Composites: Part A 74 (2015) 107–113

Contents lists available at ScienceDirect

Composites: Part A

journal homepage: www.elsevier.com/locate/compositesa

# The influence of paper type on the properties of structural paper – Polypropylene composites

Martina Prambauer [a,b,*], Christian Paulik [b], Christoph Burgstaller [a]

[a] Transfercenter für Kunststofftechnik GmbH, Franz-Fritsch Strasse 11, A-4600 Wels, Austria
[b] Institute for Chemical Technology of Organic Materials, Altenberger Strasse 69, Johannes Kepler University, A-4040 Linz, Austria

ARTICLE INFO

*Article history:*
Received 17 December 2014
Received in revised form 3 April 2015
Accepted 4 April 2015
Available online 9 April 2015

*Keywords:*
A. Polymer-matrix composites (PMCs)
A. Laminates
Natural fibers composites
B. Mechanical properties

ABSTRACT

In this work, different types of paper were used for producing cellulose reinforced polypropylene composites with MAPP as a coupling agent. The samples were prepared by a film stacking method of the polymer and paper layers, followed by hot pressing in order to fabricate structural composite laminates. Virgin copy, filter and newspaper were used as reinforcements for producing composites with varying paper content. The influence of paper type and content on the mechanical and physical properties of the laminates was investigated and discussed in detail. Remarkable results for tensile and bending test were obtained for copy and newspaper composites at a paper content of 30 and 40 vol.%, indicating a high potential for constructive and industrial applications of the laminates. Further characterization was carried out by Charpy impact test, water uptake study, TGA and light microscopic analysis. To summarize the characterization, structural paper reinforced composites with attractive properties were obtained.



## 1. Introduction

Fiber reinforced polymers are the materials of choice for numerous applications e.g. in the fields of house furnishing, construction, car, aerospace, sports, packaging and leisure industry. Their mechanical properties are very often superior to the plain polymers and further benefits, such as weight reduction, make them a versatile material for engineering aspects. Well known reinforcements are glass, carbon and aramid fibers, which are combined with thermoplastic or thermoset matrices [1,2]. However, in the past few decades polymers, reinforced with natural fibers have been the subject of multiple research projects, due to several advantages over conventional fibers. Natural fibers are renewable materials, which are low in price and density. Depending on the source, the mechanical properties are high enough to compete with synthetic reinforcement materials, such as glass fibers. Natural fibers have a wide distribution and are environmentally friendly materials meaning that they are biodegradable, they are renewable and $CO_2$ neutral materials. In addition, natural fibers are non-abrasive to processing machinery and represent no health risk through inhalation, as would be the case for glass fibers [3,4]. The source of natural fibers depends on local availability, but notably are hemp, flax, sisal, abaca and wood particles. A comparison of the mechanical properties of selected natural fibers to those of E-glass is given in Table 1, adapted from literature [3–8]. Especially in the automotive and aerospace industry, where weight reduction is an important issue, natural fibers have already been used as construction materials. But also applications of wood plastic composites, in particular, are found in floor panels, under-body parts or claddings [8,9].

However, there are also some problematic aspects considering natural fiber reinforced polymers. The wettability of the highly hydrophilic fibers by the matrix, which is very often of a rather hydrophobic nature, is restricted and results in poor adhesion at the fiber–polymer interface. Reduced mechanical properties and increased water uptake are the consequences of this incompatibility. In order to improve these shortcomings, either physical (plasma treatment, corona treatment) [10] or chemical (organosilanes, isocyanides, sodium hydroxide, permanganate and peroxide) modifications are applied to the fibers. Furthermore compatibilizers, such as maleic anhydride grafted polymers, are often used as well [2,11].

The properties of naturally grown fibers depend on several environmental parameters, such as cultivation locality, climatic influence and harvesting time. Therefore significant variations of the fiber quality are obtained even within the same plant species [7]. This leads to difficulties for applying natural fibers, as the mechanical properties of the reinforced polymer strongly depend

* Corresponding author at: Transfercenter für Kunststofftechnik GmbH, Franz-Fritsch Strasse 11, A-4600 Wels, Austria. Tel.: +43 7242 2088 1034.
*E-mail address:* martina.prambauer@tckt.at (M. Prambauer).

http://dx.doi.org/10.1016/j.compositesa.2015.04.004
1359-835X/

**Table 1**
Comparison of tensile properties of E-glass fibers to selected natural fibers [3–8].

| Fiber | Density (g/cm$^3$) | Tensile strength (MPa) | Elastic modulus (GPa) | Elongation at failure (%) |
|---|---|---|---|---|
| E-glass | 2.55 | 2000–3500 | 70 | 3 |
| Hemp | 1.48 | 550–900 | 70 | 2 |
| Flax | 1.40 | 800–1500 | 27 | 1.2–1.6 |
| Sisal | 1.33 | 600–700 | 38 | 3 |
| Jute | 1.30 | 400–700 | 26 | 1.5–1.8 |
| Abaca | 1.50 | 400 | 12 | 3–10 |
| Kenaf | 1.45 | 930 | 53 | 1.6 |

on the fiber's characteristics and a steady quality of the composite materials cannot be guaranteed. Furthermore, processing such fibers in plastics processing machinery is also complicated due to their structure and bulk density [12]. In order to circumvent these problems, the approach of using pulp or paper fibers for reinforcing thermoplastic matrices has been discussed in literature for several times [9,13–17]. The advantage of using these kinds of processed fibers is the high consistency concerning quality, fiber length, source and the mechanical properties, ensured by the restrictive standards of the paper industry. As paper is a bulk product, it is also low in price and readily available [9].

Most attempts, which are cited in literature to incorporate paper into thermoplastics, were carried out by grinding the paper to small particles and mix them with the polymer in melt. However this process reduces the fiber length of the cellulose fibers resulting in a decrease of mechanical properties. In order to avoid this effect, whole paper sheets have been used to produce structural polypropylene–paper composites for this work. The aim of the research was to create structural laminates by a hand stacking method, at which paper sheets and polymer layers are stacked up and subsequently combined by hot pressing. The laminates were characterized to get deeper knowledge about the influences of the formulation on the laminate's properties. The amount and type of paper in the composites were varied and the laminates where characterized by means of mechanical and physical testing.

## 2. Materials and methods

### 2.1. Material

For all fabricated composites, the same polymer, polypropylene (PP) HD 120 MO with a density of 905 kg/m3 and a MFR 8 g/10 min at 230 °C was used. The polymer was extruded in-house by a flat film extrusion line (P/M Plastic Maschinenbau, 35 mm screw diameter, $L/D$ ratio of 18) with a maximum barrel temperature of 250 °C and a screw speed of 80 rpm at throughput of 15 kg/h into films with 140 and 250 μm thickness and a width of approximate 18 cm. In order to improve the interaction between the hydrophobic polymer and the hydrophilic lignocellulose components of the paper, a coupling agent was used for all the composites. For this purpose, 5 wt% of maleic anhydride grafted polypropylene (MAPP, Scona TPPP 8112 GA) were added to the polymer granulate as dry blend before extrusion. Three different paper types were used as reinforcement. For paper type number one, which is termed as copy paper (CP) in the following, Xerox Transit copy paper with a mass per unit area (grammage) of 80 g/m$^2$ and a thickness of 98 μm was used. The reinforcement of the second laminate type was made of Whatman filter paper, type 4, with a grammage of 84 g/m$^2$ and a thickness of 200 μm and is referred to as filter paper (FP). The filter paper was chosen due to its porous structure, which is assumed to improve the embedding of the single cellulose fibers into the polymer matrix, which could enhance the mechanical properties. The third paper type was plain newspaper (NP) with a grammage of 45 g/m$^2$ and a thickness of 50 μm, obtained by Creative Company, Germany. The newspaper was compared to the other paper types with regard of a possible application for post used waste paper as polymer reinforcement. An overview of the applied paper types is given in Table 2.

**Table 2**
Overview of the properties of the used papers.

| Paper type | Grammage (g/m$^2$) | Thickness (μm) | Tensile strength (MPa) | Elastic modulus (MPa) | Source |
|---|---|---|---|---|---|
| CP | 80 | 98 | 62 | 5468 | Xerox Transit |
| FP | 84 | 200 | 8 | 974 | Whatman 4 |
| NP | 45 | 50 | 43 | 4774 | Creative Company |

### 2.2. Methods

For evaluating the reinforcing potential of the different paper types, a tensile test of the single paper sheets was carried out. For this purpose a Zwick–Roell Z0,5 universal testing machine, according to ISO 527-2, was used. For every paper type, 8 samples of the 1BA type were used. The elastic modulus was determined with a crosshead speed of 1 mm/min, afterward a speed of 5 mm/min was applied for the rest of the measurement until paper rupture. From the obtained results, a Weibull distribution graph was modeled from the tensile strength values. Eq. (1) shows the basic formula of a two parameter Weibull distribution with the scale parameter $\alpha$ and the shape parameter $\beta$ [18]. The distribution function $F(x)$ can be transformed to match the linear function $y = kx + d$, as shown in Eqs. (2) and (3). In order to obtain the parameters, $\ln x$ of (3) is plotted against the $y$-value of the function. This leads to the shape parameter $\beta$, which is directly obtained by the slope of the line, while the parameter $\alpha$ can be calculated with the point of intersection of the $y$-axis.

$$F(x) = 1 - e^{-\left(\frac{x}{\alpha}\right)^{\beta}} \tag{1}$$

$$\ln\left(\frac{1}{1-F(x)}\right) = \left(\frac{x}{\alpha}\right)^{\beta} \tag{2}$$

$$\ln\left[\ln\left(\frac{1}{1-F(x)}\right)\right] = \beta \ln x - \beta \ln \alpha \tag{3}$$

All composites were produced by stacking polymer layers and paper sheets inside a pre-heated steel mold (cavity area of 10 × 10 cm$^2$) until a height of 3.2 mm was reached (Fig. 1). For composites with low paper content, polymer films with 250 μm were used. For higher paper contents or very thin papers, thinner polymer films with 140 μm or 100 μm, were used for the stacking process. The fiber content of each sample was calculated according to Eq. (4), where $F_C$ is the fiber content, $N_P$ is the number of paper layers, $d_P$ is the paper's thickness and $d_C$ is the non-compacted composite's thickness (3.2 mm).

$$F_C = \frac{N_P * d_P}{d_C} * 100\% \tag{4}$$

By the stacking process, composites with approximately 10, 20, 30, 40 and 50 vol.% fiber content were produced. The exact fiber content did not vary more than 1 vol.% from these values. However, it has to be kept in mind that during the molding process the paper layers get compacted and the fiber volume fraction in the composite is slightly reduced. Therefore, the values for the fiber volume refer to the non-compacted composite.

A spacer frame was inserted for adjusting the laminate's final thickness to 3 mm before the mold was closed. The filled mold was placed into a hot press from Wickert WLP 80/4/3 and a