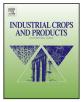
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Environmental impact assessment of end of life options for flax-MAPP composites



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ABSTRACT

Flax fibre composites are now widely used in various industries, such as automotive, in replacement of less sustainable materials. One of the remaining challenges is to define the most environment-friendly endof-life options for the flax fibre reinforced plastics based on an environmental evaluation and comparison of the available techniques. In this work, three end-of-life possibilities for flax fibre reinforced thermoplastics were investigated: chemical recycling, mechanical recycling and incineration. It was found that the chemical recycling technique is feasible as their properties do not change compared to the initial composite. However, its processing time, chemicals needed and equipment have negative effects on the environment. The second method, the mechanically recycled composite with discontinuous short fibres (flakes) leads to a decrease in properties of about 75% compared to the initial cross-ply composite. When this recycled material is compared to random mat composites (also short and randomly oriented fibres), the properties also decrease by 6–46%. The main advantage of the mechanical recycling technique is the speed of the process as very large quantities of waste can be shredded and processed into new components while reducing the environmental burden of producing fresh MAPP and flax fibre. The last EOL method studied was incineration with energy recovery and it has been found to be a good alternative as well since all the material can be fully combusted and embodies a relatively high calorific value.

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1. Introduction

Lately, the European Union's End of Life Vehicles Directive has stated that all vehicle constructors have to assure the re-usability and/or recyclability of the materials of new cars put on the market to a minimum of 85% of their total weight and the reusability and/or recoverability up to 95% of the total weight. Various materials can be used to construct a car such as steel, aluminium, plastics and composites. As for composites, mostly carbon and glass fibre reinforced composites are used, but a shift towards natural fibre based composites has been observed. This is thanks to their good mechanical properties, low density and lower environmental impact. The other motivations for the implementation of flax composites in automotive applications are driven by the fact that lightweight parts can be produced with competitive prices and reduced emission into the environment. This leads to a lower fuel consumption and recyclability of the materials which are combined to thermo-

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http://dx.doi.org/10.1016/j.indcrop.2016.09.006 0926-6690/© 2016 Published by Elsevier B.V. plastic polymers such as PP which could reduce the waste disposal problem (Bourmaud et al., 2013; Bourmaud and Baley, 2007; Kulma et al., 2015; Martin et al., 2016). PP is low cost and mechanically performant polymer that is widely used in automotive parts manufacturing. Thus, the combination of PP to flax fibres provides a recyclable composite materials with interesting properties while being green and sustainable.

Whereas the recycling of steel is well documented and researched, the recycling routes possible for composite materials and especially natural fibre composites, are much less investigated. Recycling is not merely a single process but can be seen as a chain of processes, starting with the collection of sufficient materials that has reached their end-of-life (EOL). The advantage of recycling is that the new material can be repurposed which will save energy and resources. However, recycling of composite will yield lower quality materials (Yang et al., 2012). This concept is called down-cycling. An important technological parameter affecting the EOL options for composites is the matrix material which could either be thermoset or thermoplastic. The important difference, from an EOL point of view, is the possibility to re-melt the thermoplastic polymers, whereas the thermoset polymers can be burned off to

Non-renewable energy requirements of fibres.

Fibre production type	Non-renewable Energy Requirements	Source
Flax (Hackled)	11,7 GJ/ton fibres	(Le Duigou et al., 2011)
Flax fibre mat	9,55 GJ/ton fibres	(Joshi et al., 2004)
Flax fibres	12,4 GJ/ton fibres	(González-García et al., 2010)
Glass fibre mat	54,7 GJ/ton fibres	(Joshi et al., 2004)
Glass fibres	45 GJ/ton fibres	(Le Duigou et al., 2011)

produce energy in incinerators or be digested to recuperate the reinforcing fibres. The use of thermoplastic composites is a rather recent and as such, their recycling is not yet a subject of active research. As of today, there is three main recycling methods that can be used for thermoplastic composites.

Mechanical recycling is a feasible technique for natural fibre composites and it is currently being researched in some institutes (Srebrenkoska et al., 2008). However mechanical recycling will always yield short fibres as output material. The mechanical recycling consists of cutting and crushing the composite into small pellets. These pellets can later be used as feedstock for injection moulding processes. This is considered as a downcycling of the material as the properties are decreased due to the reduced initial fibre length (Otheguy et al., 2009a). The recycled feed can also be mixed with virgin matrix material before being fed into an extruder in order to create a different fibre content in the part. A study on the mechanical recycling of boats shows that the recycled materials have good mechanical properties. However, these properties are still lower than those of the virgin material (Otheguy et al., 2009b). Furthermore, it was found that when recycling short pine fibre reinforced PP composites the Young's modulus and tensile strength decreased by only 13% and 17% respectively after eight recycling cycles. The major contribution to the decrease in properties is the breaking of the fibre into smaller pieces, and the damage introduced on the fibres (Beg and Pickering, 2007). In this study, the fibre breakage of natural fibres was evaluated. It was found that the fibres broke up into smaller fragments reducing their length but at the same time the fibre diameter was reduced, hence keeping the aspect ratio higher as expected. Because of this effect the aspect ratio only decreased by a factor of 6. The length reduction still remained higher than the diameter reduction (Morán et al., 2007).

The second method is chemical recycling, which involves attacking of the matrix with a solvent. Up to now, little work has been done on the chemical recycling of natural fibre composites with thermoplastic matrix. However, if a chemical treatment would dissolve the matrix without damaging the fibres or reducing the properties of the fibres, this could be a possible recycling method. Some work has been done on the acid attack of sulphuric acid on Carbon/PEEK composites in order to degrade the matrix and recuperate the fibres (Ramakrishna et al., 1997). The recycled material was then reprocessed into Carbon/PEEK composite with fresh thermoplastic. Its properties where compared with those of the virgin composite and it was found that the flexural modulus of the recycled material only reached 20% of that of the virgin composite. As for the flexure strength, a decrease of 90% was observed compared to the virgin composite.

The other technique is "incineration with energy recovery" which involves the complete burning of the material to produce energy and/or heat (Villanueva and Wenzel, 2007a). The incineration of composite materials is not a recycling technique since no material is recovered. As of today, this is the most widespread disposal technique for composites after landfill (Duflou et al., 2014a; Hedlund-Åström, 2005; Joshi et al., 2004). However, due to the fast treatment and easiness of use, it is a commonly applied EOL technique for natural fibre based composites. Natural fibres show

Table 2

Embodied energy per ton of fibres reported by (Dissanayake et al., 2009).

Fibre production type	Embodied Energy	
Sliver (no till + warm water retting)	59 GJ/ton fibres	
Yarn (no till + warm water retting)	86 GJ/ton fibres	
Glass fibre mat	55 GJ/ton fibres	
Continuous glass fibres	26 GJ/ton fibres	

a degradation in their mechanical properties when they are heated to medium high temperatures up to 200 °C (Gassan and Bledzki, 2001). When the incinerator design maximizes the heat recovery after combustion of the material, 50% to 70% of the heat can be recuperated as energy (Grosso et al., 2010). Both the matrix and the fibres can be combusted in the case of natural fibre. In contrast, only the matrix material of glass fibre composites can be combusted while the glass fibres form a slag which sticks to the incinerator walls and requires an extra process to remove it. The non-combustible glass residue needs further treatments. It constitutes one of the major disadvantages for the incineration of glass reinforced plastics (Duflou et al., 2014a).

Flax is often compared to glass fibres as they both share similar specific properties as reported in many studies (Arbelaiz et al., 2005; Bodros et al., 2007; Verpoest et al., 2012). Thus, many comparative studies are made between glass and natural fibres in order to see if the substitution of glass fibre by flax fibres is environmentally beneficial. Life cycle studies (González-García et al., 2010; Joshi et al., 2004; Le Duigou et al., 2011) comparing flax and glass fibres have shown that flax fibres are undoubtedly beneficial for the environment compared to glass fibres as seen in Table 1. Furthermore, Le Duigou and Baley, (2014) have found that substituting glass fibres by flax fibres in a polypropylene composite with a similar material index reduces all the environmental impacts by 10% for abiotic depletion and global warming to 20% for acidification. Non-renewable energy used was also reduced by 12% passing from 109.8 MJ/kg for glass-PP composite to 96.5 MJ/kg for flax-PP composite. It was found that using an equal stiffness criterion have shown that flax mat-PP with higher volume fraction is environmentally more advantageous (impact category of climate change) if compared to glass mat-PP composite (Duflou et al., 2012, 2014a; Yelin, 2014). Since flax fibres have a lower density than glass fibres (1.45 vs 2.55 kg/m³), flax composite lead to lighter structure and contribute to lower fuel consumption. However, if a higher quantity of flax is used to achieve the targeted properties, such as stiffness or strength, the environmental advantage of flax may be reduced.

Diverging observations were made by Dissanayake et al. (Dissanayake et al., 2009) who found that the production of flax sliver in England and glass mat is comparable in terms of energy as seen in Table 2. This is caused by the fact that the entire environmental burden was assigned the flax fibre product alone, whereas the by-products, like slivers, were considered as waste. This is not realistic as all by-products (dust, shives, woody parts, etc...) are used in the industry to produce random mat, injection moulding compounds or flax boards. On the other hand, Le Duigou et al. allocated a significant part of the environmental burden to the by-products. Furthermore, the former authors (Dissanayake et al., 2009) used wet spinning data of very fine linen yarns aimed for

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