# Mechanical Properties of Hybrid Composites Based on Polypropylene Modified with Natural Fillers

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**Abstract.** Hybrid composites based on a polymer matrix and a natural (renewable) filler can be one of the alternatives to widely used polymers. It is well known that natural fillers have poor adhesion to hydrophobic polymer matrices. In order to improve the interaction between hydrophilic fillers and hydrophobic matrix, fillers are usually pretreated with activators. Composites made by injection molding based on polypropylene and Kraft-lignin, microcellulose from hemp waste with filling from 9 to 27 wt % were investigated in work. In order to enhance the adhesion between the filler and the polymer, a special chemical treatment of the components was used with the addition of talc as a compatibilizing agent. Tensile and bending tests of the composites were carried out, as well as studies on the determination of microstructure features and evaluation of the influence of reinforcement and homogenization of mixtures using SEM. It was established that, compared to primary polypropylene, the flexural strength of composites with microcellulose practically did not change, Young's modulus (E) increased to 12% depending on the degree of filling, and the flexural strength of composites with lignin increased to 42.3 MPa (32%), E up to 1570 MPa (22%). The tensile strength is practically unchanged for the cellulose filler and slightly increased to 10% for lignin, and Young's modulus, depending on the type of filler and degree of filling, increased from 10 to 32%. Structure studies indicate a good interphase interaction of components in composites.

**Keywords:** Hybrid Composites · Kraft-Lignin · Microcellulose · Polypropylene · Flexural Strength · Tensile Strength · Manufacturing Innovation

## 1 Introduction

The global pulp and paper industry produces about 100 billion tons of lignin annually [1]. Approximately 98% lignin is stored in numerous landfills, and only a little bit of them is burned for energy, which poses a severe environmental problem [2].

The most commonly used reinforcing materials are lignocellulose materials: containing cellulose, hemicellulose, lignin, and cellulose fibers. These materials are obtained from various renewable sources, such as sawdust and cotton, technical hemp, recycled

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V. Ivanov et al. (Eds.): DSMIE 2023, LNME, pp. 221–229, 2023.
https://doi.org/10.1007/978-3-031-32774-2_22

corrugated cardboard, sugar cane, [3] and others. Unfortunately, such raw materials are not always available organizationally, economically, and technically. For example, [4], the decomposition of lignin into simpler chemical compounds (phenol, benzene, etc.) is more expensive than their synthesis from oil or gas, compared to the products' quality. By 2050, the plastics industry may need 20% of crude oil supplies to support plastics production if the trend continues [5].

Recently, the study of lignin-filled polymer composites to replace oil-derived polymers has attracted the interest of many global researchers. One of the most common thermoplastics is polypropylene (PP), which is used for various types of products but has a number of disadvantages, namely, the inability to natural degradation and low mechanical characteristics [6]. The world industry produces 100 million tons of PP annually, and if at least 30% of the polymer were replaced with natural components, it would reduce oil consumption by a third, as well as the embodied energy used to produce PP. Moreover, the synthesis of natural filler would initiate the natural degradation process of polypropylene after its use and can increase tensile strength, flexural strength, or elastic modulus. In scientific works, Dias et al. [7] found that the cost of producing a lignin composite on an industrial scale is 70–182 USD/ton, while PP is 1.210 USD/ton [8]. Thus, the grafting of lignin and PP can be cost-effective and will positively impact the carbon footprint and the environment in general. So, the process of developing hybrid composites should take into account the interfacial compatibility and dispersion of the material components.

## 2 Literature Review

The constantly growing number of scientific publications on processing, depolymerization, modification, and application demonstrate the prospects for using lignin. Several review articles on lignin compositions have reviewed the state and potential for developing lignin-containing materials [9–11], which discuss various combinations of lignin with polymers.

In paper [12] examines the effect of homogenization and adhesion on mechanical properties depending on the amount of lignin in the polymer matrix. It was found that a wide range of effects on various properties resulted from the interaction of components with lignin. For example, Young's modulus usually increases due to the stiffness of lignin molecules [13], but strength and ductility often decrease [14]. The results obtained regarding the compatibility and nature of the interaction of the components are quite controversial. Kosikova et al. [16] claimed good adhesion between lignin, PP, and organosolvent, while Jeong et al. [9] reported weak compatibility with polymers, including LDPE and PP. Most of these statements are rarely supported by real experimental evidence and mainly reflect the authors' expectations.

Many authors claim that the character of the interphase interaction of components is the main factor that determines the properties of the composite and the distribution of stresses in the composite.

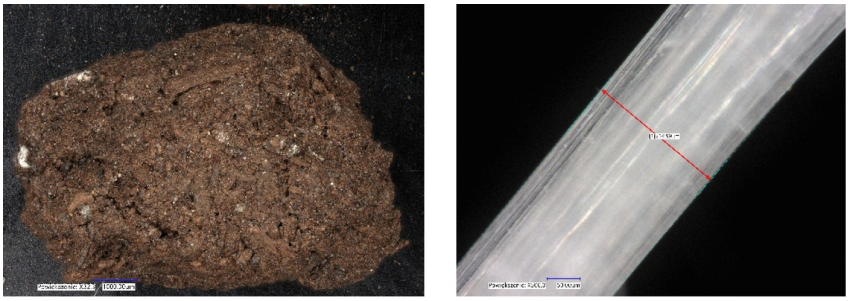
In order to improve the interaction, various agents are added as a component of the composite, or surface modification of natural fillers is used. Modifiers improve miscibility in order to form the desired mixture. Different types of lignin have a low affinity for

polymers [17], which leads to agglomeration. During the addition of lignin, more than 10% wt., it is necessary to apply the homogenizing treatment. Agglomeration of lignin is caused by the presence of intermolecular hydrogen bonds in the structure [18]. The best results were obtained by adding 25–40% lignin as a reinforcing agent to polymers [19]. However, the homogeneity of lignin in the polymer is a problem. There are many types of lignin with different characteristics between polymers and lignin, so there is a constant need to improve composite preparation methods.

This work is devoted to studying the features of the interaction of primary polypropylene as a matrix with pretreated Kraft lignin and microcellulose fibers of plant origin and determining the mechanical characteristics of the obtained composites.

## 3 Research Methodology

The following materials were used for the formation samples of composites: virgin polypropylene granule TIPPLEN H 681f, Kraft lignin, cellulose fibers obtained from the technical hemp (see Fig. 1), talc. Tetrahydrofuran (≥99.5%) and ethanol (≥93.0 wt.%) were used to treat lignin.



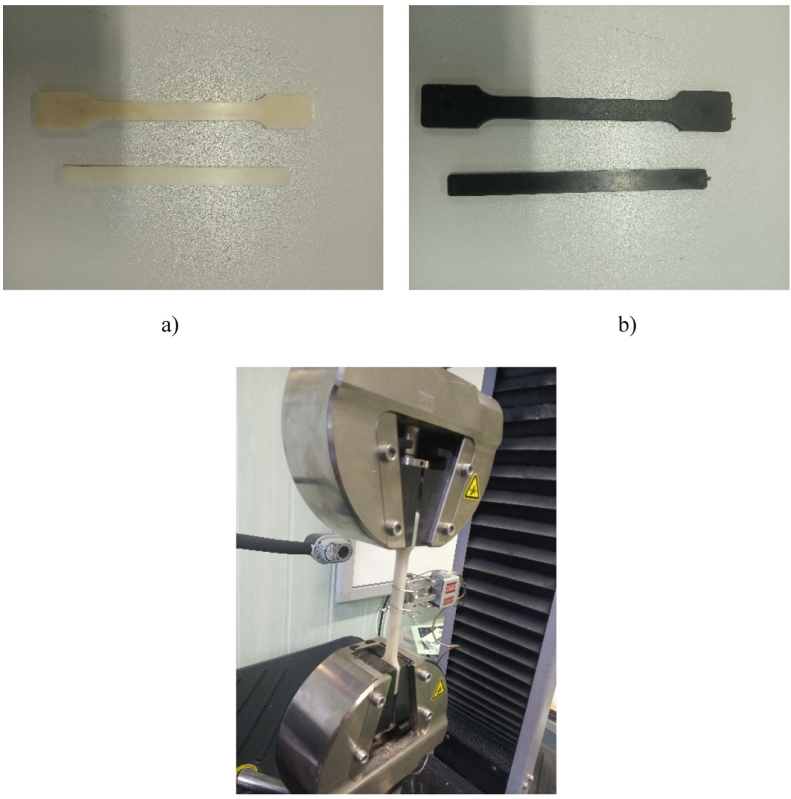
a) b)

**Fig. 1.** Image of Kraft lignin particles (a) and cellulose microfibers (b).

Lignin was treated with a solution consisting of 5 wt.% lignin 35 wt.% tetrahydrofuran, 30 wt.% ethanol, 30.0 wt.% deionized water. Then, the mixture was stirred for 3 h. It was precipitated by adding the solution to deionized water with a mass of 1.72 times greater than the solution. Afterward, water was removed via centrifugation using a refrigerated centrifuge.

The lignin particles containing the substance were extracted and transferred to Petri dishes and then placed in a drying oven at 50 °C for 24 h. The samples (Fig. 2) were obtained by extrusion at 210 °C with a screw rotation of 20 rpm. Each part of 100 g of the composite was stirred for 2 min.

The study of mechanical properties (tensile modulus, tensile strength, and flexural strength) was carried out according to EN ISO 527 and PN-EN ISO 178 standards. Flexural modulus and strength were investigated. The Charpy impact test was performed on



a) b)

c)

**Fig. 2.** General view of samples with cellulose(a) and lignin (b), tensile strength test (c).

notched specimens using a machine Zwick HIT5.5P. Study of the structure of composites after impact tests were observed by scanning electron microscopy (SEM) in a JEOL JSN5510LV. For preparing the samples for SEM were coated with a layer of gold.

## 4 Results and Discussion

For the study, compositions were prepared with the ratio of components by mass presented in Table 1. The content of the filler varied from 9 to 27 wt.% lignin or cellulose and 6 wt.% of talc.

The tensile and flexural strength of the composites, Young's modulus, and impact toughness, were determined to understand the interaction of different preparation methods and composition impact on the properties of the composites. The resulting dependencies are shown in Figs. 3, 4 and 5.

Thus, analyzing the tensile test results (Fig. 3), it can be determined that all compositions' modulus of elasticity has increased. This characteristic's highest result was obtained by composition filling microcellulose, in which the modulus of elasticity

**Table 1.** The numbers, compositions, and all the prepared composite samples.

| Number compositions | Polypropylene, wt.% | Cellulose, wt.% | Lignin, wt.% | Talk, wt.% |
|---|---|---|---|---|
| 0 | 100 | – | – | – |
| 1 | 85 | 9 | – | 6 |
| 2 | 76 | 18 | – | 6 |
| 3 | 67 | 27 | – | 6 |
| 4 | 85 | – | 9 | 6 |
| 5 | 76 | – | 18 | 6 |
| 6 | 67 | – | 27 | 6 |

increased by 32% and the tensile strength increased by 2% only for the composite filled with 9 wt% lignin. A slight decrease in strength was observed for all other compositions up to 5%.

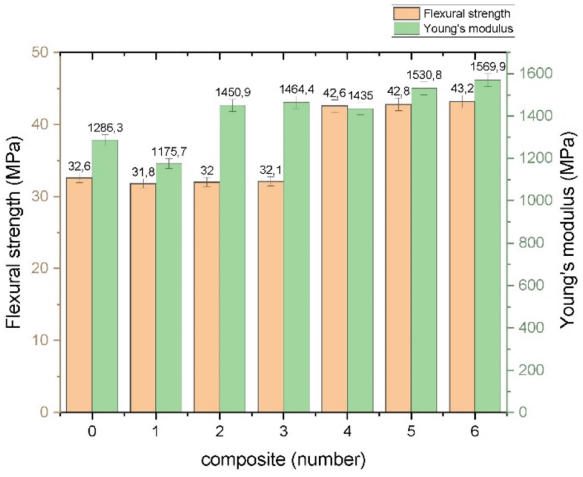



**Fig. 3.** Young's modulus and tensile strength of different composites.

According to the tensile strength results in Fig. 3, no composite with anything other than polypropylene had a higher tensile strength than virgin polypropylene. When the amount of polypropylene was reduced, as microcellulose or lignin was added, the tensile strength gradually decreased, but even at 30% filling, no substantial strength deterioration was observed. This indicates that composite-based lignin or cellulose might be an alternative to polypropylene without losing the products' tensile strength. Moreover, based on the results, it was not possible to prove that the addition of talc positively affects the strength of the composite.

Figure 4 shows the results of bending tests of PP composites at a temperature of +20 °C. The flexural modulus increased for almost all compositions from 10 to 25%. The best flexural strength and modulus of elasticity were obtained for composites No. 6 (with 27 wt %) lignin. Concerning lignin, the increase in bending strength was from 30 to 33% and was practically independent of the degree of filling, a similar dependence on the degree of filling is observed for cellulose, but without an increase in strength compared to primary polypropylene. The good adhesion of the filler can explain the obtained results to the matrix.

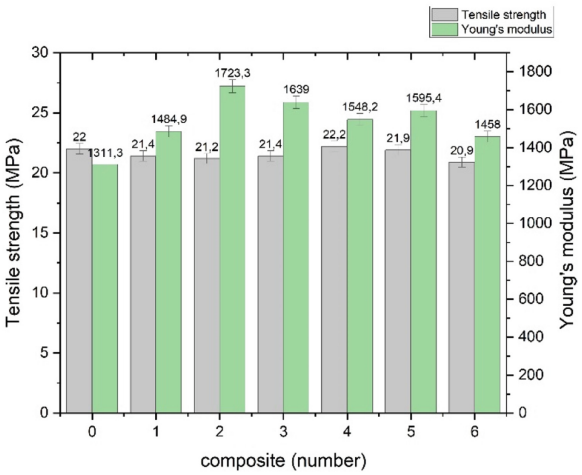



**Fig. 4.** Flexural strength and elastic modulus of different composites.

The results of determining the impact strength are shown in Fig. 5. Thus, for most composites, the impact viscosity was higher in samples with cellulose compared to lignin and polypropylene. The difference was small for dispersed composites, while for fibrous fillers, it was more significant.

The results showed that adding lignin to polypropylene reduces the impact strength of the composite. So, the result was predictable because another study reported a similar behavior and the same results for polypropylene and wood flour composites at 20 wt% and 30 wt%. [3, 4].

In order to determine how well the components are distributed in the composites and how well they interact was prepared SEM images.

The results shown in Fig. 6 (a, b) present that the distribution and dispersion of cellulose microfibers in the polypropylene matrix were the same for all composites. In the images, can be seen certain bright parts which may be cellulose aggregates.

Analyzing the structure of the composites, one can notice a difference in the images containing different amounts of filler, which characterizes the nature of the lignin distribution. It is noticeable that as the lignin content in the composites increases, the structure becomes more densely packed, and the matrix becomes elongated. However, the distribution surface of Fig. 6 d, between the particles of lignin and polymer, is relatively

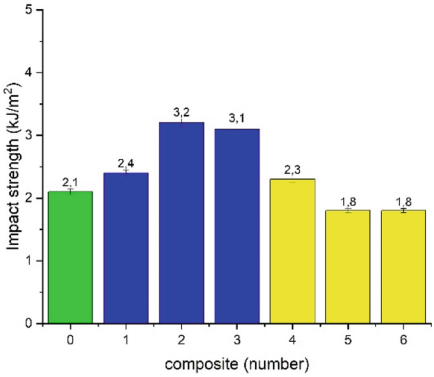



**Fig. 5.** Notched Charpy impact strength.

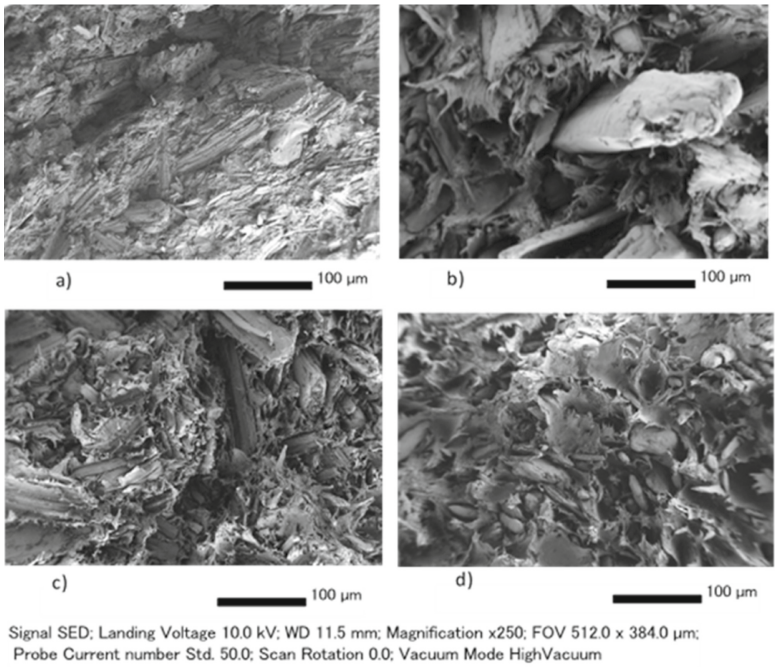



**Fig. 6.** The fracture surface of the tested tensile strength samples: (a; b) – 9 and 27% microcellulose; (c; d) – 9 and 27% lignin.

homogeneous, which is a consequence of the previous chemical treatment of Kraft lignin. In the composites with cellulose, it is observed that the number of holes created during the pulling out of the fibers during the tensile test also increased with the increase in the amount of cellulose. This may indicate that the compatibility between components in cellulose composites is worse than those containing polypropylene and lignin.

## 5 Conclusions

This study aims to develop hybrid composites made of polypropylene and paper waste and determine the effect of chemical composition on mechanical properties and structure. A strength-enhancing effect was not expected, but an improvement in stiffness was highly likely. This work established that lignin-cellulose fillers, both fibrous and dispersed in content up to 30%, do not impair tensile strength and increase bending strength and impact toughness at a certain composition. Consequently, the flexural strength of composites with lignin increased to 42.3 MPa (32%) and Young's Modulus to 1570 MPa (22%). The tensile strength limit for cellulose filler practically does not change, and for lignin, it slightly increases to 5%, and Young's modulus, depending on the type of filler and degree of filling, increases from 10 to 32%. Structure studies indicate a good interphase interaction of components in composites. As a result, it can be stated that lignin in the composition of hybrid composites has good prospects for replacing the exhaustible resource from which polypropylene is polymerized. Of course, further research is needed, as the results could be affected by unsystematic dispersion, but the perspective of using lignin in composites has been proven. In future studies, we have to study how a more significant amount of lignin will affect the composite and heat resistance, UV resistance, and water absorption.

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