timur, Zeynep Munteha Sahin. The Effect of Recycling Number on the Mechanical, Chemical, Thermal, and Rheological Properties of PBT/ PC/ABS Ternary Blends: With and Without Glass-Fiber. *Polymer Composites*, 2014, pp. 2074-2084, DOI 10.1002/pc
- [21]. Jan Rybníček, Ralf Lach, Monika Lapcikova, Josef Steidl, Zdenek Krulis, Wolfgang Grellmann, Miroslav Slouf. Increasing Recyclability of PC, ABS and PMMA: Morphology and Fracture Behavior of Binary and Ternary Blends 3. *Journal of Applied Polymer Science*, 2008, Vol. 109, pp. 3210-3223
- [22]. Mengyao Shang, Henglun Cheng, Baoqing Shentu, Zhixue Weng. Preparation of PBT/POE-g-GMA/PP ternary blends with good rigidity-toughness balance by core-shell particles. *Journal applied polymer science*. 2019, DOI: 10.1002/APP.48872
- [23]. Mauricio Vásquez-Rendón, Natalia Sánchez-Arrieta, Mónica Álvarez-Láinez. Two-step processing method for blending highperformance polymers with notable thermal and rheological differences: PEI and PBT. *Polymer-Plastics Technology and Engineering*, 2017. <http://dx.doi.org/10.1080/03602559.2017.1381256>