PLASMA-MODIFIED WOOD FIBERS AS FILLERS IN POLYMERIC MATERIALS*

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Wood fibers were used as fillers to obtain composites with polyethylene and rubber. In order to improve the compatibility between fiber and polymer matrix, a methane cold plasma was used to modify the wood fiber surface. The tensile properties of the resulted composites were comparatively studied.

Key words: methane cold plasma, polymer composites, wood fiber, tensile properties.

1. INTRODUCTION

Natural fibers are widely used as reinforcing materials in polymeric composites due to their abundance in nature, low density, good resistance to breakage during processing, low cost, etc. [1]. These fillers can strengthen the composites by transfer of stress between the fiber and the polymer matrix. The compatibility of natural fibers with polymeric materials can be improved by chemical modification of polymer or fiber surfaces [2, 3].

Wood fibers can be used for this purpose in polymeric matrices containing, for instance, polyethylene or rubber, but chemical treatment of these fibers by conventional methods requires rather complex processes. Alternatively, cold plasma treatment is a useful technique to modify the surfaces of natural polymers [4] and to graft monomer to polymeric materials [5]. For example, gases like argon, mixture of nitrogen and hydrogen, ammonia, were used to modify cellulosic fibers in plasma conditions [6] and hydrogen and oxygen plasma are used to study the adhesion between plasma-treated cellullosic materials and polyethylene [7].

Another technique used for this purpose is the plasma polymerization which uses methane, ethylene and other gases that can polymerize in plasma conditions.


In this paper, we used a methane plasma for modifying the wood fiber surface in order to improve the compatibility between the fiber and polyethylene matrix. Tentatively, unmodified and modified wood fibers are used beside carbon black as fillers for rubber.

The tensile properties of the resulted polymeric composites are comparatively studied.

2. EXPERIMENTAL

The raw materials were polyethylene of low density, natural rubber and comminuted beech wood fibers of about 80 mesh.

The plasma was produced in a rotational spherical glass vessel of 1 L with two external copper electrodes, by means of a radio-frequency generator operating at a frequency of 13.58 MHz. The pressure in plasma reactor was of 2 torr and methane was introduced at a flow rate of 10 mL/min. In a typical experiment, 30 g of wood fiber were introduced into the reactor and its rotation was started so that all the sample surface was virtually exposed for 10 min. to the plasma.

In the experiments regarding the composites based on polyethylene, the corresponding quantities of polymer and untreated or plasma-treated wood fibers were mixed at a temperature of 150°C and the resulted mixture was compression-molded into dog-bone-shaped tensile test specimens. The samples were then cooled at room temperature with the pressure maintained at the same value during the cooling.

In the second set of experiments, composites of natural rubber were prepared at room temperature and mixtures of carbon black and wood fibers in different proportions were used as fillers. The mixture contained 100 g rubber, 35 g talc, 88 g filler, 5 g zinc oxide, 1.9 g sulphur, as well as antioxidant and accelerators. The resulted material was pressed as plaques of $150 \times 150 \times 2$ mm, vulcanized for 10 min. at 160°C under pressure, and cooled. The test specimens for tensile measurements were cut from this vulcanized material.

The tensile properties were measured with a TIRA test 2162 apparatus.

3. RESULTS AND DISCUSSION

Figs. 1–3 depict the variation of tensile properties vs. proportion of filler for the composites obtained from polyethylene with both untreated and plasma-treated wood fibers. The proportions of the filler were varied between 0 and 30% with respect to the polymeric material.
For the elastic modulus (Fig. 1), the composites obtained with untreated fibers (curve 1) showed the increase of this property with the increasing of filler percentage in the mixture. Samples based on treated fibers show the same behavior, but a higher increase of modulus is observed in this case.
Elongation (Fig. 2) decreased gradually with the addition of wood fiber. A more pronounced decrease of this property values is observed for the samples containing plasma-treated fibers.

Fig. 3 shows that the tensile strength significantly increases with the addition of fillers and a better behavior is exhibited by the composites containing treated wood material.

![Graph](image)

**Fig. 3.** – Strength vs. proportion of filler in composites based on polyethylene for untreated (1) and plasma-treated (2) wood fibers.

This improved efficiency of plasma-treated fibers used as filler may be due to the higher hydrophobicity that they receive during the plasma irradiation.

The tensile properties of different composites based on rubber with untreated and plasma-treated wood-fibers used as filler beside carbon black are presented in Figs. 4–6.

It is observed in this case that the elastic modulus presents a lower value for the composites containing wood fiber or wood fiber – carbon black mixture as filler than those containing only carbon black.

Elongation at break also presents a sharp decrease in the composites containing wood fibers in comparison with those containing carbon black. In the case of mixtures of carbon black and wood fibers, the increase of elongation with the decrease of wood fiber proportion is observed. For a ratio carbon black to wood fiber of 3 to 1, a maximum of this property appears without any significant modification of the modulus. This behavior suggests that there is a better
compatibility between filler and polymeric matrix around this composition of the filler.

As for the tensile strength, this property showed a tendency to increase with the decreasing of the wood fiber proportion (Fig. 6).

Fig. 4. – Elastic modulus vs. proportion of wood fiber in filler for composites based on natural rubber containing untreated (1) and plasma-treated (2) wood fibers.

Fig. 5. – Elongation vs. proportion of wood fiber in filler for composites based on natural rubber containing untreated (1) and plasma-treated (2) wood fibers.
The same behavior regarding the variation of the tensile strength was observed for the composites containing plasma-treated wood fibers. However, in this case a slight decrease of the properties is registered, which suggests a
decrease of compatibility of rubber and wood fibers. It is known that rubber presents a lower hydrophobicity as compared with polyethylene due to the double bonds in its macromolecular structure, and, therefore, exhibits a lower compatibility with plasma-treated fibers.

Another mechanical property of composites with natural rubber, the hardness (Fig. 7), which decreases by increasing the proportion of untreated wood fiber, presents a slight increase in the case of blends containing plasma-treated fibers.

4. CONCLUSIONS

Tensile properties of polyethylene composites containing wood fiber as filler showed that the treatment of the filler in cold high frequency methane plasma improves the compatibility between fiber and polymer matrix. The behavior of polymeric blends is explained by the increased hydrophobicity of fiber surface obtained by this plasma-treatment.

In the case of rubber blends, the presence of wood fiber beside carbon black as filler material, either treated or untreated in methane plasma, diminishes the mechanical properties of the composites. This decreasing is slightly more pronounced for the blends containing plasma-treated wood fibers.

REFERENCES