Surface Laser Micropatterning of Polyethylene Terephthalate (PET) to Increase the Shearing Strength of Adhesive Joints

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Abstract. The method for increasing the shearing strength of adhesive joints in plastics was investigated. This method uses laser micropatterning of different construction materials to extend adhesive surfaces. In investigations, Polyethylene Terephthalate (PET) was used. TruMicro 5325c ultra-short pulse (picosecond pulse) laser and the SCANLAB GALVO scanning head applied in the research enable ablative removal of the material without the heat-affected zone (HAZ) in the rest of it. Ultra-short laser pulses (such as picosecond pulses) remove material without melting the rest of it. The presented method significantly increases the shearing strength of the formed joints in investigated materials made from plastics, which was proved in the results of laboratory tests. The laser device parameters used, which are described in this article, have not produced cracks in the microtreated materials. The research shows that bondings between elements with the appropriately machined microstructure are characterized by a severalfold increase in the strength of joints in relation to materials devoid of the microstructure. In addition, this study addresses practical solutions to the adhesive method used for joining of the polymers used for tests. This study could be helpful for each application where we have to connect these types of polymers.

Introduction

Over the last two decades, industrial technologies using laser machines were frequently addressed in many papers from different scientific fields [1-5]. The lasers have greatly facilitated the production in the automotive, aerospace, and electronics industry. Thanks to the rapid development of lasers, different modifications in the surface layers and in materials, which were previously considered difficult for machining for various reasons, are now possible. Currently, laser micro-treatment is an increasingly common method for modifying the surface properties of different construction materials [6-9]. The continuous improvement of the existing design solutions results in applying the latest achievements from materials engineering, especially in the above-mentioned industry branches. The use of lasers for this purpose seems to be justified. The impact of a laser beam on the surface of different materials was the subject of many scientific manuscripts presented in numerous publications [10,11]. In these publications was discussed the influence of laser parameters (such as light wavelength, frequency of impulses, duration of the impulse, etc.) on the surface of the material being machined. The use of different laser surface modification techniques of polymers was elaborated on in detail in papers [12,13].

Mechanical fasteners and welding belong to traditional joining techniques, with which many manufacturers feel comfortable. However, it has to be taken into consideration that these methods

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are not the most practical solutions to perform a modern assembly. The application of some mechanical fasteners can lead to an increase in production costs, and structure weight; and it can limit the material options to choose from or result in their fatigue, strain, or even tear. Strong adhesives or tapes can outperform mechanical fasteners in many structural applications ensuring a clean and durable structure. That is why a growing number of manufacturers from various industry branches are searching for industrial adhesives as an alternative to traditional joining techniques. Thanks to achieving high reliability of objects, more and more products are made as permanent assemblies, in which a bonding technology is widely used.

This manuscript is dedicated to the possibilities of using a laser micropatterning of a surface to obtain a stronger adhesive joint of the selected construction materials. In the tests, polyethylene terephthalate (PET) was used and joined with the Multibond 1101 epoxy glue. The surface of both sides of the material was micropatterned with a picosecond laser in order to obtain a better mechanical joint. The aim of this study was to offer an innovative joining solution that can reduce the power/pressure requirement for joining and provide a better joint. For this study, the effect of a pre-patterned surface of PET for joining will be demonstrated, and the strength tests of the joining process with and without micropatterning will be discussed. The research shows that bondings between elements with the appropriately machined microstructure are characterized by a severalfold increase in the strength of joints in relation to materials devoid of the microstructure.

This manuscript aims to present the impact of the type of laser-machined micro-texture on the material's surface on increasing the adhesive force in adhesive joints. In addition, this study addresses practical solutions to the adhesive method used for joining of the polymers used for tests. This study could be helpful for each application where we have to connect these types of polymers.

Materials and Methods

Polyethylene Terephthalate (PET), has special physical properties, and is ideal for the production of very precise parts and mechanical components, which are expected to be distinguished by resistance to very high loads and abrasion resistance. It is also used in electrical insulating components applied in electrical engineering, dimensionally stable and very precise mechanical parts (such as sleeves, guides, gear wheels, rolls, pump elements, etc.), heavily loaded bearings, sleeves, guides, and thrust washers. It is also widely applied in the automotive industry and the construction of yachts and ship vessels.

The MULTIBOND-1101 is a cold-hardened two-component epoxy adhesive with a fluid, semi-fluid, and gel-type consistency. It contains a colorless epoxy resin (component A) and yellow hardener (component B). MULTIBOND-1101 wets and even saturates porous materials and is easy to apply (mixing 1:1 by volume). The hardening process is homogeneous in the entire resin mass during the average time. It is applied for joining metals, stones, ceramics, and other hard materials except for polyolefins (PE, PP, PTFE). Typical applications include automotive elements and machine parts, elements of railway carriages, buses, and yachts. This adhesive has high resistance to water, oils, and petrol.

Micropatterning of tested materials was made on a laboratory stand equipped with a picosecond TRUMPF 5325c laser generating UV laser beam with a wavelength of 343 nm. The SNANLAB intelliSCAN 14 scanning head was used for positioning laser beams in the working area. Before laser micropatterning, the surface of the samples was cleaned with ethanol. The exemplary results of the conducted micro-treatment were shown in the images, which were taken with the use of the HIROX KH-8700 optical microscope (Fig. 1). After laser micropatterning samples were rinsed sequentially in an ultrasound bath with ethanol and deionized water. Next, an overlap joint between materials was formed using Multibond 1101 epoxy adhesive. The diagram of joining elements was

Materials Research Proceedings **24** (2022) 27-33 https://doi.org/10.21741/9781644902059-5

presented and described below (Fig. 2). Subsequently, the INSTRON 4502 testing machine was used to perform strength tests, which enabled us to determine to what extent the micro-treatment of the selected geometric shape affected the increase in force needed to break the joint.

Results and Discussion

The main aspect of our work is to obtain a type of microstructure that improves the strength of the glued joints and at the same time doesn't destroy the surface properties of the material. The microstructure should change the properties of the material surface that will allow it to better penetrate by glue and increase the adhesive strength. Improper micropatterning may change the surface properties to hydrophobic, which will significantly impede the spread of the adhesive on the surface. The authors selected two common shapes parallel-line and circles which are presented in Fig. 1.

The effect of micro-treatment was obtained by ablative removal of material from the sample surface [10] by linear beam movement. This is one of the available options to perform microtexture measurements on this laboratory stand. Further research will be focused on other variants of shape, density, and depth of laser machined microtexture of other polymers. The parameters of the TruMicro 5325c laser machine, which was used to carry out microtexture measurements are following. Pulse energy – 12.6 µJ. Pulse repetition rate – 200 kHz. Laser scanning speed – 1000 mm/s. Type of shield gas – air.

According to the procedure described above, microstructures showing parallel lines perpendicular to the direction of the peeling force with a distance of approximately 50 µm and an average depth of 30 µm were performed on the surface of machined materials intended for bonding. They can be found in the following images. The microtexture that revealed circles covered 50% of the area intended for bonding. A diameter of a singular element was approximately 1 mm, and its average depth was 70 µm. The distribution and density of these elements were selected based on experimental tests conducted by the authors of this manuscript. Both parallel-line and circles textures were performed on both joined sample surfaces.

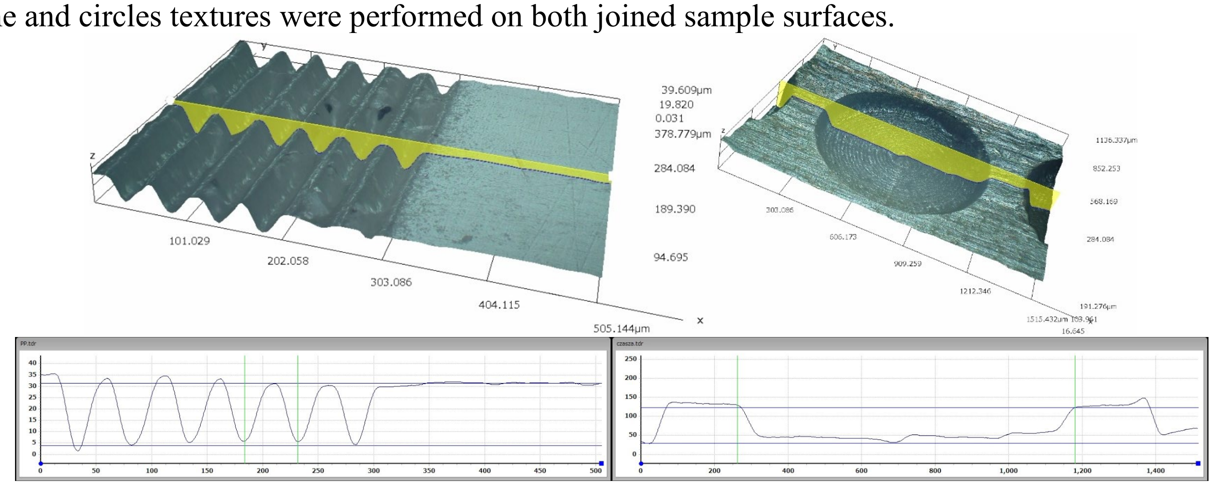

Fig. 1. *General view of the lateral profile of micropattern like parallel lines and circles made on the sample surface.*

Irrespective of the type of the applied adhesive, surface preparation is of key importance for ensuring a durable and stable adhesive joint due to the fact that the joint strength is determined to a large extent by the adhesion rate between the surface and the adhesive. Dirty surfaces inhibit adhesion and require cleaning to ensure an optimal joint. Some adhesives can bond by surface impurities, and others can require pre-cleaning before bonding. Each sample was cleaned with

Materials Research Proceedings **24** (2022) 27-33 https://doi.org/10.21741/9781644902059-5

isopropanol to remove impurities before conducting the laser micro-treatment process. After that, the samples were purified with compressed air in order to remove any material residues. The samples prepared in this way were bonded in specially designed equipment, which enabled a constant and reproducible thickness of the 1 mm adhesive joint. This ensured a visual inspection of the bond. No additional pressure was necessary. Samples were left to fully dry for 24 hours. Then, the samples were placed in the INSTRON 4502 testing machine jaws and strength tests were performed.

During the tensile test, the sample was subjected to uniaxial strain. Measurements of forces were recorded. Such investigation is one of the fundamental sources that can provide information on the mechanical properties of plastics and adhesive joints. The values measured in these samples include strain (elongation) and strain force (fracture connection). Thus, tensile strength is the maximum stress that the material transfers during static tension. Tests were conducted for five samples with microstructure and five samples without microstructure.

Diagrams below show the tensile force of the formed joint. The plastic flow of the sample was not observed due to brittle cracking both in the applied binder and the examined material. The diagram of bonded elements was presented in Fig. 2, where 1 is texturing zone and 2 is the sample gripping section. Texture measurements were performed on the 12 mm x 12 mm surface on one side of bonded elements. Texturing zone was coated with adhesive, and an overlap joint was created, which was marked on a diagram. Samples for the static tensile test were prepared according to the requirements of PN-EN ISO 6892-1 standard.

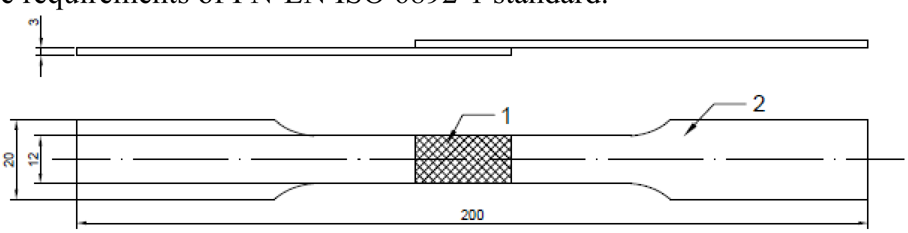

Fig. 2. *Diagram of the lap joint.*

Table 1 presents a comparison of the average results of the breaking strength of the joint made and the value of the increase in strength of the joint with the texture in relation to the joint without texture.

Diagrams in Figs. 3 and 4 illustrate the relationship between sample displacement and tensile force.

Table 1. *Summary of the results of strength tests.*

	PET without micropattern	PET with micropattern like circle	PET with micropattern like perpendicular lines
Measurement 1 [N]	176.7	519.9	393.9
Measurement 2 [N]	188.2	532	386.4
Measurement 3 [N]	119.4	540.9	454.8
Measurement 4 [N]	216.6	555.8	347.4
Measurement 5 [N]	143	553	*X
Average [N]	168.78	540.32	395.62
Standard deviation [N]	38.18	14.89	44.40
Min [N]	119.4	519.9	347.4
Max [N]	216.6	555.8	454.8
The average increase in strength [%]	-	320.13	234.4

*X – sample accidentally destroyed during mounting in the testing machine holder

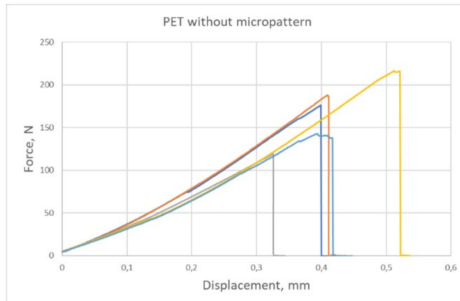

Fig. 3. *PET samples joined without laser micro-treatment.*

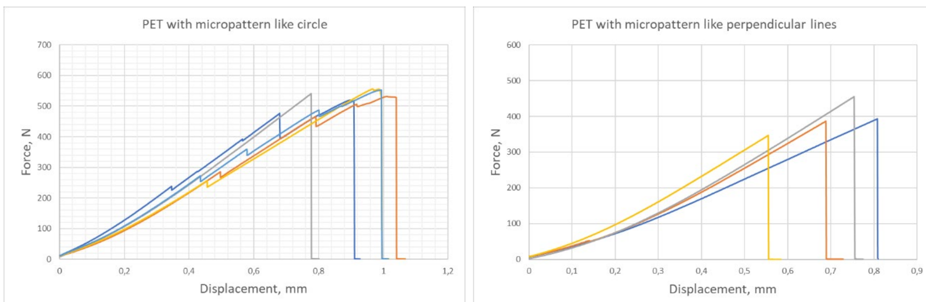

Fig. 4. *PET – PET joints with laser micro-treatment.*

Adhesively-bonded samples without micro-texturing were fractured with an average tensile force of 168.78 N. Specimens that were adhesively bonded with the structure in the form of circles were damaged with an average force of 540.32 N. The average force needed to destroy a sample, which is adhesively bonded with the structure in the shape of circles is higher by 320%. Specimens that were adhesively bonded with the structure in the form of parallel lines were damaged with an average force of 395.62 N. The average force needed to fracture a sample, which is adhesively bonded with the structure in the shape of circles is higher by 234%.

Most polymers are inherently either hydrophobic or only mildly hydrophilic, but they can be made at least temporarily hydrophilic by exposure to a laser beam. The laser treatment method is satisfactory to achieve superhydrophilic patterned surfaces. The implementation of the appropriate geometric shapes of the pattern increases the hydrophilic properties of the surface. The created micro-texture also increases the surface area of the bonded material, which has a considerable impact on the force needed to break the bonding. Unfortunately, improper fabrication of micro-textures affects changes in contact angle values and thus, changes in surface energy of the material. Due to the above, an additional series of tests, which would provide a more in-depth analysis of the obtained results, should be performed. Despite slight divergencies, the authors of the paper received satisfactory results in the perspective of conducting further research on the modification of shape and density parameters of the micro patterns. The authors of the paper noted that the appropriate modification of both the laser parameters and the texture itself will allow for obtaining even more durable connections. Both changing the operating parameters of the laser device and changing the properties of the pattern are of great importance for the properties of the resulting adhesive joint. These properties are closely correlated with each other. A slight change in one parameter can have a big impact on the end result of the process [13].

Conclusions

Ultrashort laser surface micropatterning is a very promising method for creating more advantageous hydrophilic conditions for better adhesives wetting. Many studies have shown their excellent properties for the ablative removal of thin layers of materials. Many methods have been developed for fabricating micro-pattern on the surface with the aim of industrial production. The studies that were conducted prove that the micro-texture type has a significant impact on creating a more durable adhesive bonding using laser micropatterning on the surface of bonded material. As regards PET material, the bonding between surfaces with the micro-texture exhibit much higher tear resistance during the static tensile test than the same bonding without surface patterning. Micropattern-like circles for PET are higher by 320% and it is a very promising result that gives hope of conducting future research on the modification of the shape and density of the microtexture distribution and the possibilities of its implementation on the processed material. Each of these factors contains one or more elements that must also be evaluated. Further strength tests of adhesive joints for other material pairs are also planned: metal - plastic; ceramics - plastic and metal - ceramics, with or without a textured surface, as well as with the use of another type of adhesive agent or without the use of additional adhesive.

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