Influence of various forms of cellulose on the mechanical properties of polymer composites modified in a constant magnetic field

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Abstract: The paper presents the study of polymer composites on epoxy resin matrix, polymerized in the environment of a constant magnetic field. The composites contained admixtures of various forms of cellulose – microcrystalline cellulose and cellulose from waste hemp straw fibers – in an amount ranging from 10 to 30 wt%. Changes in the mechanical properties due to the effect of a constant magnetic field with a magnetic induction value of $B = 0.5$ T. The composites additionally contained magnetic particles in the form of carbonyl iron, in the amount of 10 wt%.

Keywords: cellulose, polymer composite, constant magnetic field.

The aim of the study was to find polymer composites with new properties. Biodegradable polymer composites are more and more frequently becoming an alternative to plastics, which are difficult to dispose of. The desired features of the novel composites include cheaper, environmentally friendly materials, at least partially biodegradable, functional, resistant to mechanical loads, with additional, e.g. antibacterial properties. Cellulose, added in various quantities but also in various forms, can be a biodegradable filler. Cellulose is a chemical compound with the chemical formula $(C_{6}H_{10}O_{5})_{n}$, made up of 3000–14000 glucose molecules (Fig. 1), connected by $\beta$-1,4-glycosidic bonds, typical of carbohydrates. The molar mass of cellulose is 160–560 kg/mol. The cellulose chains are about seven micrometers long, and the $\beta$ bond forms rigid, long threads arranged in parallel.

The constant magnetic field used for modification can be one of the external factors such as temperature, pressure, under the influence of which the properties of the formed polymer composites are altered. Such novel composites can be used: in the chemical industry, in the household, in the construction, road, sewage or electrical industries. Tests were carried out on composites on various polymer matrices, also made of polyethylene [1, 2].

The properties of composites [3] as well as their surfaces or layers were investigated [4, 5]. Biodegradable composites also had antibacterial properties [6, 7]. Cellulose biopolymer materials attract more and more attention due

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Fig. 1. Chemical formula of the cellulose chain, where $n$ is the amount of glucose molecules

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to their numerous applications in a wide range of areas. The use of cellulose in biocomposite materials has been an object of international research since the mid-1980s [8]. Recently, composite fibers, derivatives of cellulose, have been used in composites as the matrix and fillers. Numerous advanced, functional, cellulose-based polymeric materials have also been obtained. These polymers are improved by the inclusion of glycopolymers obtained from a glucose monomer derived from cellulose, or various cellulose forms that act as matrices [9]. Various types of waste, e.g. flax fibers, are used as fillers [10, 11]. Biodegradable polymer-flax type composites demonstrated mechanical properties comparable to, or better than polypropylene composites. In addition, a comparison of the interfacial tensile properties of the composite showed that fiber-matrix adhesion played an important role in biodegradation [12]. As it has been observed, magnetic field can increase the rate of the polymerization process by up to 55–70%, due to the orientation of the fibers along the force lines of the magnetic field [13–15]. The method of application of carbonyl iron particles using epoxy silane was developed by Charles A.D.M et al. [16]. Experimental results of mechanical tests have demonstrated that the use of these particles increases rigidity, fracture resistance and induction heating rates as compared to other composites. An example of the effect of the addition of magnetic particles and natural particles on the properties of materials are composites consisting of a biopolymer chitosan matrix and a hybrid filler CoFe$_2$O$_4$ - cellulose. The introduction of cellulose into the oxide-chitosan composite modifies significantly its magnetic and mechanical properties. The presence of the filler hindered the movement of the molecules, which resulted in a decrease in the activation energy. The addition of cellulose increased the coercive field (Hc) compared to pure CoFe$_2$O$_4$ powder from 0.1453 to 0.2033 T. The introduction of cellulose filler resulted in an improvement in the Young’s modulus and tensile strength compared to chitosan without the filler. This strength was more than twice higher for the composites with cellulose filler than for pure chitosan [17]. The effect of a constant magnetic field on the gradient structure of composites on an aluminum matrix was also studied. The results of the experiment showed that the application of a constant magnetic field during the solidification of the metal caused the formation of a gradient structure. The flow forced by the magnetic field significantly changed the structure of the solidifying aluminum matrix of the composite under the influence of the magnetic field [18].

**EXPERIMENTAL PART**

**Materials**

The components for preparation of the polymer composites included: epoxy resin “Epidian 5”, Organika-Sarzyna, (Nowa Sarzyna, Poland); IDA hardener, Organika-Sarzyna, (Nowa Sarzyna, Poland); Avicel PH microcrystalline cellulose – 101 (Merck Life Science Sp.z.o.o., Darmstadt, Germany); waste hemp straw fiber cellulose and carbonyl iron (magnetic particles), (97%, Alfa Aesar, Thermo Fisher Scientific, Winsford, UK)

### Table 1. Types of the tested polymer composites with various forms of cellulose on “Epidian 5”epoxy resin matrix

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Addition of magnetic particles (carbonyl iron) % wt.</th>
<th>Type of cellulose added</th>
<th>Cellulose content % wt.</th>
<th>Magnetic induction B, T</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 and 1’</td>
<td>10</td>
<td>Microcrystalline cellulose “Avicel PH-101”</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>2 and 2’</td>
<td>10</td>
<td></td>
<td>20</td>
<td>0</td>
</tr>
<tr>
<td>3 and 3’</td>
<td>10</td>
<td>Cellulose from hemp straw waste fibers</td>
<td>30</td>
<td>0</td>
</tr>
<tr>
<td>4 and 4’</td>
<td>10</td>
<td></td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>5 and 5’</td>
<td>10</td>
<td></td>
<td>20</td>
<td>0</td>
</tr>
<tr>
<td>6 and 6’</td>
<td>10</td>
<td></td>
<td>30</td>
<td>0</td>
</tr>
</tbody>
</table>
Microcrystalline cellulose (Avicel PH – 101) is purified, partially depolymerized cellulose. It is an insoluble white powder with a density of 1.5 g/cm³ (Fig. 2).

Cellulose in the form of waste hemp straw fibers has been used for centuries to manufacture extremely durable and strong products (Fig. 3.). The fibers are characterized by high strength and protect against the penetration of ultraviolet radiation.

All polymer composites were prepared according to the same procedure. First, the composition of each of them was developed, the individual components were weighed and then added in the appropriate order (Epidian 5 epoxy resin, magnetic particles in the form of carbonyl iron, the filler and the catalyst). The whole was mixed mechanically and then the liquid mass of the polymer composite was placed in previously prepared molds according to PN-EN ISO 10210: 2018-1. Some of the samples were placed, for the polymerization period, between the poles of a laboratory electromagnet, in a constant magnetic field environment with magnetic induction B. Some of the samples were left for the polymerization period outside the electromagnet, where the magnetic induction was B = 0.

**Methods**

Mechanical strength tests of composites with respect to tensile strength (according to PN-EN ISO 527-2) and flexural strength (according to PN-EN ISO 178:2003+A1:2005) were carried out on a strength testing machine Zwick/Roell Z050, KL 0.05, with a 50 kN measuring head, test speed of 50 mm/min, tensile module speed of 5 mm/min. Impact strength was tested according to PN-EN ISO 8256:2006. The constant magnetic field with magnetic induction B, in the environment of which some of the samples were studied, was produced using an LS-EM-7V model laboratory electromagnet from Lake Shore Cryotronics. The set included also an electromagnet control power supply, model LS - 648, a teslameeter, model LS - F41 - FC and a Hall probe, model LS - FP - 2X - 250 - TF15.

**RESULTS AND DISCUSSION**

Within the framework of the study of polymer composites, mechanical strength tests including the measurements of tensile, flexural and impact strength were performed. The composites were produced on “Epidian 5” epoxy resin matrix. The admixtures were microcrystalline cellulose, added in the amount of 10, 20 and 30 wt%, or cellulose in the form of waste hemp straw fibers, added in the same quantities. Moreover, magnetic particles in the form of carbonyl iron were always added in the amount of 10 wt%. The samples polymerized both in a constant magnetic field environment with magnetic induction B = 0.5 T and outside the magnetic field (B = 0).

**Tensile strength**

Tensile strength is the greatest stress that a material sample can withstand when stretched. This stress is determined by the formula (I):

\[ R_t = F_r / A \ [kG/cm^2] \]  

where: \( F_r \) – maximum tensile force [kG], \( A \) – cross-section of the stretched sample, perpendicular to the direction of force [cm²].

The tests of polymer composites are presented on the graph of stress dependence on the type of composite (Fig. 4), where (\( \sigma_m \)) means the stress in the material.
during stretching. The tests were carried out according to DIN EN JSO 527-1, the initial force was 0.1 Pa, the test speed was 50 mm/min., the distance of the handles with the sample was 60 mm.

As demonstrated by the tests, the tensile strength of the “Epidian 5” epoxy resin alone was 20.1 MPa. The addition of microcrystalline cellulose increased the tensile strength of the composites. An increase in microcrystalline cellulose content in the amount of 10 to 30 wt% increased the tensile strength by an average of approx. 25%. The constant magnetic field reduced the tensile strength of the composites by approx. 13%. On the other hand, the addition of cellulose in the form of waste hemp straw fibers reduced the tensile strength. The average reduction in tensile strength was approx. 79%. The constant magnetic field further increased the tensile strength of the composite containing added cellulose in the form of waste hemp straw fibers by approx. 22%.

**Flexural strength**

Flexural strength is the greatest stress that a material sample can withstand during flexural. This stress is determined by the formula (2):

\[ R_z = \frac{M}{W} \]  \[ \text{[kG/cm}^2\] (2)

where: \( M \) – flexural moment \([\text{Nm}]\) \([\text{kGcm}]\), \( W \) – indicator of the cross-sectional strength of the bent element \([\text{cm}^3]\) (3),

\[ M = \frac{(F \cdot l)}{4} \]  \[ (3) \]

where: \( F \) – destructive force \([\text{kG}]\), \( l \) – beam span between supports \([\text{cm}]\),

\[ W = \frac{(b \cdot h^2)}{6} \]  \[ (4) \]

where: \( b \) – beam width \([\text{cm}]\), \( h \) – beam height \([\text{cm}]\).

As demonstrated by the tests, the flexural strength of the “Epidian 5” epoxy resin alone was 12.6 MPa. The addition of microcrystalline cellulose increased the flexural strength of the composites. An increase in microcrystalline cellulose content in the amount of 10 to 30 wt% increased the flexural strength by an average of approx. 302% (for an admixture of 10% cellulose), 82% (for an admixture of 20% cellulose) and 33% (for an admixture of 30% cellulose). The constant magnetic field reduced the flexural strength of the composites by approx. 11%. On the other hand, the addition of cellulose in the form of waste hemp straw fibers increased the flexural strength. The average increase in flexural strength was approx. 269%. The constant magnetic field further increased the flexural strength of the composite containing added cellulose in the form of waste hemp straw fibers by approx. 35%.

**Impact strength**

The polymer composites were also tested for impact strength by the Charpy method. The tests were carried out using a QC 639F type Charpy hammer, pendulum mass – 5 J, pendulum impact velocity on the sample – 2.9 m/s. Calculations according to the formula (5):

\[ U = \frac{L}{A} \]  \[ [\text{J/m}^2] \] (5)

where: \( U \) – impact strength, \( L \) – work needed to break the standardized sample \([\text{J}]\), \( A \) – cross-sectional area of the sample at the notch site \([\text{m}^2]\).

As demonstrated by the tests, the mechanical impact strength of the “Epidian 5” epoxy resin alone was 2.61 kJ/m². The addition of microcrystalline cellulose increased the impact strength of the composites. An increase in microcrystalline cellulose content in the amount of 10 to 30 wt% increased the impact strength by approx. 61%. The constant magnetic field additionally increased the impact strength of the polymer composites by approx. 17%. The addition of cellulose in the form of waste hemp straw fibers increased the impact...
Additionally increased the impact strength of polymer straw fibers by approx. 35%. The constant magnetic field by approx. 61% and cellulose in the form of waste hemp lulose increased the impact strength of the composites strength by approx. 35%. Moreover, microcrystalline celulose added in the form of waste hemp straw fibers increased the flexural strength of the composites with microcrystalline cellulose added by approx. 23% and with cellulose in the form of waste hemp straw fibers by approx. 23%.

CONCLUSIONS

The paper presents research on new polymer composites based on “Epidian 5” epoxy resin matrix. The composites contained admixtures, in the amount of 10 to 30 wt%: microcrystalline cellulose (Avicz PH-110), or cellulose from waste hemp straw fibers. The composites additionally contained magnetic particles in the form of carbonyl iron in the amount of 10 wt%. The obtained polymer composites were tested for tensile, flexural and impact strengths. The constant magnetic field with magnetic induction B used in the tests was an additional external factor, under the influence of which some parameters could be changed. Microcrystalline cellulose increased the tensile strength of the composites approx. 25%. In contrast, the addition of cellulose in the form of waste hemp straw fibers reduced the tensile strength by of approx. 79%. The constant magnetic field further increased the tensile strength of the composite with the content of cellulose added in the form of waste hemp straw fibers by approx. 22%. Microcrystalline cellulose and cellulose in the form of waste hemp straw fibers increased the flexural strength of the composites by approx. 269%. The constant magnetic field further increased the flexural strength by approx. 35%. Moreover, microcrystalline cellulose increased the impact strength of the composites by approx. 61% and cellulose in the form of waste hemp straw fibers by approx. 35%. The constant magnetic field additionally increased the impact strength of polymer composites with microcrystalline cellulose added by approx. 17% and with cellulose in the form of waste hemp straw fibers by approx. 23%.

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REFERENCES

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