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The effect of water on electrical properties of polymer composites with cellulose fibers

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Polymer composites with cellulose fibbers are environmental friendly materials with improved mechanical properties. The use of polymer composites with natural fibbers in electrotechnics is hindered by the inherent high moisture absorption of cellulose fibbers, which determines a reduction of electrical properties of composite material. By appropriate chemical surface treatment of cellulose fibbers the effect of water could be diminished. Polypropylene composites with a uniform dispersion of surface treated cellulose fibbers were obtained and characterised. The mechanical properties (tensile strength, modulus of elasticity, hardness) and electrical properties (volume and surface resistivity) of polymer composites are reported. Mechanical and electrical tests point out the efficiency of physical and chemical treatment of cellulose fibbers. The volume resistivity of polymer composite remains almost unchanged after immersion in water when treated cellulose fibbers were used. The treatment with agents capable of creating chemical bonds with cellulose fibbers gives the best results.

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1. Introduction

Nanostructured composites from polymers and cellulose fibbers are breakthrough materials with a low impact on the environment, improved mechanical properties, low density, reduced tool wear, reduced dermal and respiratory irritation when compare with traditional reinforcing materials [1-6].

The interest of using cellulose fibbers for polymer matrix reinforcing is rapidly growing in the last decade because of the above mentioned advantages and because the natural fibbers are from annually renewable resources. The influence of water on mechanical properties of polymer-cellulose composites and the influence of environmental aging on the durability of such composites have been the subject of many papers [7-8]. Less work is concerned with the electrical behaviour of polymer-cellulose composites.

The usage of polymer composites with natural fibbers in electrotechnics is hindered by the inherent high moisture absorption of cellulose which could determine o reduction of electrical properties: increase of electrical conductivity, dielectric losses and dielectric constant and decrease of the breakdown voltage [9]. By appropriate chemical surface treatment of cellulose fibbers the effect of water could be decreased.

The aim of this paper is to present the effects of surface treatment of cellulose fibbers on the mechanical and electrical properties of polypropylene-cellulose composites in dry and wet ageing conditions.

2. Experimental

Composite samples were prepared in the following stages:

1. treatment of cellulose fibbers from hard wood (beech wood from Romania) by adding the coupling agents (agent 1 – $HS(CH_2)_3Si(OCH_3)_3$ from Aldrich, agent 2 - $H_2N(CH_2)_3Si(OC_2H_5)_3$ from Dow Corning) to a 10% cellulose suspension in a 90/10 ethanol/water solution (in volume), stirring for 2 hours, centrifuging and drying 4 hours at 70 ^oC in an oven with air circulation. The heat treatment consisted of curing at 120 ^oC for 2 hours with the purpose to realise chemical bonding;
1. mixing of PP J 700 (Midia Romania) with treated and untreated cellulose fibbers in a Brabender Plasticorder (temperature 175-180 ^oC, rotor speed 60 rpm, processing time 10 min.);
2. mould-pressing at 180 ^oC for 8-10 min and quenching in cold water.

The samples of polymer composites were cut from the pressed composite materials according to standard procedures. They contain polypropylene and 30% untreated cellulose fibbers (***PP/CF***) or polypropylene and 30% cellulose fibbers treated with polypropylene grafted with maleic acid anhydride (***PP/CFm***) or polypropylene and 30% cellulose fibbers treated with agent 1 (***PP/CF1***) or polypropylene and 30% cellulose fibbers treated with agent 2 (***PP/CF2***).

The composites were investigated from the standpoint of mechanical properties and electrical conductivity. Tensile properties of the composites were determined according to SR EN ISO 527:2000 on specimens type I with 25 mm/min, modulus of elasticity at ultrasonic frequencies (apparent dynamic modulus) was determined according to ASTM E494 and Shore hardness was determined according to SR EN ISO 868:1995. Conductivity measurements were carried out using a Keithley 6517 electrometer with a measurement cell connected to a PC-Pentium III computer as described in [10].

Sample sheets 150 mm × 150 mm × 1 mm were immersed in distilled water in an immersion tank at room temperature for different periods of time (0, 72, 168, 336, 1800 hours). Samples were positioned on special grids to avoid contact between them. After the immersion, the sample sheets were dried between two sheets of absorbent paper and conditioned at room temperature for 2 hours.

3. Results and discussion

3.1. Mechanical properties

The mechanical characteristics of the composite materials from polypropylene and treated and untreated cellulose fibbers are presented in Table 1.

The addition of polypropylene grafted with maleic acid anhydride (sample PP/CFm) that determines physical compatibility at polymer-filler interface leads to an increase in the tensile strength, modulus of elasticity and hardness. The addition of curing agents 1 and 2 leads to a significant increase of mechanical strength and hardness (samples PP/CF1 and PP/CF2). This could indicate that a reaction takes place at the interface.

Table 1.

Sample	Tensile strength at break [MPa]	Modulus of elasticity at ultrasonic frequencies [GPa]	Shore hardness [^{0}Sh]
PP/CF	20	7.1	68
PP/CFm	26	7.4	71
PP/CF1	34	7.7	73
PP/CF2	32	7.7	72

Filler treatment conditions provide the hydrolysis of silane coupling agent, the condensation of silanol and the cellulose fibber surface grafting [2]. The reaction between Si-OH and OH groups of cellulose giving irreversible chemical bonding of the silane onto the cellulose surface (Si-O-Cellulose) was pointed out by FTIR analysis [1]. In the processing conditions of polypropylene-cellulose composites the hydrophobisation introduced by mercapto and amino groups could explain the increase of mechanical properties of PP/CF1 and PP/CF2 composite materials.

The treatment with agents capable of creating chemical bonds with cellulose fibbers gives the best results in improving mechanical properties.

3.2. Electrical properties

DC electrical measurements were performed by applying a voltage of 500 V for 2 hours on samples of 100 mm × 100 mm × 2 mm before and after water immersion. The time of 2 hour voltage application were established after the first series of experiments. Differences between the values of volume (or surface) resistivity after 1 hour (3600 s) and 2 hours (7200 s) are significant (Table 2).

Table 2.

	Befor immersio				After immersio 72 h			
Sample	t [s]	ρ_v [Ωm]	t [s]	ρ_s [Ω]	t [s]	ρ_v [Ωm]	t [s]	ρ_s [Ω]
PP/CF	1812	5.01×10^{14}	1805	3.18×10^{16}	1812	1.23×10^{13}	1810	1.28×10^{13}
	3608	6.61×10^{14}	3610	3.08×10^{16}	3603	1.89×10^{13}	3602	2.32×10^{13}
	7206	8.00×10^{14}	7208	3.18×10^{16}	7202	3.87×10^{13}	7209	4.51×10^{13}
PP/CFm	1802	4.05×10^{14}	1811	1.39×10^{16}	1801	1.55×10^{14}	1803	2.18×10^{16}
	3600	5.88×10^{14}	3610	1.79×10^{16}	3613	2.08×10^{14}	3608	3.06×10^{16}
	7033	7.91×10^{14}	7210	1.68×10^{16}	7205	2.75×10^{14}	7202	5.75×10^{16}
PP/CF1	1813	3.35×10^{14}	1800	2.18×10^{17}	1805	2.06×10^{14}	1800	3.93×10^{15}
	3607	4.66×10^{14}	3602	3.69×10^{17}	3200	2.97×10^{14}	3602	5.24×10^{15}
	7033	6.20×10^{14}	7209	6.16×10^{17}	7202	4.95×10^{14}	7213	1.17×10^{16}
PP/CF2	1801	4.75×10^{14}	1801	1.81×10^{16}	1812	2.3×10^{14}	1802	5.12×10^{15}
	3603	6.43×10^{14}	3603	2.61×10^{16}	3611	3.63×10^{14}	3604	7.21×10^{15}
	7207	8.26×10^{14}	7210	4.08×10^{16}	7205	5.51×10^{14}	7208	1.36×10^{16}

Volume and especially surface resistivity of PP/CF composites, which contain untreated cellulose fibbers, decrease with more that an order after 72 hours of water immersion. Polymer composites with treated fibbers (physical or chemical treatment) present similar values for volume and surface resistivity before and after water immersion (Table 1).

The results of electrical measurements on composite samples before and after immersion in water different periods of time are presented in Fig. 1 and 2.

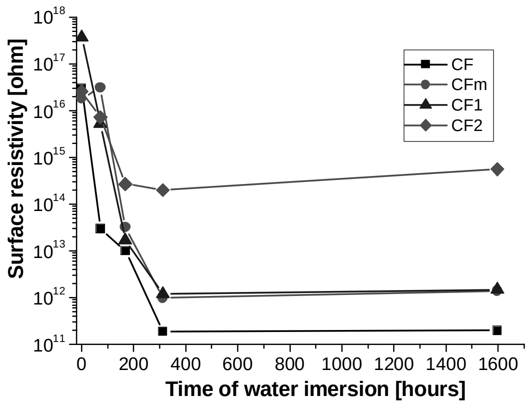

Fig. 1. Surface resistivity of PP-cellulose composite samples before and after water immersion.

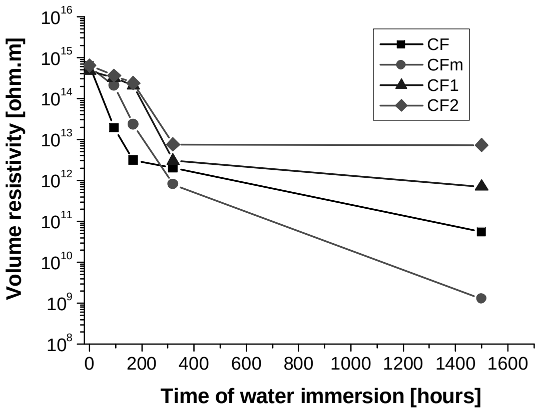

Fig. 2. Volume resistivity of PP-cellulose composite samples before and after water immersion.

The immersion in water of polymer composite samples affects the electrical behaviour of composites, especially during the first 14 days and especially the surface resistivity. The values of volume (only samples with treated fibbers) and surface resistivity rest almost unchanged between 2 weeks and 75 days of immersion in water.

The treatment with agents capable of creating chemical bonds with cellulose fibbers gives the best results in improving electrical properties: PP/CF2 sample presents the greatest surface (Fig. 1) and volume resistivity (Fig. 2) after water immersion of all tested samples.

4. Conclusions

The treatment of cellulose fibbers is essential for improving mechanical properties (tensile strength, modulus of elasticity, hardness) of polymer composites. The treatment with agents capable of creating chemical bonds with cellulose fibbers gives better results in improving mechanical properties.

The immersion in water of polymer composite samples affects the electrical behaviour of composites. The samples of polymer composites with treated cellulose fibbers keep better resistivity values after immersion in water.

The treatment of cellulose fibbers with agents capable of creating chemical bonds (especially with amino groups) improves mechanical and chemical properties of polymer composites immersed in water.

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