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A comparison of processing and performance for lignocellulosic reinforced polypropylene for injection moulding applications ☆

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ABSTRACT

Using various reinforcements to tailor plastics properties to specific needs is a widespread approach in the plastics industry. Due to ecological considerations, natural reinforcements like wood particles or natural fibres have been investigated due to their reinforcing potential, but are usually not discussed in terms of processability. Therefore, the aim of this work is to compare a selected processing pathway for lignocellulosic reinforcements in terms of processability and composite properties. Composites were produced via compounding on a co-rotating twin screw extruder and injection moulding. Materials properties like tensile strength and modulus were assessed, as well as the processing specific properties were recorded. We found, that wood particles show a very good ratio between processability and properties, while sisal fibres outperform the other reinforcements in terms of mechanical properties due to their fibrous shape, but also using reinforcements like milled rice husks in composites can be feasible, especially when wood is not a locally available resource.

1. Introduction and aim of the work

Using reinforcements in thermoplastics is a very widespread approach in plastic industry. Such materials can be glass fibres, calcium carbonate or talc, and can improve the mechanical performance as well as the cost of the material, so one gets tailor made materials. While glass fibres are used if high mechanical performance is needed, talc and calcium carbonate are often used if only the elastic modulus is the target value. Examples in application of reinforced materials can be found nearly everywhere in our everyday lives, like casings for electrical appliances, various automotive parts, pipes and technical profiles, but also in packaging application, where for example plastic pallets are reinforced to withstand mechanical loads.

Over the last decades, due to economic as well as ecological considerations, also lignocellulosic reinforcements got some attention from industry as well as from university researchers. In economic terms, the typical filler approach, i.e. using a material which is much cheaper than the base resin to extend the latter, was the starting point for this development several decades ago. Such fillers were wood sawdust or particles, which are dealt with by several researchers [1–4], and found their main application in extruded profiles, which are used mainly for semi load-carrying applications, like decking or siding.

The ecological approach, which is still more widespread in academia, is to substitute glass fibres with their natural counterparts. Wambua et al. [5] were reporting on that issue some years ago, and they come up with the question if natural fibres can be a feasible alternative to glass. They found, that there are some properties, like coming from a renewable source, CO_2 neutrality and easier disposal, which can be held in favour of natural fibres. Also composite density is sometimes accounted for natural fibre use, especially because the raw fibres have high specific properties [6]. Typically, at least some part of this density advantage is sacrificed when natural reinforcements are processed via thermoplastic processing routes like injection moulding or extrusion, because these need to apply pressures typically in the range between 100 and 1000 bar due to the viscosity of the thermoplastics. With this, the cells of cellulose are collapsing to some extent, and density is increased, sometimes up to the maximum of the cell wall density, which is about 1.5 g/cm^3 [7]. Also other problems can be associated with the use of natural reinforcements. Due to their chemical nature, cellulose reinforcements provide a number of functional groups, mainly hydroxyl, at their surface. In combination with the typical polymer matrices, i.e. polyolefins, the interfacial interaction is weak, therefore the mechanical properties are limited. The typical approach to overcome this is to use compatibilizers, typically maleic anhydride grafted polyolefins, which is described

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extensively in literature from various researchers [1–2,8], and it is also well known in the wood plastic composites industry. Further, also the coefficient of thermal expansion and its effect is discussed, which influences the interfacial properties and therefore reduces the composite performance [9].

While in academia the gaining of knowledge about more ecological fibre alternatives is further followed, industrial interest is more lying on the use of low cost reinforcements, to somehow attenuate the effects of high crude oil and therefore resin prices. Due to the use of wood not only for construction, but also for other industrial pathways, like the production of pulp or regenerated cellulose fibres, as well as producing energy, these materials, even the residues, became more expensive over time. Therefore, a strong interest in using other natural reinforcements is still persisting.

Such materials, which are possibly suitable for reinforcing thermoplastics can be, asides wood flour, plant fibres like sisal, flax or hemp or agricultural by-products as rice husks or straw. To classify such materials like natural fibres, one can apply the biological structures, which decide between fibres originating from stems and leaves, for example [10], but in the authors' opinion these classification is not the best for thinking of reinforcements. Therefore, to classify the materials in a more processing and industry related approach, for this work we will use a simplified classification for cellulose based reinforcements, given in the following:

- Agricultural residues (of undefined form), such as rice husks or flax shives.
- Particulate reinforcements, such as wood particles.
- Natural fibres, e.g. sisal, hemp or flax.
- Semi man-made cellulose fibres, i.e. fibres which grow that way, but have to be refined to be useable as fibres, e.g. kraft pulp fibres.
- Man-made cellulose fibres, i.e. regenerated fibres like Lyocell® or Cordenka®.

The cellulose based reinforcements here are listed in ascending order of aspect ratio from particulate to fibrous, i.e. such materials, which have a macroscopically more powder like appearance, over such which are appearing as particles to the ones which can be clearly seen are fibres. These are again ordered from the coarser ones, sisal or flax fibres to the finer ones, like the Lyocell®. Further, with increasing aspect ratio there is a tendency to increased cost of the materials, simply explained by the fact that the effort to yield such materials is higher for the pulped and regenerated fibres than for the sawdust for example.

While there are several examples of publications dealing with the final composite properties of such cellulose materials in different thermoplastic matrices [11–14], the issues of processing are not addressed very often in literature. This has some reasons; in academia, different alternative processing routes, like film casting from solution, are applied besides typical thermoplastic processing. Such methods cannot be transferred in a cost effective way to industry in comparison with melt processing, especially in the case for polyolefins. The reasons for that are manifold. First of all, solvents to be used for solvent casting are expensive, to some extent harmful, and cannot be fully regenerated. In addition, polyolefins need elevated temperatures to be solved, as well as a high excess of the solvent. Secondly, the effort in terms of the necessary safety equipment of such a production site is much higher, because explosion safe equipment and venting systems have to be used. Such systems are not part of the standard installations in plastics processing facilities, because when melt processing granules to semi-finished parts, explosive atmospheres are of no concern.

Further, an optimized throughput in typical thermoplastic processing routes such as extrusions is not the main parameter we are interested in academia. The idea behind using such routes is to yield a material to investigate for various properties. Therefore materials will be run with any setup or parameter adjustments necessary, because the goal is to get some material for further investigations. To reach said goal, also modifications to the standard machine setup are quite common, to yield the possibility of using the material of interest in composite production. Such modifications can be some manual interaction, because for the typically small batches required, the effort to manually feed a material into the intake of an extruder to produce a compound is justifiable. It is to mention here, that this kind of approach is definitely necessary to gain new knowledge about materials, so to build the basis for industrial applications of a material. Such approaches are typically not undertaken in industry. The reason for this is, that profitable production is achieved via throughput and stable, carefree processing, also at much bigger machines, where due to the higher throughput any reject production is of much higher volume and therefore value than in academia. While the throughput may be the main parameter at a first glance, the processing stability is even more important, because every distortion needs some human attention and interaction, which is costly and is also another source of failures, which can influence the productivity. The last point here is the alteration of setups to self-to-develop specialty solutions, which is very unlikely in industry. The reasons for this are, that such developments take up production time without earning money due to actual production, and that the requirements for work place safety are very strict, which will result in a high effort to certify such self-built machinery, especially for a company producing compounds or semi-finished products.

Therefore, the aim of this work is to compare a selected processing pathway, namely extrusion compounding and injection moulding, for lignocellulosic reinforcements in terms of processability and composite properties. Correlating the outcome of processing and testing should give us a better picture about the suitability of a material for processing via the selected route with the focus on the compounding step.

2. Materials and methods

As reinforcements, rice husks, wood flour and sisal fibres were chosen, so one representative from every group, i.e. particulate reinforcement, natural fibres and agricultural by-products, is within the scope of this study. The decision not to include (semi-)man-made cellulose fibres like pulp or Lyocell® was due to the fact that these are typically not considered for the same applications as wood plastic composites due to the higher materials cost. Further, due to the fibres in these materials being single, elementary fibres, these are also extremely hard to feed into extruders without any further, laborious pre-processing, which make them less likely for application in such thermoplastic composites. Wood particles were softwood grade Arbocel C320 from J. Rettenmaier and Sons, Germany. Rice husks were a non-commercial material from India, which were milled in an air jet mill to yield particles with an increased bulk density, therefore suitable for feeding into an extruder. Sisal fibres were supplied as a threaded yarn with 2.6 g/m (*Seilerei Sammt*, Germany), which were cut to 6 mm long fibres by means of a self-built fibre guillotine cutter.

As the matrix, a general purpose polypropylene homopolymer (HD120MO, supplied by *Borealis*, Austria) with a melt flow rate of 8 g/10 min at 230 °C and 2.16 kg piston weight was used. This grade was chosen due to its universal applicability for different processing routes, e.g. injection moulding and extrusion. To ensure reasonable interaction between the non-polar matrix and the polar reinforcements, a maleic anhydride grafted polypropylene (Exxelor PO1020, *ExxonMobile Chemicals*, USA) with 0.9 wt% of maleic anhydride and a melt flow rate of 150 g/10 min at 190 °C and 2.16 kg piston weight. The compatibilizer was added in a share of 1/10 of