

Influence of matrix polymer on melt strength and viscosity of wood flour reinforced composites

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The paper presents results of our recent studies on the viscoelastic characteristics of thermoplastic composites filled with 30 wt.% of wood flour (WF), as a part of the foaming program. PP, HDPE and PVC were used as polymer matrices. Melt processability has been characterized by MFI tests in temperature of 180°C, 190°C and 200°C. It has been shown that the composites based on PP and PVC were easier flowing than HDPE in that range of temperature. Melt viscosity results suggested a thermal degradation of PVC. However, wood flour exhibited a stabilizing effect on PVC in PVC/WF composites. Melt strength tests have shown that the compatibilized PP/WF composite should be useful for foaming. Set of experiments with a chemical blowing agent has been performed in the single-screw extruder, which confirmed that supposition.

Key words: melt strength, wood flour, composites, blowing agent, foam extrusion.

1 INTRODUCTION

In recent years an increasing interest in foaming of polymer composites has been observed. Since extrusion is one of the most economical processes for production of plastics, the commercial utility of foamed polymer/wood flour composites would be further enhanced, if these could be produced using a continuous extrusion technology [1].

Polymers filled with wood flour (WF) exploit cellulose as a reinforcing filler for the polymer matrix. This component allows to obtain the highly filled, low cost materials. However, the composites of WF and polyolefins needs to be compatibilized in order to improve a normally poor adhesion between the hydrophobic polymer and a hydrophilic wood flour [2]. Such modification allows obtaining the high quality materials exhibiting good mechanical properties.

Natural component of wood flour is water which can act as a uncontrolled blowing agent. Therefore, filler drying is an important factor when preparing a composite. It excludes certain amount of water participating in foaming process [3].

Thermoplastic foams have a cellular structure generated by the expansion of gas or a chemical blowing agent, which usually is a solid compound [4,5]. Foamed composites possess unique properties like low density, moderate material cost and good mechanical properties [6].

In a course of foaming, at the last stage of a growth process, the cellular structure has a tendency to collapse [4, 6] During expansion of a gas bubble within the material, a cell wall can be destroyed by pressure. In general, creation of a cellular structure depends on several parameters (temperature, pressure, quantity of a blowing agent), while one of the most important is the melt strength, which strongly influences the polymer foamability.

Melt strength during stretching under uniaxial stress measures the force applied to the extruded material stream up to the break of a material. Measurements of this parameter are based on a principle of two rotating wheels, which draw down the polymer melt with a defined velocity and thereby produce an elongational deformation [7, 8]. Ability to evaluate this parameter depends on the rheological properties (viscosity), physical properties (glass temperature, melting point) and material structure (linear or three-dimensional chains).

Very important is a selection of the initial values of the velocity and acceleration conditions suitable to interpretation of the melt strength. The velocity of routing wheels should be comparable to the outflow velocity of a composite. The values of acceleration should be selected from a wide range of possible values for the particular material. Too high value of this parameter may cause a growth in the velocity and quick break of the melt stream. Low rates of acceleration cause an opposite effect [7].

Melt strength is strongly related to temperature,

therefore temperature is a critical factor for the viscosity of most polymers (including PP and HDPE), but for others (PVC) can induce the degradation process.

Melt strength depends also on pressure. Some investigations have shown that for polyolefins the tearing off force in the melt rises markedly when the pressure diminishes [7]. On a basis of the melt viscosity and melt strength at defined temperature and pressure, conditions of flow and melt extension of different materials can be compared, which may be useful for selection of a proper one for foaming. Measurements of the melt strength allow to predict the material behaviour during a cell generation and growth as a result of the gas bubble expansion in a composite.

2 MATERIALS AND METHODS

2.1 Materials

Polypropylene (Moplen HP500J) and high density polyethylene (Lupolen 3721c) were provided by Basell Orlen Polyolefins (Poland).

Poly(vinyl chloride) plasticized with 40% ethylhexyl phthalate was produced by Inco Veritas (Poland). Wood flour (Lignocel HB120) was provided by J. Rettenmaier & Sohne (Germany).

Compatibilizers used for an enhancement of the interfacial adhesion were polyolefins grafted with maleic anhydride (MA): PP-g-MA was provided by Aldrich Chemicals. HDPE-g-MA was produced at site by reactive extrusion.

Chemical blowing agent Hydrocerol CF 40E from Lifocolor Farbplast, Poland was used for foaming.

2.2 Manufacturing of composites

Composites of PP, HDPE or PVC with wood flour have been prepared using the internal mixer Rheomix 600 of HAAKE Rheocord 9000 system (Thermo, Germany). The content of wood flour was 30 wt.% for all composites. Mixing was performed with 60 rpm at 180°C for polyolefins and 170°C for poly(vinyl chloride). The mixing time amounted to 5 min. for PP and PVC or 8 min. for HDPE. Final loading of all components was equal to 75% of the mixing chamber volume. Roller blades rotors were used for mixing purposes.

Compatibilizers were added in an amount of 3 wt.% (PP-g-MA) or 50 wt.% (HDPE-g-MA).

2.3 MFI test

Melt flow index (MFI) tests were performed for all materials at three temperatures (180°C, 190°C and 200°C) at a loading of 5kg.

2.4 Viscoelastic measurement

Measurements of melt viscosity were performed with the rotational rheometer Haake Rotovisco RT20 at temperatures 180°C, 190°C and 200°C. The sensor system was plate-plate working in CR (controlled strain mode) conditions [9].

2.5 Melt strength test

For characterisation of the extensional flow properties the test was performed with Rheotens and the capillary rheometer Gottfert RheoTester 1000 (nozzle length 20 mm and nozzle diameter 1 mm).



Fig. 1. Melt strength test – Goettfert RheoTester 1000.

This measurement simulates extrusion and spinning process. Polymer melt is extruded through a die with a piston velocity of 0,1 cm/s and pulled out by two rotating wheels with initial acceleration of 1,2 cm/s² and the elongation speed of 0,1 cm/s (Figure 1).

2.6 Foaming process

The foaming process was performed with a single screw extruder Rheomex 252 in two stages. The first one was mixing of all components with the blowing agent. The temperature profile in a barrel allowed

only for its partial decomposition. This stage provided also an equal distribution of the blowing agent within the composite. The extrudate was finally pelletised.

At the second stage the material was extruded again and calibrated after expansion. In a meantime a cellular structure was formed in the extrudate. The temperature profile was high enough to allow a complete decomposition of the blowing agent.

3 RESULTS AND DISCUSSION

3.1 MFI tests

Table 1 exhibits an influence of the test temperature on the melt flow index.

Table 1. Influence of temperature on MFI

Composition	MFI, g/10 min		
	180°C	190°C	200°C
PP	3,92	5,50	6,48
PP/WF/comp	1,29	2,04	2,34
HDPE	0,51	0,64	0,78
HDPE/WF	0,24	0,33	0,44
HDPE/WF/comp	0,11	0,16	0,21
PVC	4,23	16,36	38,20
PVC/WF	1,10	6,00	11,04

On the basis of MFI results one can conclude that the composites based on PP and PVC are quite well flowing materials. In contrary a flow of HDPE and all composites with HDPE matrix is only marginal because of a temperature range, too low for HDPE.

3.2 Melt viscosity

Melt viscosity decreased for all composites with an increase of temperature. The lowest viscosity at 180°C has been reported for PP and PVC as well as for PVC/WF and PP/WF/comp. Results of the melt rheology have been presented in Table 2.

Table 2. Influence of temperature on melt viscosity at 1 rad/s

Composition	Melt viscosity, Pa s		
	180°C	190°C	200°C
PP	8962	6703	4907
PP/ WF/comp	30950	22250	14700
HDPE	18060	20550	24030
HDPE/WF	99560	59700	43420
HDPE/ WF/comp	79750	62450	47820
PVC	6044	59170	74110
PVC/WF	25970	21300	8635

The melt viscosity of PVC at 190°C and 200°C behaved differently to other materials. Viscosity increased for approximately ten times in comparison to that of 180°C, whereas for PP a decrease for ca. 50% has been observed. This is an evidence for a severe PVC degradation. However, melt viscosity of PVC/WF composites decreased with increasing temperature, thus suggesting a stabilizing effect of cellulose to poly(vinyl chloride) thermodegradation.

3.3 Melt strength

Influence of temperature on the melt strength of composites has been presented in Table 3.

Table 3. Influence of temperature on melt strength

Composition	Melt strength, N		
	180°C	190°C	200°C
PP	4,27	0,28	0,47
PP/WF/comp	5,00	2,48	1,72
HDPE	0,16	0,18	0,24
HDPE/WF	-	0,14	0,21
HDPE/WF/comp	0,22	0,42	0,15
PVC	0,16	0,22	-
PVC/WF	0,87	0,18	0,10

Results of the measurements have shown that the composite with PP matrix requires the highest force to breaking down the extrudate. This feature has been observed at all temperatures studied. The highest melt strength was measured for PP/WF/comp at 180°C (Figure 2). Other composites exhibited much lower values of the breaking force.

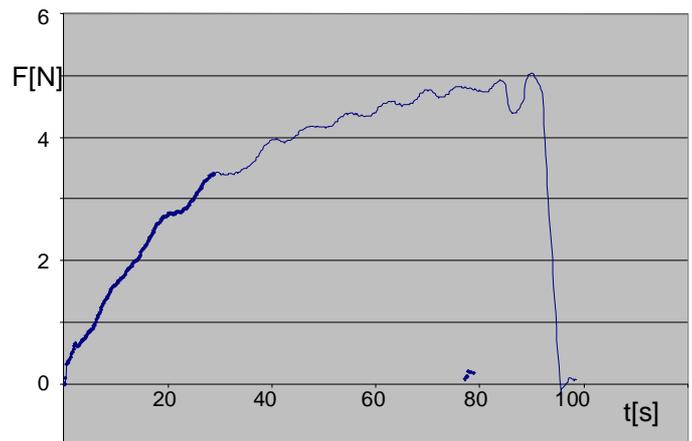


Fig. 2. Melt strength plot for PP/WF/comp at 180°C

With increasing temperature the melt strength decreased for PP and PVC composites. For HDPE composites very low breaking force was registered and only a little change in the melt strength was

observed. Basing on the melt strength results PP/WF/comp material has been selected for the foaming experiments.

3.4 Morphology of the cellular structure

For characterisation of the cell structure SEM pictures of the cold fracture surface have been made.

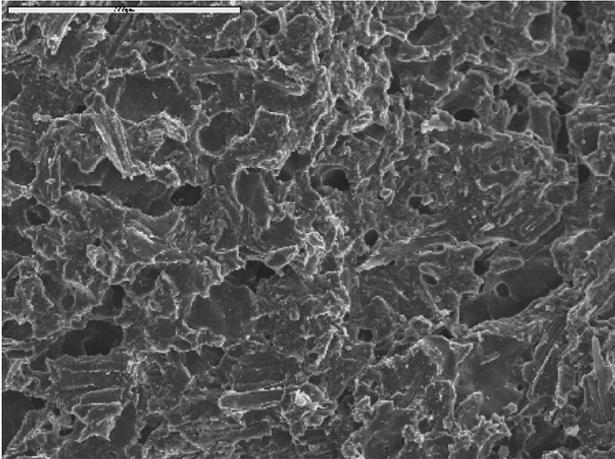


Fig. 3. Cell structure of PP/WF/comp composite

Cell morphology of PP/WF/comp composite foam has been presented on Figure 3. Large differences in the cell size and distribution have been observed.

4 CONCLUSIONS

- The highest melt strength was found at 180°C for PP/WF/comp material and this composite was selected for foaming.
- Unequal porous structure in cellular composites can result from a high content of a

filler, which induces heterogeneity in the material. Additional compatibilization between the components should be favorable.

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