

# The effect of blending lignin with polyethylene and polypropylene on physical properties

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## Abstract

Lignin, a natural polymer was used as a filler in low-density polyethylene and polypropylene up to 30 wt%. The influence of lignin blending on processing stability, mechanical properties and light and long-term heat degradation are described for both polymer blend types. It was shown, that there is a different degradation behaviour between polyethylene-lignin and polypropylene-lignin blends. The lignin concentration influenced both the tensile strength and melt flow index. © 2000 Elsevier Science Ltd. All rights reserved.

**Keywords:** Polyethylene/lignin blends; Polypropylene/lignin blends; Degradation

## 1. Introduction

Lignin is an amorphous natural polymeric material that is based on phenylpropane derivatives. Lignin has many functions in wood, but its commercial utilisation is relatively low. In biomass lignin is present together with polysaccharides in the amount 20–30% lignin to 60–80% polysaccharides [1]. Various properties of lignin designate it for different technical purposes. Lignin can be utilised as a stabiliser (antioxidant) for plastics and rubber [2], the modified lignins have been used in crosslinked polyurethanes [1] or phenolic resin [3] or as a mechanical properties modifier for rubber mixtures [4,5]. An American patent [6] describes a modification of natural lignin, which is grafted by olefin monomer. Lignin, modified by this method is able to create, by a polymerisation process, a fully biodegradable copolymer. Kosikova et al. [7] describe the influence of lignin addition to polypropylene (PP) on the mechanical properties of PP films, where lignin content varied up to 10 wt%.

The objective of the present paper is to describe the preparation of lignin–polyethylene (low density polyethylene, LDPE) and lignin–polypropylene blends with lignin content up to 30 wt% and the data on the characterisation of their basic properties at various lignin concentration levels.

## 2. Experimental

### 2.1. Materials

LDPE in the powder form, melt flow index = 1.7–2.3 g/10 min, (Bralen RA 2-19 P3, Slovnaft Bratislava, Slovakia) and non-stabilised PP in powder form, melt flow index = 8.0–11.5 g/10 min, (Tatren HPF, Slovnaft Bratislava, Slovakia) were used as polymer matrix. Lignin PLE, powder product of prehydrolysis of beech wood, was developed in the Chemistry Institute, Slovak Academy of Sciences,  $M_w = 2000$  [8].

In addition the following additives were used. Loxamid E, amide of erucic acid, (Henkel KGaA, Germany); Irganox 1010, phenolic antioxidant, (Ciba Speciality Chemicals, Switzerland); and Irgafos 168, phosphitic antioxidant, (Ciba Speciality Chemicals, Switzerland).

### 2.2. Samples preparation

Polymer blends were prepared by extrusion of PP powder or PE powder blends with lignin in a single screw extruder. The obtained extrudates were cut into pieces by a granulator.

Extrusion parameters:

Extruder diameter: 19 mm;  
L/D ratio: 25;  
Compression ratio: 1:4.

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Table 1  
Composition of PE and PP–lignin blends

Samples	PP (wt%)	PE (wt%)	Lignin (wt%)	Loxamid E (wt%)	Irganox 1010 (wt%)	Irgafos 168 (wt%)
1	89	–	–	1	–	–
2	89	–	–	1	0.1	0.1
3	89	–	10	1	–	–
4	–	89	–	1	–	–
5	–	89	–	1	0.1	0.1
6	–	89	10	1	–	–

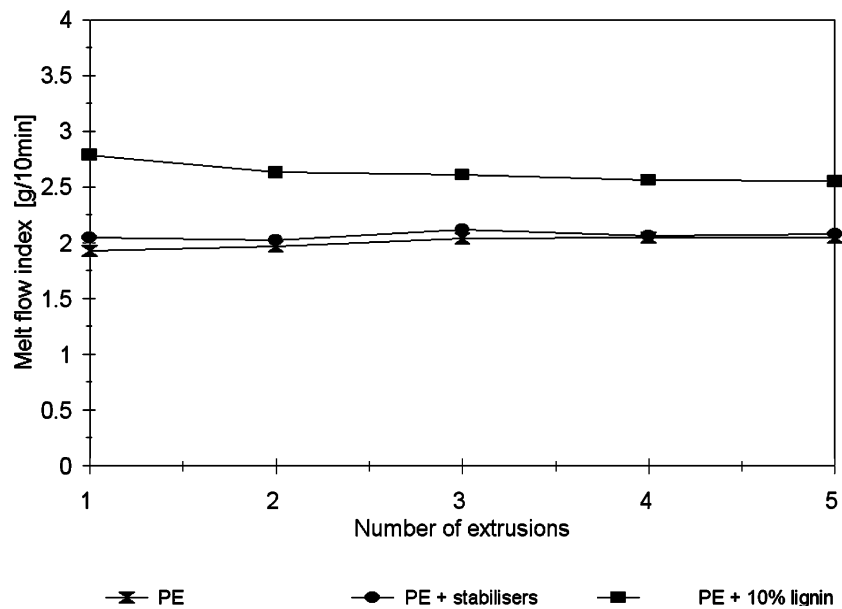


Fig. 1. Dependence of melt flow index on number of extrusions for LDPE mixtures.

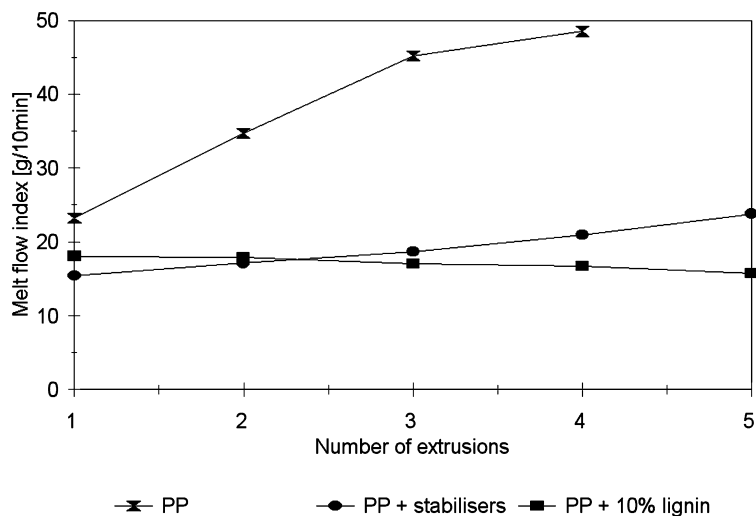


Fig. 2. Dependence of melt flow index on number of extrusions for PP mixtures.

Table 2  
Compositions of PP and PE blends for lignin concentration influence study

Samples	PP (wt%)	PE (wt%)	Lignin (wt%)	Loxamid E (wt%)	Irganox 1010 (wt%)	Irgafos 168 (wt%)
1	99	–	–	1	0.1	0.1
2	–	99	–	1	–	–
3	–	89	10	1	–	–
4	–	79	20	1	–	–
5	–	69	30	1	–	–
6	–	84	15	1	–	–
7	–	74	25	1	–	–
8	89	–	10	1	–	–
9	79	–	20	1	–	–
10	69	–	30	1	–	–
11	84	–	15	1	–	–
12	74	–	25	1	–	–

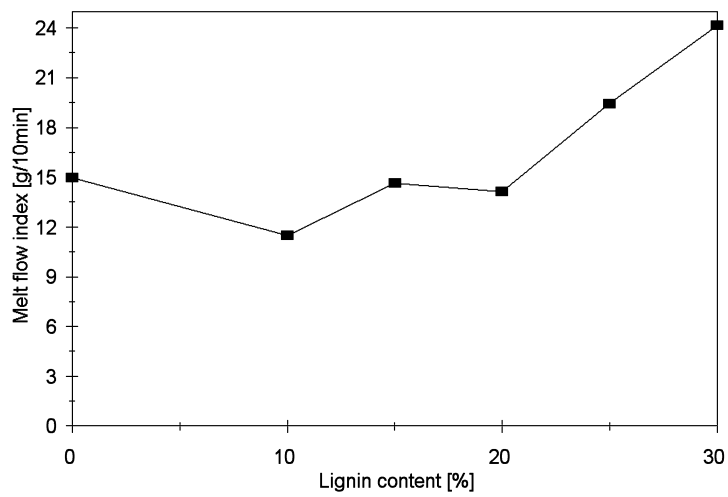


Fig. 3. Dependence of PP melt flow index on lignin content.

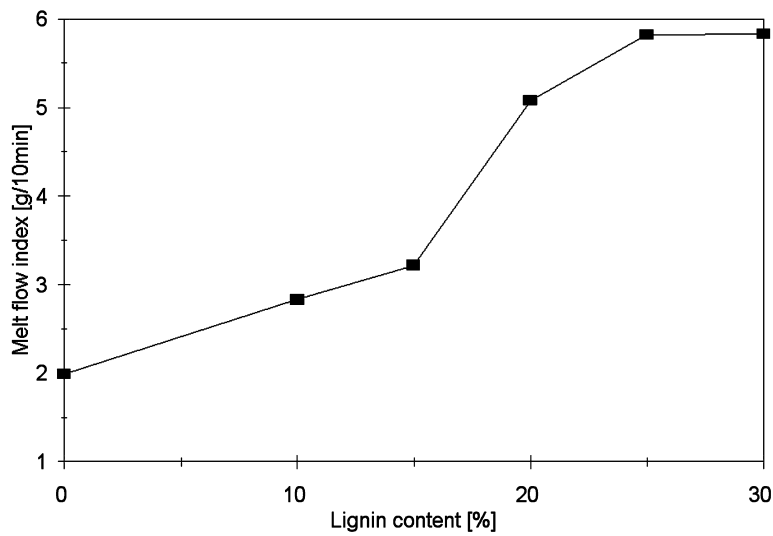


Fig. 4. Dependence of PE melt flow index on lignin content.

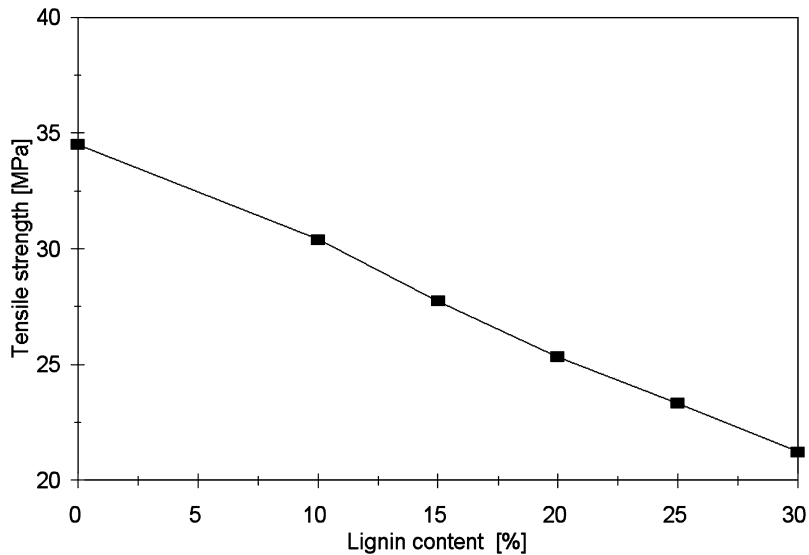


Fig. 5. Dependence of tensile strength on lignin content for PP blends.

Temperature scale for PP from filing part to head was 230–250–260–210°C.

Temperature scale for PE from filing part to head was 150–160–180–170°C.

Screw operation speed: 50/min

The rheological properties and processing stability of the prepared blends were measured according to ASTM D1238 at 190°C and 21.19 N for PE and at 230°C and 21.19 N for PP according to ASTM D 1238/L. Prepared blends were moulded between plates, using the following conditions, in an electrical press.

Temperature for PP: 200°C;

Temperature for PE: 180°C;  
 Pressure: 10 MPa;  
 Preheating time: 2 min;  
 Pressing time: 3 min.

Plates were cooled in a cold press at 10 MPa for 5 min. The tensile strength of samples was then measured using the Tensometer MONSANTO 10 tester according to ASTM D 638. Long-term heat stability was evaluated as tensile strength changes after 500 h exposure in the oven at 130°C. Light stability was tested with a QUV tester, QUV Company, UK, using UVA lamps at 63°C, in 8 h light–dark cycles. Degradation was detected as CO group increment, which was

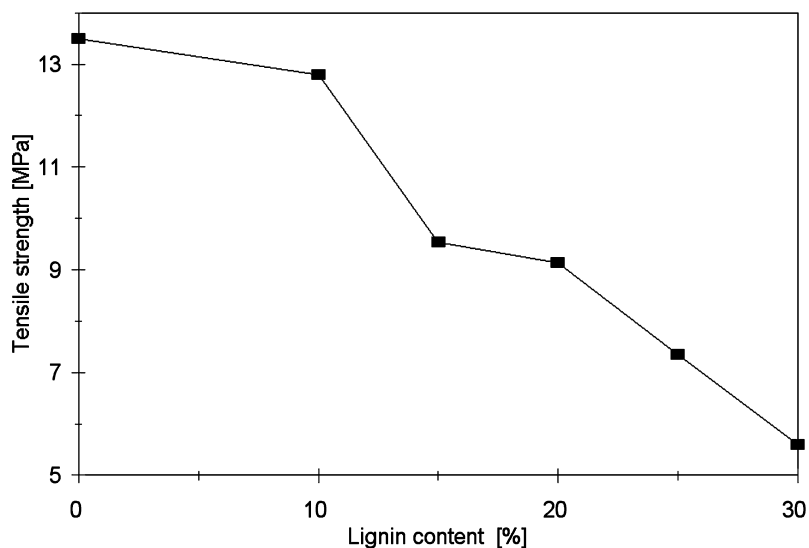


Fig. 6. Dependence of tensile strength on lignin content for PE blends.

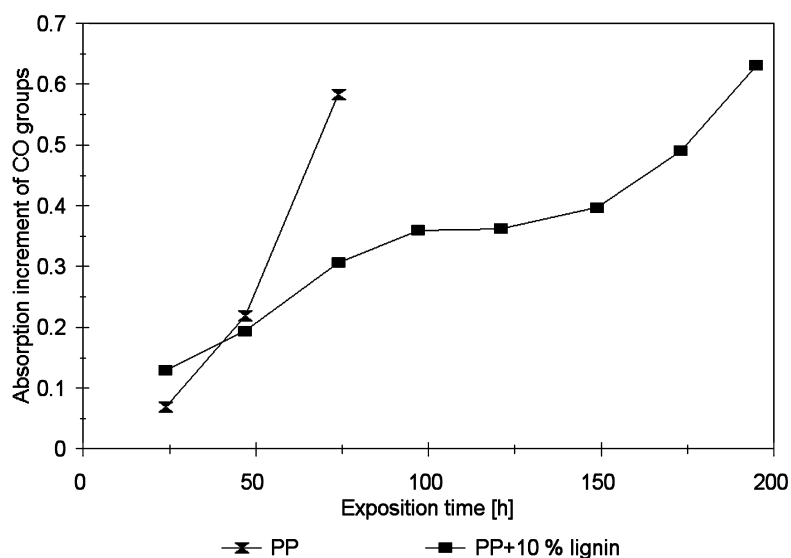


Fig. 7. Dependence of carbonyl group increment on exposure time in QUV tester, for PP–lignin blends.

measured by the IR spectrophotometer Specord M80 at 1730/cm.

### 3. Results and discussion

#### 3.1. Processing stability of lignin blends

The influence of lignin addition to LDPE and PP, on the processing stability of LDPE and PP was studied at a 10 wt% concentration level. The composition of the prepared mixtures is shown in Table 1. The processing stability was determined as a melt flow index (MFI) dependence on the number of successive extrusions. Each sample

was extruded 5 times. The behaviour of compositions containing 10 wt% of lignin were compared with the behaviour of stabilised and non-stabilised raw synthetic polymers (LDPE and PP). Measured dependencies of MFI on the number of extrusions are shown in Figs. 1 and 2. From these two figures it is seen that the addition of lignin and conventional stabilisation system has a similar effect to the stability of LDPE. On the other hand, processing stability of PP is more sensitive to presence of lignin. The MFI of non-stabilised PP increases very rapidly with the number of processing cycles. The addition of conventional stabilisers strongly reduces the MFI increase. The 10 wt% lignin addition to non-stabilised PP changes the MFI increase to a slight MFI decrease.

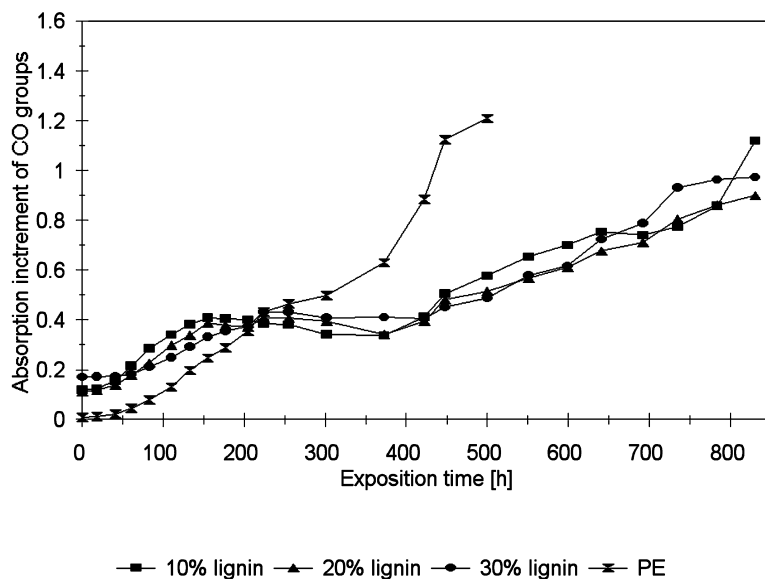


Fig. 8. Dependence of carbonyl group increment on exposure time in QUV tester for PE–lignin blends.

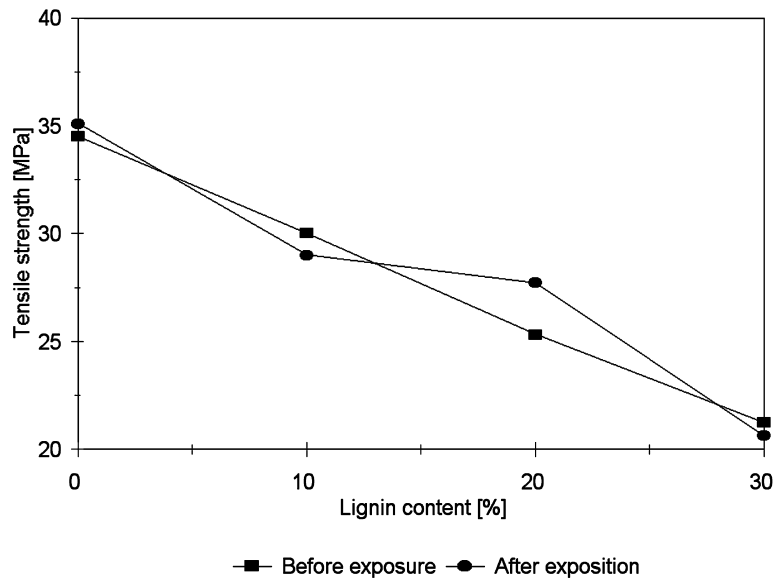


Fig. 9. Dependence of tensile strength of PP, before and after QUV exposure, on lignin contents.

From these results it is clear that lignin acts as an effective processing stabiliser in PP. In the case of PE the material is essentially stable both with and without lignin addition.

### 3.2. Mechanical and processing properties of PP and PE dependencies on lignin concentration

For the study of the effect of lignin addition on the basic mechanical and processing properties, PP and PE–lignin blends were prepared. The composition of mixtures is shown in Table 2. In all PP blends Loxamid E was used as a processing agent. PP–lignin blends were prepared in the

absence of a conventional stabiliser. Pure PP was stabilised with Irganox 1010–Irgafos 168 blend.

MFI measurements were made as a measure of processing characteristics for various lignin concentration levels. Both dependencies (for PP and PE blends) are shown in Figs. 3 and 4. Lignin in PP (Fig. 3), up to 20 wt% concentration, has practically no effect on the MFI of the PP blend. Above this concentration the MFI starts to increase. A different behaviour was observed for the PE–lignin blends (Fig. 4), where the MFI increased continuously with lignin concentration.

The tensile strength dependence on lignin concentration is very similar for both type blends (PP and PE) (Figs. 5 and

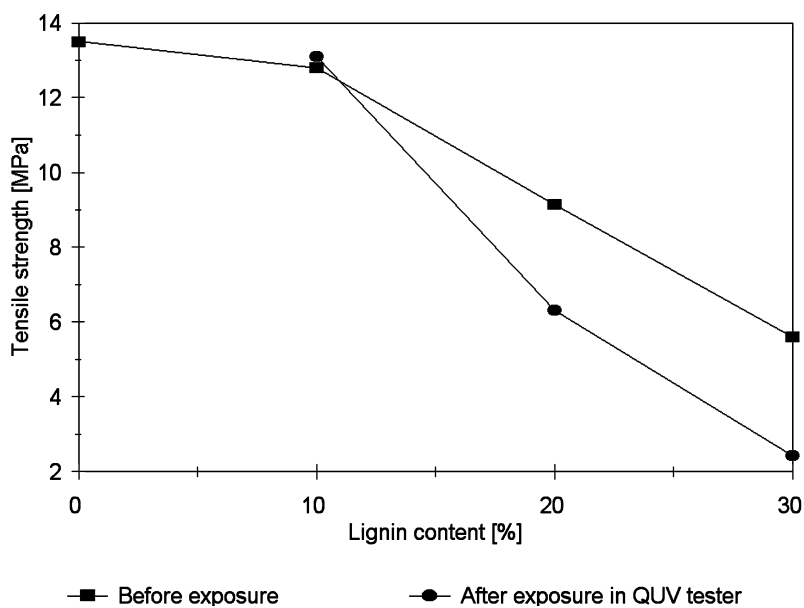


Fig. 10. Dependence of tensile strength of PE, before and after QUV exposure, on lignin contents.

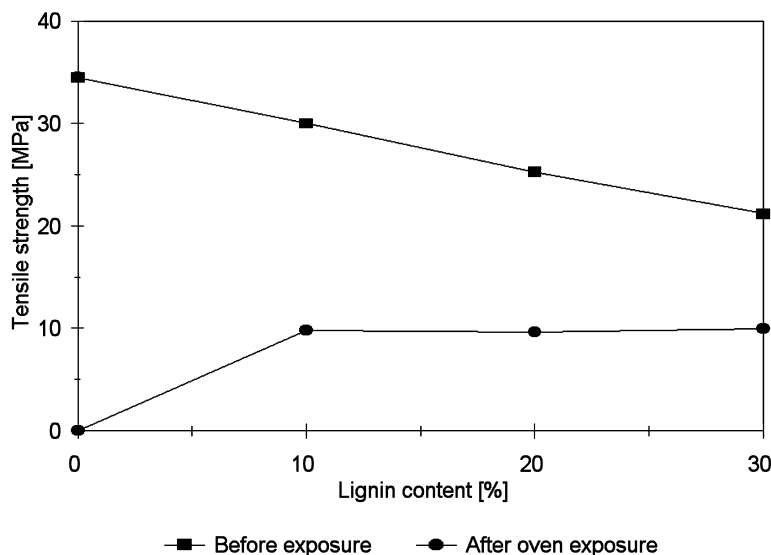


Fig. 11. Dependence of tensile strength of PP, before and after oven exposure, on lignin contents.

6). The tensile strength continuously decreasing with lignin for both polymer types.

### 3.3. Light stability and long-term heat stability of PP and PE–lignin blends

Chosen mixtures from Table 2 were tested for the light stability (LS) and the long-term heat stability (LTHS) determination of the effect of lignin concentration. The effect of lignin concentration on LS (measured as dependence of carbonyl groups increment) is shown in Fig. 7 for PP and Fig. 8 for PE blends.

According to the data showing in Figs. 7 and 8, the degradation process starts earlier for the pure polymer than for polymers with lignin blends. Based on these results it is suggested that lignin acts as a light stabiliser in both the polymers. The effect of UV-initiated degradation on mechanical properties of tested samples was also studied. Figs. 9 and 10 show the changes of tensile strength before and after 113 h QUV tester exposure of samples with various lignin contents. In contrast to the previous evaluation method (carbonyl groups increment), the influence of UV radiation on tensile strength is not the same in PP and PE. While for the PP blends UV radiation there is no effect on tensile strength of blends containing lignin up to 30 wt% content (Fig. 9), for PE some differences can be observed (Fig. 10). UV exposure has practically no influence on tensile strength up to 10 wt% lignin content. At higher levels of lignin concentration there is a significant decrease of tensile strength observed for samples that were UV exposed in comparison with samples without UV exposure.

Based on the LS results obtained it can be said that lignin acts, in PP, as a light stabiliser over a large concentration range and is able to protect the mechanical properties

after a short UV exposition of PP–lignin blends. On the contrary, in the PE–lignin blends, lignin is able to protect the mechanical properties after UV exposition at only a relatively low concentration level. From 20 wt% content, lignin causes, after short UV exposure, a decrease of tensile strength in spite of no effect on the carbonyl group increment.

Obtained results suggest differences in lignin influence on PP and PE matrix. These differences were observed in the LTHS evaluation too.

In Fig. 11 it is shown that the presence of lignin in PP causes differences between the tensile strength of non-exposed samples and samples after 500 h oven exposure, independent of the lignin concentration. The exposed samples have much lower values of tensile strength than non-exposed samples. If PE was used as polymer matrix (Fig. 12), the exposed samples have lower tensile strength only up to 10 wt% lignin content. From 20 wt% lignin content, exposed samples have higher tensile strength in comparison to non-exposed samples. Tensile strength increment, after exposure, increases with lignin content in the PE–lignin blend.

## 4. Conclusion

In earlier works [9] lignin was identified, for PP, as a stabiliser in general at low concentration and as an initiator of the degradation process of PP at higher lignin concentration. Results in this paper described and identified the type of degradation (processing, light or long-term heat) in which lignin acts as a stabiliser. Experimental work was extended to PE as well. Based on the results obtained it can be said that lignin acts, mainly in PP, as a good processing stabiliser and a light stabiliser. At long-term heat stress lignin acts as

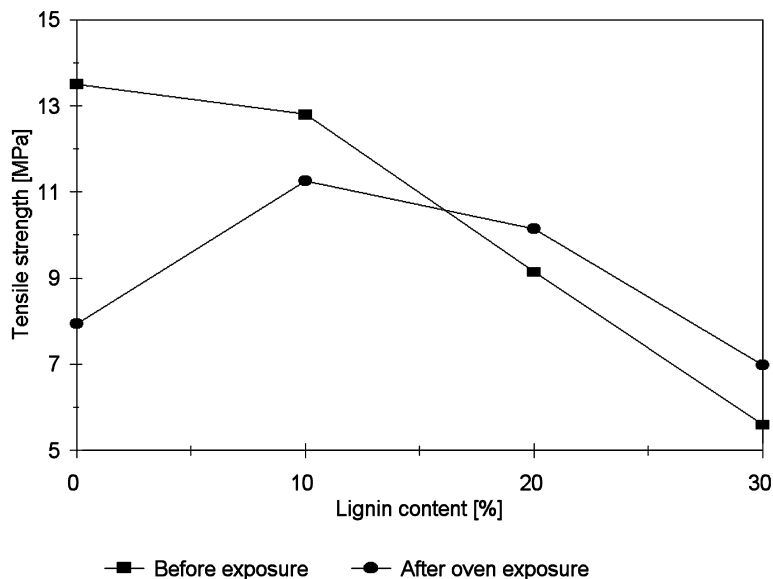


Fig. 12. Dependence of tensile strength of PE, before and after oven exposure, on lignin contents.

an initiator of the degradation process in PP, mainly at higher concentration levels. In the case of PE blends where lignin has practically no effect on the processing stability, it acts as an initiator of the degradation process after short UV light exposure of samples, if the lignin content is more than 10 wt%. At long-term heat stress, lignin in PE acts as a stabiliser and its effect is predominately at higher concentrations.

The influence of lignin on the mechanical properties, represented by tensile strength, was described. For both polymers (PP and PE), the mechanical properties decrease with increasing lignin content.

The MFI of PE–lignin blends increases in the entire tested lignin concentration range continuously. The MFI of PP–lignin blends is stabilised up to 20 wt% lignin content, and increases above this concentration.

Results obtained help to clarify the interaction between lignin and PE or PP matrix, mainly in the degradation process and show that the degradation process of pure lignin has an influence on the degradation of polymer matrix. From this point of view the results obtained show that the final degradation characteristics of polymer blends containing lignin as a filler can be modified by the polymer matrix

selection and the type of stress condition of the final product.

#### Acknowledgements

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