

INFLUENCE OF RECYCLED PA6 AND PBT PLASTIC MATERIAL ON THE BONDING STRENGTH

GABRIEL DITTRICH¹, LUBOSLAV STRAKA¹, FRANTISEK BOTKO¹, JAN DUPLAK¹, JAROSLAV BIRCAK¹, FAHAD ALDAIHAN²

¹Technical University of Kosice, Faculty of Manufacturing Technologies with a seat in Presov, Slovak Republic

²Korea Maritime and Ocean University, Faculty of Mechanical Engineering, 727 Taejong-ro, Yeongdo-Gu, Busan 49112, South Korea

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e-mail: gabriel.dittrich@tuke.sk

Nowadays, due to the requirement to achieve specific product properties or to reduce the weight of machine parts, conventional metal materials are replaced by plastics. Various methods are used to connect them to assemblies. The task of the joint is to provide a perfect, but particularly reliable, compact unit. One of the relatively effective methods of bonding plastic parts is their bonding. In addition to the properties of the adhesive used, the quality and reliability of the bonded plastic components also depend on the properties of the plastic materials used. The aim of the experimental research was therefore to contribute to the knowledge in the field of strength properties of bonded joints using PA6 and PBT plastic materials with different amount (wt%) of recycled polymer.

KEYWORDS

bonded joints, recycled plastic, polyamide 6 (PA6), polybutylene terephthalate (PBT), shear strength, testing.

1 INTRODUCTION

Modern machines and equipment require more and more components to ensure their operability, safety and reliability. However, each of these components is involved in increasing their weight. To avoid an enormous increase in weight, machine and equipment manufacturers are mainly focused on light non-ferrous alloys and plastics. At present, this trend is particularly apparent in the automotive industry. In automotive production, one of the requirements placed on the vehicle is its low total weight. Parts of machinery and equipment made of steel, where functionality and stability allow, are gradually replaced by materials of several times lower specific gravity. These materials also include plastics. However, the replacement of parts made of steel with parts made of plastic also implies the need to maintain the required strength and stability. This can be achieved relatively efficiently by the use of components made of composite materials bonded by means of a suitable fastener. Moreover, the integration of additives into the adhesive bonding layer can have a positive impact on the distribution of the forces in the bonded layer, thereby increasing its adhesion. At the same time, thanks to a number of advantages that the bonding brings with it in the form of high wear resistance and high strength to weight ratio, this method of interconnecting plastic parts is currently quite often used.

2 BASIC CHARACTERISTICS OF BONDED JOINTS

The quality and reliability of bonded joint depends on many factors. In particular, these are parameters related to its static and dynamic resistance to external forces [Knezo 2016, Michalik 2016]. These basic parameters need to be taken into account at the design stage of the bonded joint. In special cases, it is also advisable to carry out an analytical study of shear stress and bond strength distribution [Reza 2014, Campilho 2015]. New possibilities in the area of bonded joints opened the development of synthetic adhesives, epoxy, polyvinyl acetate and various types of modified synthetic adhesives as well as rubber-based synthetic adhesives based on special synthetic elastomers. This issue was described in detail by the authors in the publications [Jagatap 2018, Morfini 2018]. An important advantage of the bonding technology is that it does not interfere with the structure and integrity of the bonded components, such as in riveting or welding. At the same time, this method of joining components retains their original appearance. The application of bonding technology brings not only a number of technological advantages, but also undoubtedly brings a favorable economic effect [Quinia 2012]. In general, the vast majority of plastics can be bonded [Miller 2014]. An important condition for their bonding is a satisfactory chemical composition which ensures sufficient adhesion to the adhesives [Barbosa 2018]. Although plastic bonding is basically a simple polymer bonding with polymers, this process is much more complicated in practice [Panda 2017]. For example, unlike metals, plastics contain a number of additives [Knapčíková 2018], such as colorants, plasticizers, stabilizers, etc., which in many cases prevent their perfect adhesion [Chumble 2017]. In addition, the strength of the adhesive bond is affected by the adhesion of the adhesive used and the surfaces to be bonded [Clauß 2011]. The size of the adhesion forces is also influenced by electrostatic and chemical effects, therefore, consistent preparation of bonded surfaces is crucial [Beber 2016]. At the same time, the vast majority of machine parts are exposed to voltage [Lehocka 2016], bending, compression and shear forces, or a combination thereof. Since the bonded joint has the highest shear strength [Panda 2018], it must be sensitively selected with regard to the direction of the loading forces of the bonded components [Akhavan-Safar 2017]. However, a suitably designed bonding joint with adequate treatment of bonded surfaces provides a prerequisite for a solid and reliable joint [Na 2016]. Conversely, improperly designed adhesive joints, untreated or poorly treated surfaces often result in incoherence joint [Fernandes 2015]. Cleaning and degreasing of plastic materials can be considered a suitable preparation of bonded surfaces. In some cases, it is also necessary to carry out chemical treatment of the surfaces, for example by etching, or to activate the surfaces by flame or corona discharge. In terms of difficulty making the adhesive bond can be based on the results of several studies concluded that bonding thermoplastics is compared with thermoset bonding much more complicated [Neto 2012]. In practice, thermoplastics are joined exclusively by reactive adhesives [Argoud 2018]. In addition, when bonding two thermoplastics, solvent-free adhesives are exclusively applied [Arash 2014]. In terms of bonding integrity, the thermoplastics can be divided into two main groups [Reis 2011]. The first group consists of medium polarity thermoplastics (PS, PMMA, PVC, ABS, PC), which have good adhesion properties. The second group consists of thermoplastics strongly polar, such as e.g. PA and PET, which can only be bonded after demanding treatment of bonded surfaces.

3 TESTING OF BONDED JOINTS

The purpose of testing bonded joints is to determine their quality parameters [Esmaili 2018, Dobránský 2016]. In general, the quality of the bonded joint can be characterized as a summary of properties [Knapčíková 2017], which provide a suitable precondition for satisfying in general the required or predetermined requirements [Straka 2011]. First, a visual inspection of the bonded joint is performed. Subsequently, due to the practical demands placed on the quality of the adhesive bond is carried out verification of its adhesive properties [Budhe 2017]. In practice, there are several methods that test these joints in particular for strength [Anes 2016]. Strength testing of bonded joints is standardized, with a technical standard describing the test procedure. The basic test is the shear strength test according to STN EN 1465. It is a test in which the bonded joint is stressed by shear stress static tensile. The load of the bonded joint is performed in the direction of the longitudinal axis until the test sample is completely broken. The loading rate of the sample is $10\text{mm}\cdot\text{min}^{-1}$, and the bonding failure should occur within $65\pm 20\text{s}$. The surface of the bonded surfaces of the samples must be treated in accordance with STN EN 13887. On the following Figure 1 shows the basic dimensions of the experimental sample in accordance with STN EN 1465 and the distribution of forces in its shear strength test.

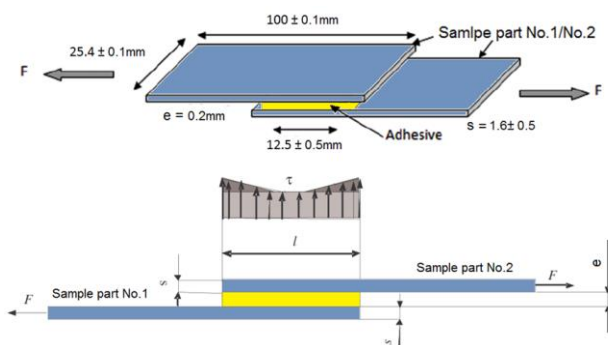


Figure 1: Basic dimensions of the experimental sample in accordance with STN EN 1465 and the force distribution in the shear strength test

In certain specific cases, or in terms of a comprehensive assessment of the bonding quality, in addition to the standardized shear strength parameter, the bonded joint must also be assessed for other parameters. This is, for example, the parameter of brittleness of the bonded joint according to STN EN 66 8511. The peel resistance of the bonded joint at 90° is also assessed (STN EN 28510-1) and 180° (STN EN 1465), or its shear resistance at cyclic load according to ISO 9664:1993. The difference between the tensile strength test according to STN EN 1465 and ISO 9664: 1993 is that the test specimen is subjected to cyclic loading so that the resulting tension is a superposition of static (middle) and pulsating tensile stresses. The test is performed on a device capable of producing a sinusoidal load at a frequency of about 30Hz. It is not recommended to exceed 60Hz to avoid bonded joint breakage due to heating.

Table 1: Basic physical and mechanical properties of samples of materials PA 6 and PBT

Material	Density ($\text{kg}\cdot\text{m}^{-3}$)	Melting Point ($^\circ\text{C}$)	Yield strength (MPa)	Tensile modulus of elasticity (MPa)	Tensile strength (MPa)	Elongation (%)	Thermal conductivity ($\text{Wm}^{-1}\text{K}^{-1}$)	Electrical resistance (Ωcm)
PA 6	1140	220	38	3200	50	35	0.25	$10^8\text{-}10^{14}$
PBT	1330	225	34	2500	61	50	0.27	$10^{13}\text{-}10^{16}$

4 MATERIAL AND METHODS

In the experiment, bonded joints were made by mutual non-detachable connection by combining two plastic materials of the company Xiamen Keyuan Plastic Co, Ltd. labeled PA 6 and PBT by the 2-component adhesive REPLAST® Easy companies Würth. In the case of PA 6 plastic material, it was a black granulate with improved mechanical properties, dimensional stability, heat and aging resistance, whose fatigue resistance is 2.5 times higher than that of conventional polyamide PA6. This material is often used in the automotive industry for the production of rear view mirror housings, lighting controls, door handles, heating and air conditioning components, seat belt tensioners and the like. In the case of PBT (Polybutylene terephthalate) plastic material, it was a pure plastic polymer, which is characterized by good dimensional stability, chemical resistance and high tensile strength. This material is also often used in the automotive industry to produce mechanically stressed body parts such as fenders, bumpers, and the like. Due to their good chemical stability, they are also used to manufacture fuel system parts. The following table 1 shows the basic physical and mechanical properties of the plastic material samples used.

A total of 21 combinations experimental samples with different amount (wt%) (0, 50 and 100) of recycled polymer PA 6 and PBT were produced. Samples were made by Metalis Slovakia with its headquarters in Prešov. On the following Figure 2 shows experimental samples made of plastic material PA 6 and PBT for testing according to STN EN 1465.

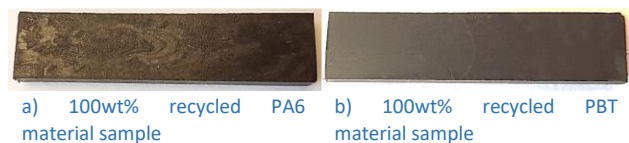


Figure 2: Preparation of experimental samples for testing according to STN EN 1465

Prior to bonding, all contact surfaces of the samples were treated in accordance with STN EN 13887. This was followed by gluing with REPLAST® Easy companies Würth. It is a two-component polyurethane adhesive with a density of $1.225\text{g}\cdot\text{cm}^{-3}$ and a temperature resistance of -30 to 100°C . On the following Figure 3 shows one of the experimental samples, which was made of two (a/Sample part No.1 and b/Sample part No.2) 100wt% recycled PBT plastic materials.



Figure 3: Experimental adhesive bonded sample from two 100wt% recycled PBT plastic materials

Table 2: Combination of PA6 and PBT bonded samples with amount (wt%) of recycled polymer

Material designation	The proportion of recycled material in the first part (a/sample part No.1) bonded samples (wt%)	Material designation					
		PA6			PBT		
		The proportion of recycled material in the second part (b/sample part No.2) bonded samples (wt%)					
		0	50	100	0	50	100
PA6	0	PA6 ₀ /PA6 ₀			PA6 ₀ /PBT ₀	PA6 ₀ /PBT ₅₀	PA6 ₀ /PBT ₁₀₀
	50	PA6 ₅₀ /PA6 ₀	PA6 ₅₀ /PA6 ₅₀		PA6 ₅₀ /PBT ₀	PA6 ₅₀ /PBT ₅₀	PA6 ₅₀ /PBT ₁₀₀
	100	PA6 ₁₀₀ /PA6 ₀	PA6 ₁₀₀ /PA6 ₅₀	PA6 ₁₀₀ /PA6 ₁₀₀	PA6 ₁₀₀ /PBT ₀	PA6 ₁₀₀ /PBT ₅₀	PA6 ₁₀₀ /PBT ₁₀₀
PBT	0				PBT ₀ /PBT ₀		
	50				PBT ₅₀ /PBT ₀	PBT ₅₀ /PBT ₅₀	
	100				PBT ₁₀₀ /PBT ₀	PBT ₁₀₀ /PBT ₅₀	PBT ₁₀₀ /PBT ₁₀₀

After curing of the adhesive, the samples were tested according to STN EN 1465, which specifies the procedure for determining the shear strength at the tensile stress of the overlapped adhesive joints. The strength test of bonded joints of samples was performed in laboratory conditions of the Faculty of Manufacturing Technologies with the seat in Prešov on the equipment of VEB Thüringer Industrierwerk Rauenstein with the designation FM 1000, which is suitable for tensile, compression and bending tests with a maximum load range of 0 to 10kN. During the tensile test, a loading force of 10kN was selected with a clamp speed of 10 mm.min⁻¹.

Samples were prepared within the experiment, which represented individual combinations of bonded joints of plastic materials PA6 and PBT with different amount (wt%) of recycled polymer. The following Table 2 lists the various combinations of bonded samples of material PA6 and PBT.

After curing of the adhesive according to the instructions given by the two-component adhesive manufacturer, the samples were then subjected to a shear strength test according to STN EN 1465. It is a test device that is used for tensile, compression and bending tests with a maximum loading force of 10kN. The following Figure 4 shows the VEB Thüringer Industrierwerk Rauenstein FM 1000, which was used for experimental testing of samples.



Figure 4: The machine VEB Thüringer Industrierwerk Rauenstein FM 1000 used in the shear strength test of bonded joints of experimental samples

Since the test equipment used allows only the force $F(N)$ and the maximum load force $F_{max}(N)$ to be recorded, it was necessary to calculate the stress acting on the bonded joint according to formula (1):

$$\tau_{is} = \frac{F_{i\max}}{S_{is}} \text{ (MPa)} \quad (1)$$

τ_{is} – shear stress at the point of i -th bonded joint (MPa),
 $F_{i\max}$ – maximum force measured when loading i -th sample (N),
 S_{is} – the size of the bonded area (mm²)

In the visual inspection of the broken part of the samples in the area of the bonded joint that occurred after the shear strength

test, signs of a combination of cohesive and adhesive fracture were observed. In some parts, the adhesive has been partially detached from its substrate. On Figure 5 shows the broken PBT₁₀₀/PBT₁₀₀ experimental sample in the bonded area after shear strength test.



Figure 5: Broken sample PBT₁₀₀/PBT₁₀₀ in the bonded area after shear strength test

5 RESULTS AND DISCUSSION

Maximum force values $F_{i\max}$ (N) recorded by the VEB Thüringer Industrierwerk Rauenstein FM 1000 and the empirically determined shear stress values τ_{is} (MPa) at which the i -th experimental sample in the bonded area was bonded by a combination of materials PA6 and PBT with 0 to 100wt% recycled material are shown in the following Table 3.

Table 3: Maximum load force values $F_{i\max}$ and empirically determined shear stress values τ_{is} recorded in the shear strength test of the bonded joint of experimental samples

Parameter	Sample part No.1	Sample part No.2		
		PA6 ₀	PA6 ₅₀	PA6 ₁₀₀
$F_{i\max}$ (kN)	PA6 ₀	2,42		
	PA6 ₅₀	2,24	2,02	
	PA6 ₁₀₀	1,94	1,84	1,72
τ_{is} (MPa)	PA6 ₀	7,30		
	PA6 ₅₀	6,76	6,09	
	PA6 ₁₀₀	5,85	5,55	5,19
		PBT ₀	PBT ₅₀	PBT ₁₀₀
$F_{i\max}$ (kN)	PBT ₀	3,95		
	PBT ₅₀	3,68	3,54	
	PBT ₁₀₀	3,32	3,04	2,90
τ_{is} (MPa)	PBT ₀	11,95		
	PBT ₅₀	11,10	10,68	
	PBT ₁₀₀	10,02	9,17	8,75
		PBT ₀	PBT ₅₀	PBT ₁₀₀
$F_{i\max}$ (kN)	PA6 ₀	3,31	3,11	2,92
	PA6 ₅₀	2,89	2,68	2,44
	PA6 ₁₀₀	2,33	2,19	2,07
τ_{is} (MPa)	PA6 ₀	9,98	9,38	8,81
	PA6 ₅₀	8,72	8,08	7,36
	PA6 ₁₀₀	7,03	6,61	6,24

Dependent on empirically determined maximum shear stress values, in which the i -th experimental sample in the bonded area was breached; created by bonding the combination of materials PA6 and PBT with 0 to 100wt% recycled material, there were constructed graphical dependencies Figure 6 to 8.

From the graphical dependencies shown in Figure 6, a decrease in the empirically determined maximum shear stress values τ_s of the bonded joint can be observed with increasing wt% of the recycled PA6 material. Their values ranged from 5.19 to 7.30 MPa. The highest value of $\tau_s = 7.3$ MPa was achieved in a sample made by bonding from two parts (PA6₀/PA6₀) with 0wt% recycled PA6 material. Conversely, the lowest value of $\tau_s = 5.19$ MPa was achieved in a sample made by bonding from

two parts (PA6₁₀₀/PA6₁₀₀), both of which contained 100wt% recycled PA6 material.

From the graphical dependencies shown in Figure 7, a decrease in the empirically determined maximum shear stress values τ_s of the bonded joint can be observed with an increasing wt% of recycled PBT material. Their values ranged from 8.75 to 11.92 MPa. The highest value of $\tau_s = 11.92$ MPa was achieved in a sample made by bonding two parts (PBT₀/PBT₀) with 0wt% recycled PBT material. Conversely, the lowest $\tau_s = 8.75$ MPa was achieved in a sample made by bonding from two parts (PBT₁₀₀/PBT₁₀₀), both containing 100wt% recycled PBT material.

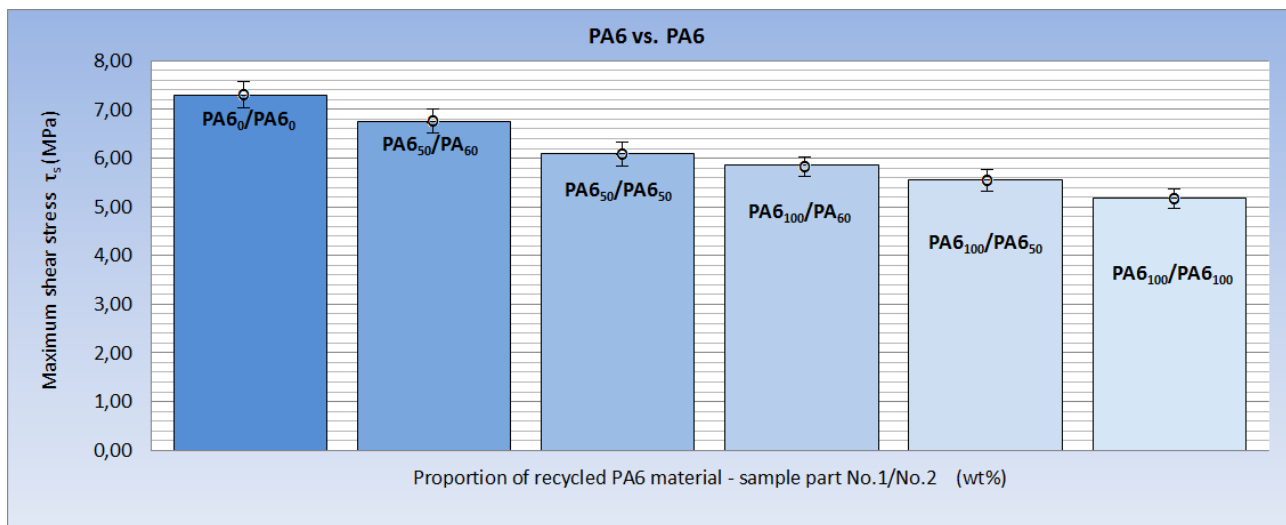


Figure 6: Dependence of maximum shear stress τ_s of bonded joint on wt% of recycled PA6 material of individual sample parts (sample part No.1/No.2)

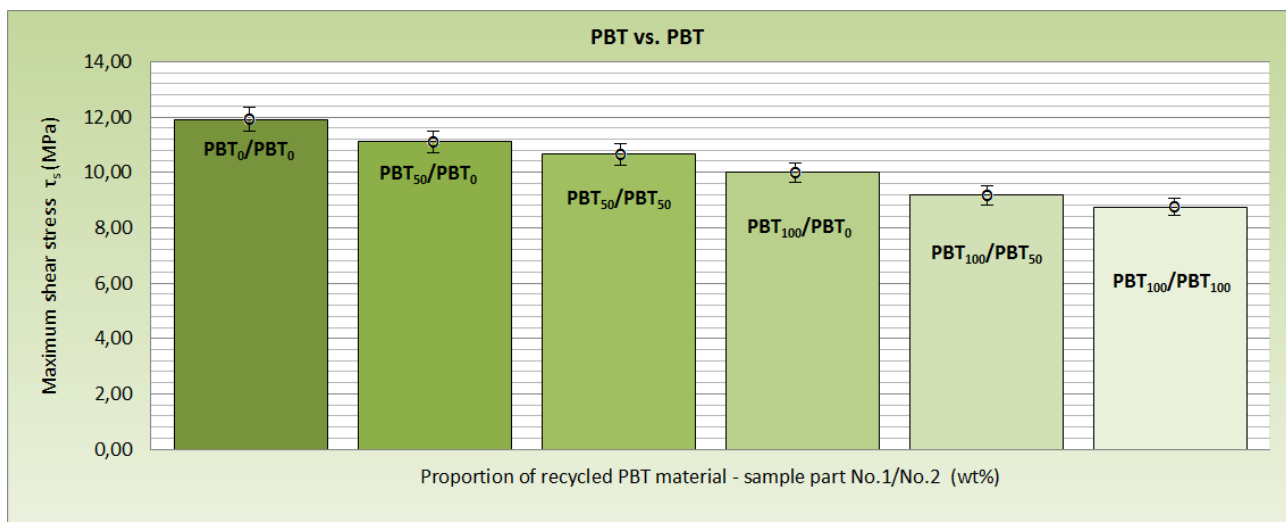


Figure 7: Dependence of maximum shear stress τ_s of bonded joint on wt% of recycled PBT material of individual sample parts (sample part No.1/No.2);

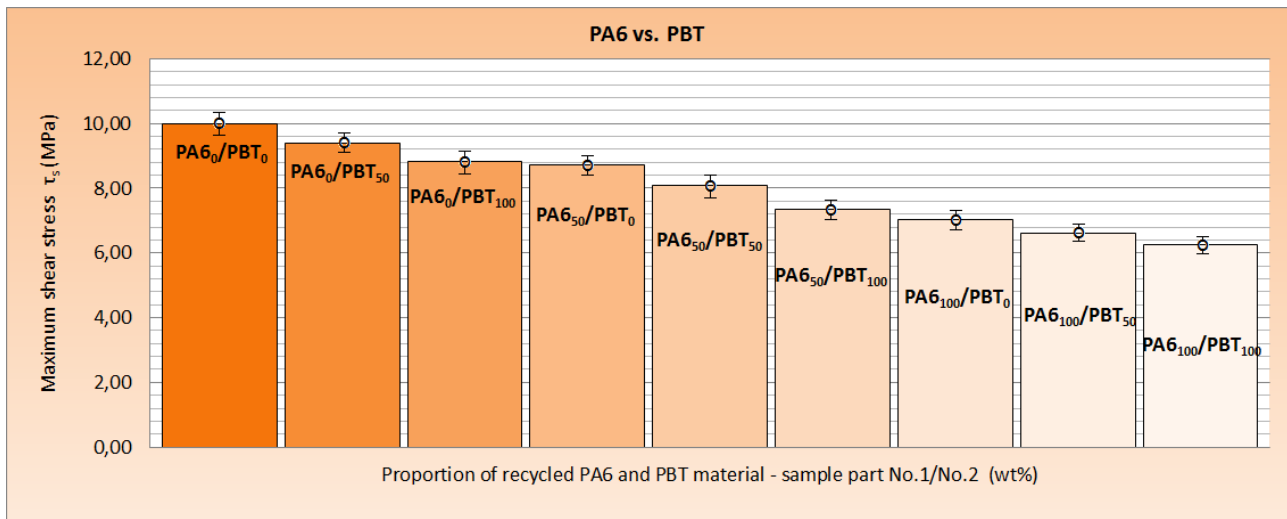


Figure 8: Dependence of the maximum shear stress τ_s of bonded joint on the wt% of recycled PA6 and PBT material of each sample part (sample part No.1/No.2)

From the graphical dependencies shown in Figure 8, which describes the effect of the wt% of recycled PA6 and PBT material of the individual sample parts on the size of the maximum shear stress τ_s of the bonded joint can be observed by these facts. With the increasing wt% of recycled PA6 and PBT material there is a slight decrease in the maximum shear values τ_s of the bonded joint. Their values ranged from 6.24 to 9.98 MPa. The highest value of $\tau_s = 9.98$ MPa was achieved in a sample made by bonding two parts (PA6₀/PBT₀) with 0wt% recycled PA6 material as well as 0wt% recycled PBT material. On the other hand, the lowest value $\tau_s = 6.24$ MPa was achieved in a sample made by bonding two parts (PA6₁₀₀/PBT₁₀₀), with both PA6 and PBT containing 100wt% recycled material. Based on a comprehensive assessment of the graphical dependencies shown in Figure 6 to 8, it can be stated that the highest bond strength in shear $\tau_s = 11.95$ MPa was achieved with a bonded joint (PBT₀/PBT₀) with 0wt% recycled PBT material of both parts. Conversely, the lowest bond strength of shear $\tau_s = 5.19$ MPa was achieved with a bonded joint (PA6₁₀₀/PA6₁₀₀) with 100wt% recycled PA6 material in both parts. It follows that the difference in shear values τ_s of the bonded joint with the 0wt% share of recycled PBT material of both parts compared to the bonded joint with 100wt% recycled PA6 material of both parts is at the level of 6.76 MPa. The reason for the decreasing tensile strength of the bonded joint with the increasing wt% of PA6 or PBT recycled material is assumed in increasing contaminants, which have a negative impact on the adhesive properties of the surface layer of the plastic material. Therefore, further research will be directed towards identifying these contaminants to achieve the same bonding properties using virgin and recycled plastic. At the same time, it is necessary to look for possibilities to increase the strength of the bonded joint made from the combination of PA6 and PBT materials.

6 CONCLUSIONS

At present, plastics have their irreplaceable position in the manufacture of machinery and equipment. At the same time, they become more and more important in modern society. The plastics industry is becoming one of the most developing industries. Advances in this area allow the emergence of new products, systems and technologies. As the demand for these products increases, their quality demands are also increasing. One of the decisive quality parameters of the finished

production is their precise and reliable bonding. On this basis, the primary choice is to choose the technology of joining them. One of the milestones in joining plastic materials was the development of synthetic adhesives. This method of joining components is now commonly used in the automotive and aerospace industries, but also in construction, electrical engineering, etc. Therefore, the aim of the experimental research was to contribute to the knowledge in the field of strength properties of bonded joints using selected types of plastics with different amount (wt%) of recycled polymer. Based on the results of experimental tests carried out in laboratory conditions in accordance with the strength test of bonded joint in accordance with the standard STN EN 1465, a significant difference was found in the properties of bonded joints of PA6 and PBT plastic materials with different amount (wt%) of recycled polymer or the combination of bonded plastic materials. It has been found that with increasing wt% of both recycled PA6 and PBT material, there is some decrease in the maximum shear stress τ_s of the bonded joint. At the same time, it was found that bonded joints, both of which were made of PBT material, had a higher τ_s value of 4.15 MPa compared to a PA6 pair. When applying PA6 and PBT combined bonding with varying wt% of recycled material, somewhat higher τ_s were observed than with PA6 bonding with varying wt% of recycled material. However, these values were slightly lower compared to the τ_s values recorded for PBT bonded joints with different amount (wt%) of recycled polymer. Future research in this field should be focused mainly on the possibilities of increasing the strength of bonded joints of said plastic materials PA6 and PBT.

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CONTACTS

Ing. Gabriel Dittrich; assoc. prof. Ľuboslav Straka, PhD.; Ing. František Botko, PhD.; Ing. Ján Duplák, PhD.; Ing. Jaroslav Birčák; Ing. Fahad Aldaihan

Department of Automotive and Manufacturing Technologies Faculty of Manufacturing Technologies of the Technical University of Košice with a seat in Prešov, Štúrova 31, 080 01 Prešov, Slovakia, tel.:+421 55 602 6365

e-mail: gabriel.dittrich@tuke.sk; luboslav.straka@tuke.sk; frantisek.botko@tuke.sk; jan.duplak@tuke.sk; jaroslva.bircak@tuke.sk