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COMPOSITES THEORY AND PRACTICE

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# TESTS OF SELECTED MECHANICAL PROPERTIES OF PLA-PLA TYPE COMPOSITES

**This paper presents the results of tests on PLA-based composite materials produced by mixing a base polymer (PLA - polylactide) with PLA fiber. The main aim was to obtain composites with improved mechanical properties. The tested materials contained various amounts of filler in the form of continuous fibers and nonwovens. The samples for the tests were manufactured using the extrusion method. Assessment of their structure and mechanical properties was performed and the results indicate a two-phase internal structure and the filler impact on the increased strength parameters of the obtained composite materials. This offers a possible solution to improve the biofunctional properties of such materials in medical applications.**

**Keywords: PLA-PLA type composites, extrusion, mechanical properties**

## BADANIA WYBRANYCH WŁAŚCIWOŚCI MECHANICZNYCH KOMPOZYTÓW TYPU PLA-PLA

**W pracy przedstawiono wyniki badań materiałów kompozytowych na bazie PLA. Kompozyty otrzymywano poprzez mieszanie bazowego polimeru (PLA) z napełniaczem włóknistym z PLA. Podstawowym celem tych zabiegów było otrzymanie kompozytów o podwyższonych właściwościach mechanicznych. Badane materiały zawierały różne ilości napełniaczy w postaci włókien ciągłych i włókniny. Próbki do badań otrzymano metodą wytłaczania. Oceniano ich strukturę oraz właściwości mechaniczne. Wyniki badań wskazują na dwufazową strukturę wewnętrzną oraz wpływ napełniaczy w kierunku podwyższenia parametrów wytrzymałościowych otrzymanych materiałów kompozytowych. Stwarza to nadzieję na polepszenie cech biofunkcjonalnych takich materiałów w aplikacjach medycznych.**

**Słowa kluczowe: kompozyty typu PLA-PLA, wytłaczanie, właściwości mechaniczne**

## INTRODUCTION

Polylactide (PLA) is currently one of the most popular polymers and its possible applications in medicine are an important direction of research [1, 2]. Interest in the material can be observed in, among others, tissue engineering, orthopaedics, surgery, cardiac surgery, and pharmacology. It has also found use as, for example, subcutaneous implants, scaffolds, surgical sutures, stents, drug carriers for controlled release, clamps, clips, surgical masks, bandages, compresses, clothing for medical staff, diapers, paper handkerchiefs, or cosmetic wads [3-5]. PLA has also found widespread use in industry, e.g. in the agriculture and food industry, or home appliances [6, 7]. Despite its popularity and numerous beneficial properties, there are practical problems with its real-life applications. Among the characteristics described in literature as insufficient, ones that merit mention are PLA's load deflection temperature, strength properties, resilience, barrier properties, sensitivity to moisture, rapid physical aging, or brittleness [8]. The issues of the technological usability of PLA are mainly connected with its mechanical properties [9]. As far as bioengineering applications are concerned, despite the fact that PLA could possibly be used as a resorbable biomaterial, its mechanical properties are insufficient for this purpose. Literature data [10-12] show that the values of Young's modulus for the material are within the range of 2÷4 GPa, its tensile strength can reach 60 MPa, while deformation at break is 1÷7%. In addition, some of its configurations behave like vitreous and brittle materials, which is exacerbated by the fact that its strength properties are further reduced during decomposition.

For the reasons mentioned above, the possibilities of improving PLA's biofunctional characteristics are a focus of research in medical applications. As far as the general applications of composite materials of this type are concerned, their mechanical properties are commonly improved through the addition of a powder filler in the form of wood flour, chalk, mica, cellulose, starch, etc. However, in the case of bioengineering applications, especially bioresorbable implants, it is required that after a certain time the biometal should be absorbed without causing harm to the body. This imposes high requirements for the biofunctional characteristics of such biomaterials.

Literature reports concerning the production of PLA-based mixtures [13-15] contain descriptions pointing to the appearance of two-phase structures, or problems connected with miscibility of the ingredients or its complete absence. Generally however, the presented test results may indicate directions for research into optimal solutions for PLA-based composites.

Considering the aforementioned issues, this study is an attempt to produce a PLA-PLA composite of increased mechanical properties in comparison with base PLA. In order to achieve this goal, the conventional extrusion technique for thermoplastics was used. An additional technological problem was the fact that the processed material ranges of processing temperatures were similar to those of the polymer base and the filler.

a)
b)
c)

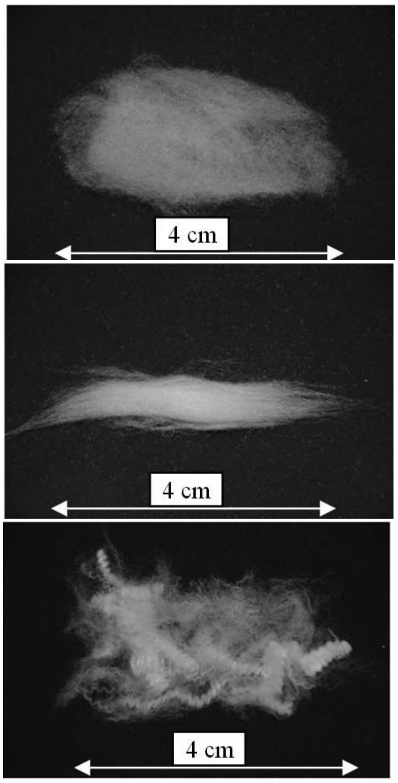


Fig. 1. View of filler used for tests: a) fleece, b) tape, c) cut fiber

Rys. 1. Widok wypełniaczy wykorzystanych do badań: a) runo, b) taśma, c) włókno cięte

## RESEARCH MATERIALS AND METHODOLOGY

### Materials

The materials used for the tests were 3052D type PLA pellets by NatureWorks and fibrous PLA structures. Materials in the following forms were used as fibrous filler (Fig. 1): fleece (R), tape (T), and cut fiber (C). The filler samples were obtained from the Faculty of Material Technologies and Textile Design of Łódź University of Technology.

The composites were produced by extruding a mixture of pellets and fibrous filler. Before commencing the extrusion process, the pellets and filler were conditioned in laboratory conditions (at temperature of 40°C, laboratory dryer) for a period of 24 hours. The form of the obtained filler required it to be shredded into the form of shavings of several millimeters to facilitate processing. Due to the fact that, as is commonly known, using a popular fibrous filler improves the mechanical properties of plastic elements, it was assumed that provided that producing a material according to the chosen methodology was at all possible, then this would most probably lead to improvement of the mechanical properties. In this context, the key problem was to produce the designed material. It was expected that after this obstacle was overcome, tests leading to achieving the best properties could be performed, with the use of a fiber with varied mechanical characteristics. For this reason, a description of the mechanical properties of the used fibers was omitted at this stage.

### Research methodology

The extrusion process was carried out by means of an EHP25Eline laboratory extruder (L/D = 25 ratio). During the extrusion process, the temperatures of the cooking zones of the extruder cylinder and the rotational speed of the screw were controlled. The temperatures of the cooking zones of the cylinder and the head were adjusted within the range of 140÷155°C, the rotational speed of the screw was approx. 10 rpm, i.e. 5% of its maximum value. The low feed rate of the material in the extruder cylinder ensured a longer time for a better yield. At the same time, the time the material stayed in the extruder cylinder (approx. 3÷4 min) was too short for significant processes of temperature degradation to occur. This had an indirect impact on the alloy pressure in the extrusion head. Due to the fact that two materials with similar plastification temperatures were extruded, it was expected that full plastification and a homogenous mix of the components would take place during extrusion. For this reason, the tests did not determine the proportional contents of the base polymer and the filler. It was assumed that the maximum filler content may be 50% and the mixtures were prepared accordingly. Moreover, the point was not to assess the impact of the amount of filler on changes in the mechanical properties of the composite, but rather to verify whether it was possible to produce a composite from two materials of the same kind with similar plastification temperatures (without additional fiber processing modification). For this reason, the focus was on finding a configuration of parameters that would make it possible to obtain a two-phase structure, understood as a clear separation between the polymer base and the filler. It was expected that in the event of success, the probable effect would be obtaining a new material with more beneficial mechanical properties than the base material. Owing to the compatibility of the mixed materials, the process of fiber preparation was omitted. Moreover, no other modifiers nor special methods of filler preparation were used.

In order to assess the internal structure of the obtained samples, tests were performed that consisted in extruding two thin strips of base PLA (a thickness of approx. 4 mm), which were then cut off from the extrusion head and air cooled. A filler sample in the form of a single 20 mm x 80 mm strip was then inserted between the strips (this is hitherto defined as the interlay method). A three-layer structure thus prepared was then formed by means of a hydraulic press (a pressure of 2 kN for 30 s) in a special mold. An element with an external wall thickness of approx. 2 mm was obtained in this manner. External visual examination allowed the authors to directly assess whether filler was present between the transparent PLA layers. The tests were continued until a two-phase structure was obtained, with the fibers clearly visible. The temperature of the cooled PLA strips was assessed using a thermal imaging camera, which allowed the authors to pre-determine the range of temperatures that would make it possible to obtain the expected material. The authors obviously knew the temperatures of the physical states of the mixed materials. The aim of the test, however, was to assess the behavior of materials in conditions approximating real, complex processing and to produce samples of composites by means of the interlay method. These tests enabled the authors to determine the actual temperature increases resulting from, for instance, the processes of friction in the extruder cylinder. After determining the temperatures at which the fibers embedded in the matrix become clearly visible, samples were produced using the interlay method as well as by mixing pellets and filler in the traditional manner, and then the extrusion process was carried out.

After producing the target materials, microscopic observations of fractures and cross-sections were performed (in order to confirm the presence of PLA fibers) using an OLYMPUS Lext OLS 4000 confocal microscope. Before commencing the microscopic observations, the samples were subjected to cutting, grinding and polishing with the use of traditional polishing grinders. In order to assess the mechanical properties of the obtained composites, tensile testing was carried out on a Zwick Roell Z010 universal testing machine, pursuant to DIN EN ISO 527-1 and DIN EN ISO 527-4. It should be added that the authors are aware of the fact that the performed tests are only an initial assessment of the mechanical properties of the produced samples, and more comprehensive tests are required in order to perform a full analysis and draw detailed conclusions.

## RESEARCH RESULTS

Figure 2 shows selected results of microscopic observations of the prepared composite samples.

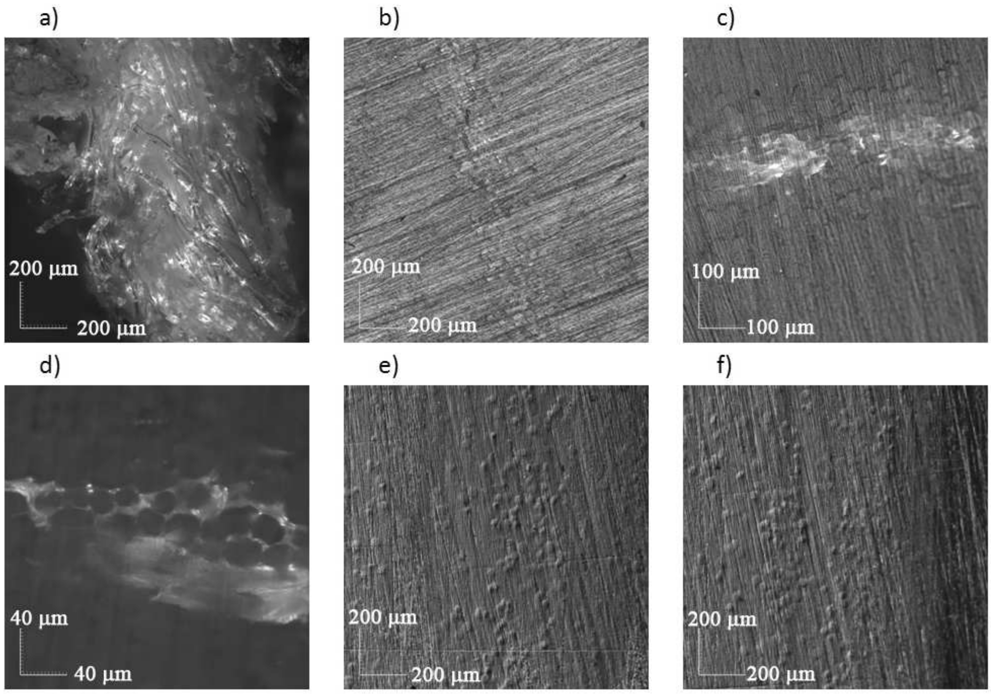


Fig. 2. Results of microscopic observations: a) fibers embedded in matrix (brittle fracture); b), c), d) cross-sections of samples obtained using interlay method; e), f) cross-sections of samples obtained using traditional method

Rys. 2. Wyniki obserwacji mikroskopowych: a) włókna zatopione w osnowie (kruchy przełom); b), c), d) przekroje poprzeczne próbek otrzymanych metodą przekładania; e), f) przekroje poprzeczne próbek otrzymanych metodą tradycyjną

 

Figure 2a shows fibers embedded in the matrix. Figures 2b-d are photographs of cross-sections of a sample obtained with the use of the interlay method. The densification of fibers and their orientation in the same direction result from their arrangement between layers of PLA strips. In the center of the photographs (Figs. 2b-d), round shapes of diagonally cut fibers can clearly be seen. This indicates the possibility of forming a composite from materials with similar plastification temperatures. This result allowed the authors to carry out the next stage of tests, whose goal was to produce composites in the traditional manner. Figures 2 e, f show cross-sections of samples produced in the traditional manner, where transverse cross-sections of fibers distributed throughout the sample can clearly be seen. The orientation of fibers in the same direction is the result of travelling through the cylinder and the process of material extrusion through the opening in the boring head. Dispersion throughout the whole volume indicates a fairly high heterogeneity of material, however, greater clusters of fibers were observed locally. These observations constituted the final proof that producing composites of this type is indeed possible. With this in mind, composite sample sets with fibers described as R, T, and C were prepared, then the samples were prepared for tensile testing by being subjected to machining.

Figure 3 shows selected results of the tests of mechanical properties of the prepared composites.

a)

b)

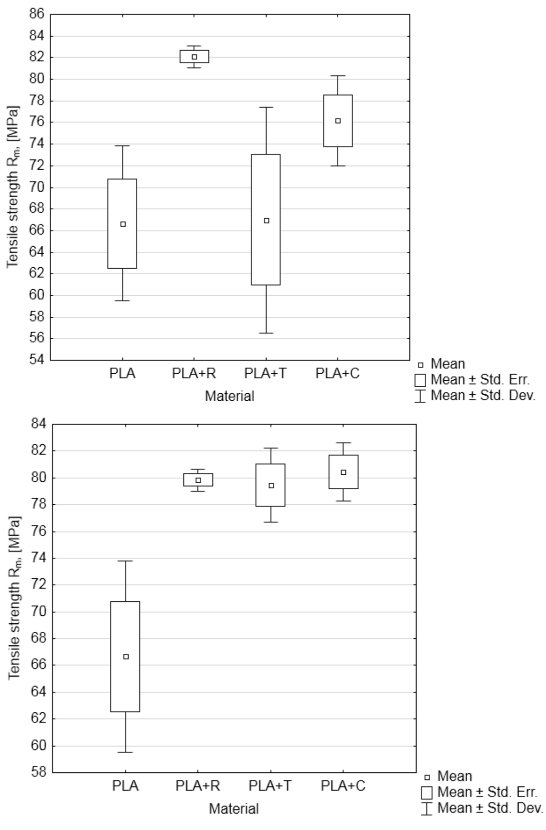


Fig. 3. Values of tensile strength for tested composites: a) obtained using interlay method, b) obtained using traditional method

Rys. 3. Wartości wytrzymałości na rozciąganie dla badanych kompozytów: a) otrzymanych metodą przekładania, b) otrzymanych metodą tradycyjną

Figure 3a compiles the results obtained for the interlay method. An overall increase in tensile strength for the PLA+R and PLA+C composites can be observed. In the case of the PLA+T composite, however, there is a less beneficial influence of filler on the $R_m$ value. This occurred because two PLA layers with filler between them were in some cases torn (Fig. 4) as a result of the alloy temperature being too low for precise connection between the two agglutinated layers to be made, or too great a filler thickness. This phenomenon was observed regardless of the form of filler.

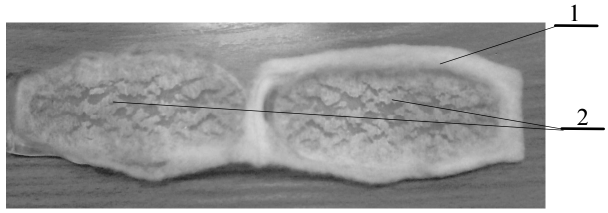


Fig. 4. Two separated composite layers: 1 - filler (fleece), 2 - PLA strips

Rys. 4. Oddzielone dwie warstwy kompozytu: 1 - wypełniacz (runo), 2 - paski PLA

Problems of this kind were not present in the case of the samples produced in the traditional manner. Figure 3b shows the values of tensile strength for the samples obtained with the use of the traditional method. The data show a clear beneficial influence of the used fibers on the $R_m$ values. Figure 5 compiles the $R_m$ values for the prepared composites.

a)

b)

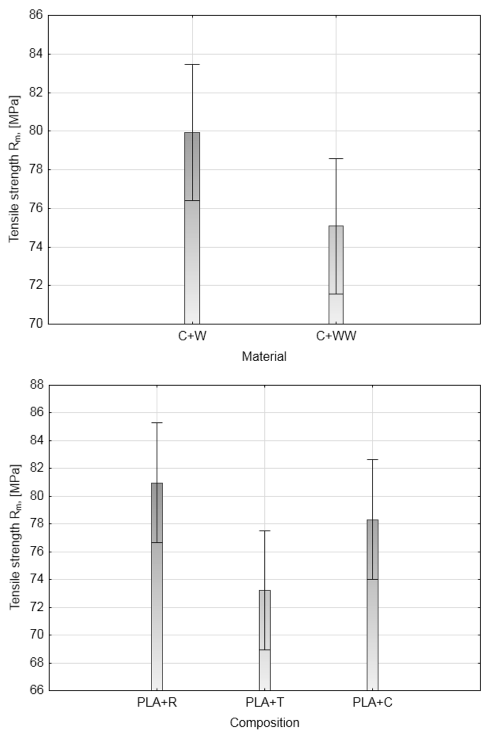


Fig. 5. Values of tensile strength: a) for composites obtained using: traditional method - C+W, and interlay method - C+WW, b) for each prepared composite depending on filler form

Rys. 5. Wartości wytrzymałości na rozciąganie: a) dla kompozytów otrzymywanych metodą tradycyjną - C+W oraz metodą przekładania - C+WW, b) dla wszystkich przygotowanych kompozytów w zależności od postaci wypełniacza

 

Figure 5a pertains to the composites obtained with the traditional method (C+W), and those obtained with the interlay method (C+WW). The presented test results (Fig. 5a) aim at showing that the produced materials, regardless of the method of preparation, are characterized by an increased tensile strength in comparison with pure PLA.

Furthermore, the obtained results allow the conclusion to be drawn that the traditional method yields better results than the interlay method as far as the $R_m$ values are concerned. Figure 5b illustrates the influence of the form of filler on the $R_m$ values. These results show that fleece produces the most beneficial results for tensile strength.

In conclusion, it should be reiterated that the methodology presented in the paper and the obtained materials constitute only an initial attempt at producing composites of the kind in question. The presented test results are a pilot evaluation of the mechanical properties of the tested samples. For a full assessment of their functional quality and full substantial inference, more comprehensive tests should be carried out.

## SUMMARY AND CONCLUSIONS

The paper presents the methodology for producing PLA-based composite materials with the use of PLA fibers. A laboratory extruder was used in order to plastify PLA and two methods were tested, i.e. the method defined as the interlay method, and the conventional extrusion of a pellet and fiber mixture. Problems arising from the similar ranges of plastification temperatures for the base material and the filler were also indicated. On the basis of the performed tests and their analysis, the following general conclusions were formulated: It is possible to produce composites from polymers with similar ranges of plastification temperatures (PLA-PLA type), without additional modifiers or fiber preparation. Processing temperatures can be adjusted in such a way as to obtain a two-phase structure, understood as a structure with a clear distinction between the matrix and the filler, without additional processing or modifiers.

- The composites produced with the interlay method were characterized by less stable mechanical properties than the composites produced in the traditional manner.
- The tests of the mechanical properties show an increase in tensile strength for the prepared composites in comparison with samples produced from the base material. Obviously, this does not mean that the overall performance of such a type of materials is improved, at this stage, however, the focus was solely on $R_m$ assessment. An increase in tensile strength is one of the characteristics that determines the usability of PLA in bioengineering and owing to the improved mechanical properties of PLA through the use of a bioresorbable and biocompatible filler, a possibility for the use of materials of the type in question in medical applications is created.
- Among the tested fiber structures, the most beneficial mechanical properties were obtained for the filler marked as R (fleece). This corroborates the commonly held theory that the form of filler, provided that the material is identical, has an influence on the mechanical properties of the produced composite.
- The results presented in this paper and the composite production methodology are but an initial attempt at assessing the usability of same type materials (biocompatible, bioresorbable, with similar processing temperatures) for producing PLA-PLA type composites on an industrial scale. Elaboration of the subject matter is planned in order to develop a technological process for the industry.

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