PROPERTIES OF HIGH-DENSITY POLYETHYLENE REINFORCED WITH PINE-WOOD FILLERS

Wood-polymer composites based on high-density polyethylene and two types of pine-wood filler were investigated. It is shown that the addition of long wood fibres to common polyethylene leads to a substantial increase in stiffness, i.e. from 504 MPa for PE-HD up to 1526 MPa for PE-HD with 60% wood fibres. Moreover, morphological investigations revealed that superficial wood particles were sufficiently embedded in the base polymer ensuring protection against higher moisture uptake rates.

Keywords: polyethylene, pine wood, mechanical properties

Introduction

Wood-polymer composites (WPCs) are materials which have been on the market for many years, and are used in the construction industry, for example, for moulded components or deckig profiles [Błędzki et al. 2006; Zajchowski and Ryszkowska 2009; Oszust et al. 2011; Michalska-Pożoga and Czerwińska 2015]. For furniture application (plates and sheets), thermosetting materials, such as phenol formaldehyde resin, are usually used as a polymeric matrix. Thermoplastic polymers such as polypropylene, polyethylene or polyvinyl chloride are the most common, are characterized by good commercial properties and may be shaped by conventional processing methods such as injection moulding or extrusion [Gozdecki et al. 2011; 2012; 2015; Kociszewski et al. 2012] at moderate temperatures not exceeding 210°C. In comparison with synthetic commodity polymers, wood is a cheaper, stiffer and stronger material, which makes it a good candidate for use as a polymer filler or for reinforcement. The properties of a wood-polymer composite depend on the type of matrix and filler. In the case of a reinforcing agent, not only is the volume fraction important, but also the size, shape and surface area of the particles, as well as the compatibilization method [Bula and Jesionowski 2010; Bula et al. 2015; Knitter and Dobrzyńska-Mizera 2014]. The filler used to prepare WPC can be of any
origin, and have different forms: splinters, chips and flour. It can be obtained from conifers (pine or spruce) and much less frequently from deciduous trees (oak or maple) [Stark and Rowlands 2003; Liber-Kneć et al. 2006; Migneault et al. 2008]. The properties of the composite are formed at the stage of processing where the material in a molten state is characterized by specific rheological properties which affect the accurate shape of the product. The selection of processing tools determines the level of distribution of the wood filler in the polymer matrix as well as its orientation, which in turn impacts the heat dissipation rate of the moulded product.

The main purpose of this study was to compare the reinforcing effect of two kinds of pine-wood particles, differing in particle length, on the mechanical properties of high-density polyethylene.

 Materials and methods/Research methodology

The high-density polyethylene (PE-HD) used in this study was Hostalen GD 7255 (obtained from Basell Orlen Polyolefin, Poland) with a density of 0.955 g/cm³ and a melt flow rate of 4 g/10 min (190°C/2.16 kg). Ethylene-propylene copolymer grafted maleic anhydride (PE-g-MAH) Fusabond P353 (DuPont) with a density of 0.904 g/cm³, a melt flow rate of 470 g/10 min (190°C/2.16 kg), and a grafting degree of 1.4% wt was used as a coupling agent.

Two kinds of pine-wood particles were used as fillers: Lignocel C120 short wood fillers (SWF), and Lignocel C300 long wood fillers (LWF), (grades with a mean fibre length of 120 and 300 µm, for C120 and C300, respectively). The fillers from softwood, were supplied by J. Rettenmaier & Söhne GmbH, Rosenberg, Germany.

The WPCs were produced in a two-stage process. In the first stage, both wood particles were compounded into pellets at 20%, 40%, and 60% by weight with the PE-HD using a single-screw extruder. The barrel temperatures of the four zones ranged from 180°C to 200°C from the feeding to the die zone. The screw speed was 60 rpm. Following this, the pure PE-HD and composites were injection moulded into dumbbell-shaped specimens using an ENGEL 80/25 HL injection moulding machine, with 20 tons of clamping force. The specimens obtained were 4 mm thick and 10 mm wide. All the dimensions were in agreement with the ISO 527-2 standard. The compositions of the samples, including the percentage contribution of all the materials used, are listed in table 1.

The post-processing shrinkage was measured at room temperature using specimens with an initial width of 10 mm and 150 mm length, after cooling. The change in the linear dimensions of the injection moulded part $S_l = ((l_0 - l)/l_0) \cdot 100\%$ and width $S_w = ((w_0 - w)/w_0) \cdot 100\%$ were calculated; where $l_0$ and $w_0$ are the length
Table 1. Composite formulations

<table>
<thead>
<tr>
<th>Material code</th>
<th>Type of wood particles</th>
<th>Wood content [%wt]</th>
<th>PE-HD content [%wt]</th>
<th>PE-g-MAH content [%wt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PE-HD</td>
<td>–</td>
<td>–</td>
<td>100</td>
<td>–</td>
</tr>
<tr>
<td>PE/20SWF</td>
<td>small wood fillers</td>
<td>20</td>
<td>76</td>
<td>4</td>
</tr>
<tr>
<td>PE/40SWF</td>
<td>small wood fillers</td>
<td>40</td>
<td>57</td>
<td>3</td>
</tr>
<tr>
<td>PE/20LWF</td>
<td>long wood fillers</td>
<td>20</td>
<td>76</td>
<td>4</td>
</tr>
<tr>
<td>PE/40LWF</td>
<td>long wood fillers</td>
<td>40</td>
<td>57</td>
<td>3</td>
</tr>
<tr>
<td>PE/60LWF</td>
<td>long wood fillers</td>
<td>60</td>
<td>38</td>
<td>2</td>
</tr>
</tbody>
</table>

and width of the injection mould cavity, and \(l\) and \(w\) are the length and width of the specimen after cooling. The melt flow rate (MFR) was determined in a plastometer (model MP-IIRT-M, Russia) according to ISO 1133. The test conditions were set at a load of 2.16 kg and a temperature of 190°C for all the samples. Tensile tests were carried out using an Instron universal testing machine (model 4481, Canton, USA) at room temperature using a crosshead speed of 10 mm/min. The tensile tests were performed as per ISO 527-2. Brinell hardness tests were carried out using an HPK 8206 Brinell tester as per the standard PN-EN ISO 2039-1:2004P. During the tests, a ball indenter with a diameter of 5 mm and a test force of 49 N was used. The investigations of sample surface morphology were carried out and microphotographs were taken using a reflected-light microscope. The morphology was examined using a Nikon Eclipse E300 microscope.

Results and discussion

Processability properties

The melt flow rate and thermal shrinkage are presented in figures 1 and 2. The melt flow rate is inversely related to the sample viscosity and can be used to estimate the interaction between phases in the polymer composites. Figure 1 shows that the unfilled PE-HD had the highest MFR of all the tested materials. The incorporation of lignocellulose fillers led to a gradual decrease in the melt flow rate. This indicates that during injection moulding a very low flow length value could be expected, or higher injection pressure should be applied for thin-walled parts.

In both cases, the dimensional stability, evidenced by longitudinal and transverse shrinkage, decreased substantially with the pine-wood filler content compared to the unfilled PE-HD. The above findings provide important data concerning the dimensional correction of tools for the processing of these composites in comparison to the ones used for pure PE-HD.
Fig. 1. MFR values for PE-HD and its composites

Fig. 2. Effect of filler content on shrinkage of PE-HD and its composites

Mechanical properties

The selected tensile and hardness properties of the pure matrix and its composites, obtained from the static tensile and Brinell hardness tests, are
presented in figure 3. For both wooden fillers, the Young’s modulus and tensile strength increased according to the composition, while the elongation at break decreased. The noteworthy improvement in stiffness was probably due to the high volume ratio of pine-wood particles in the polyethylene matrix, as well as in the filler anisotropy, especially for the LWF particles [Zhang et al. 2008; Kaseema et al. 2015]. On the other hand, a drastic limitation in the elongation values for the composites, in comparison to the unfilled PE-HD, is attributed to poor adhesion between the matrix and fillers, and the creation of small voids around the rigid filler particles. The large number of inclusions in the PE-HD matrix were potential nucleation sites, which contributed to a decrease in the plastic deformation of the composites.

Fig. 3. Effect of filler on the Young’s modulus (a), Brinell hardness (b), tensile strength (c), and elongation at break (d) of PE-HD and its composites

**Surface morphology**

Figures 4a and b present the surface morphology of the sample filled with 40 wt.% of pine wood. The pine-wood particles, covered with the polymer layer, can be clearly seen. Therefore, the lignocellulose filler is probably protected
against moisture and little or no water absorption should take place during exposure to the weather.

Fig. 4. View of the PE/40LWF composite surface, magnification 40×

Conclusions

The application of pine-wood particles as a reinforcement for PE-HD, especially with a filler content of 40 wt.% or more, markedly reduced the flow ability of the PE-HD/pine-wood composites. This means that highly filled PE-HD composites should be used exclusively for thick walled products, such as hollow and siding profiles or garden furniture. Moreover, the composites revealed superior stiffness and markedly improved tensile strength, as well as hardness. Hence, these composites may be used as engineering materials with the potential to replace wood products. This is even more reliable, if one takes into account that the pine-wood particles in the composites were covered by a hydrophobic matrix. Therefore, no additional protection coating is needed for these materials.

References

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List of standards

ISO 1133 [2011] Plastics – Determination of the melt mass-flow rate (MFR) and the melt volume-flow rate (MVR) of thermoplastics


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