# POLY-PAPER: AN INNOVATIVE THERMOPLASTIC ECO-COMPOSITE MATERIAL RECYCLABLE WITH PAPER AND CARDBOARD

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## ABSTRACT

The research is focused on the development of an innovative eco-composite material based on circular polymers reinforced with cellulose fibres. Poly-paper is a biodegradable and recyclable material that can be processed as a conventional thermoplastic polymer, but recyclable into paper and cardboard recycling chain.

The new composite was called "Poly-paper" because it brings together the main properties of two materials families that play a key role in today's packaging: processability of plastics and virtuous recycling in the chain of paper. Poly-paper specific application is related to packaging industry: it can replace various plastic components in a responsible way and it can be easily integrated and recycled with paper and cardboard packaging.

*Keywords*: eco-composite, cellulose fibers, polyvinyl alcohol, Renewability, injection moulding.

# **1 INTRODUCTION**

Packaging materials typologies are various, as many are the functions they need to fulfil. These materials come from different origins, have different functions, but also a different recycling channel.

In industrial case studies, different materials are usually combined in a single packaging: corrugated cardboard, printed paper, polystyrene foam, polyethylene and various other plastic components [1].

Each of these materials follows a different path for the collection, disposal and recycling. If used in a single package, these materials are not always easily recognised and distinguished by the consumer that is usually uninformed about the correct disposal in the recycling cycle. In a sustainable design process, all packaging materials should be disassembled and collected in a specific

recycling channel. Cellulosic packaging is a perfect example of circularity: cellulosic recycling channel represents the most active and constantly growing recycling channel both for its virtuosity and for the high percentage of the recycling rate set by ISPRA, Urban Waste Report at 80% in 2016.

Nowadays, designers are called to develop responsible packaging materials: circular plastic market represents a growing trend for future years as reported by European Bioplastics, nova-Institute, 2016.

# 2 BACKGROUND

In the last 20 years, economic, environmental, technical and legislative issues can be considered drivers for the development of sustainable packaging materials reinforced with natural fibers or fillers.

The use of natural fibers in substitution, for example, of aramid and glass fibers makes possible to obtain sustainable composite materials. They have multiple advantages over artificial fibers as they are renewable, have low density and high strength and modulus, and are non abrasive [2]. In particular, cellulose fibers advantages in sustainable composites are toughness, flexibility, easy processing, recyclability, eco friendliness. This green reinforcement has been applied to several searches in the last few decades [3].

Several researches deal with the formulation of composites using cellulose fibers.

Some of these investigate biodegradability, others, improved mechanical characteristics, thermal or acoustic insulation in various sectors of application. A study by Grapner et al [4] describes the mechanical characteristics of PLA composites reinforced with different kinds of natural fibres in mass proportion of 40 wt% by compression moulding. Zhang et al in 2011 [5] formulated PVA/cellulose composites with improved mechanical performances, the percentage of cellulose added was fixed at 30 wt% of PVA used. Curvelo et al [6] proposed composites prepared with regular cornstarch plasticized with glycerin and reinforced with short cellulosic fibers (16% w/w), results show an increase of 100% in tensile strength and more than 50% in modulus with respect to non-reinforced thermoplastic. Huda et al in 2005 [7] obtained a green composite prepared from poly(lactic acid) (PLA) and recycled cellulose fibers (reclaimed from newspaper/magazine or kraft paper stock), the average length of fibers is about 850 microns used up to 40 wt%.

The properties of polymer composite materials are closely linked to the polymeric properties of the matrix and the type, shape, size, concentration, distribution, orientation of the reinforcement. In Fibres reinforced composites the reinforcement can be of natural or synthetic origin, or both combined [3]. The polymer composites are further classified into 100% renewable composite, partly renewable composite, non-renewable polymer composites based on the renewable or non-renewable nature of the polymer matrix / reinforcement [3]. Several studies focused on the possibility of applying natural fibers as reinforcement in a composite and processing it by injection molding, this is closely linked to the fibers used in the matrix. Injection molding is suitable and investigated for natural fiber reinforced thermoplastic biocomposite [8]. It is therefore necessary to investigate different production processes of natural fiber reinforced composites: typical processes include extrusion, injection molding, compression molding and filament winding. Before moving towards a specific production process, we need to focus on different criteria including desired properties, size, and shape of the resultant composites, the production rate, processing raw materials and the manufacturing [8].

The aim of this work is the development of a sustainable composite material for packaging based on renewable fibers in a water-soluble and eco-compatible polymeric matrix. The composite formulation will ensure the recyclability in water, typical of the paper industry. The possibility of combination with cellulosic products is needed to get packaging intended for the same disposal chain.

Another objective is to establish possible manufacturing processes for the composite according to the methods used in the following work. The best processes are those that guarantee large scale production, typical of the packaging sector. This would apply the composite to replace traditional plastic packaging items.

The project idea is a material suitable for fulfilling the functions of packaging: contain, protect, transport, which will be insured according to the mechanical characterisations.

## **3** MATERIALS AND METHODS

The polymer selected as matrix for the composite is an experimental formulation based on water-soluble polyvinyl alcohol (PVA), with degree of hydrolysis between 75% and 90%, average molecular weight (Mw) between 75 and 150 kDa and a polydispersity index (Mw/Mn) between 2.5 and 4.3. It is a polymer-based material produced without taking fertile land for agriculture, with water soluble properties and forming non-toxic composites [9], [10]. PVA (polyvinyl alcohol) has been investigated to be used as a potential matrix for sustainable composites, thanks to its biodegradability, bio and eco compatibility, high tensile strength, excellent adhesive properties, and chemical resistance and gas barrier properties [5], [11], [12]. The reinforcement selected is cellulose fibres, to ensure and amplify the recycling of the entire composite in water. The cellulose fibers selected as reinforcement had dimensions lower than 45  $\mu$ m (>45  $\mu$ m 0%-0.1%), and bulk density in the range of 232-248 g/L. Different percentages of cellulose were investigated (from 0 up to 60 wt%).

The obtained eco-composite materials have been processed by extrusion by a corotating twinscrew extruder (Lab-Compounder KETSE 20/40D EC; Brabender, Duisburg, Germany). The composites were pelletized from the extruded filament, to be processed by injection molding.

From all formulations, dog bone specimens with dimensions according to ISO 527-3-1B (150mm x 20mm max) were fabricated using Ray Ran test Sample Injection Moulding Apparatus. Speciments were prepared at 180°C melt temperature and 70°C mold temperature with injection and holding pressure of 8 bar.

Tensile properties of the injection molded specimens were measured using an Instron 5985 universal testing machine, equipped with a load cell of 10 kN in accordance to ASTM D638 standard.

#### 4 **RESULTS**

Mechanical test results exhibited a good stress-strain behaviour as well as a close correlation between cellulose fibre content and tensile properties.

Young modulus showed an increase of three folds from 10 to 30 wt% cellulose content, as shown in Figure 1. Furthermore, the trend of the tensile modulus has been determined up to 60 wt% cellulose content by other methods reaching a 10 folds increase.

At the same time, as shown in Table 1 a slighter increase in stress at break (from  $9.78 \pm 0.13$  MPa to  $11.94 \pm 0.67$  MPa) together with a significant reduction in elongation at break was detected (from  $276.1 \pm 17.0$  % to  $57.7 \pm 2.5$  %). All the eco-composite formulations show a good feasibility that has been optimized in the definition of process temperature:  $180^{\circ}$ C for the barrel and 70 °C for the mould in case of injection moulding apparatus.

Fibers	Yeld stress (MPa)	Tensile modulus (MPa)	Strain to failure (%)
0 wt%	$9,21 \pm 0,62$	$24,35 \pm 3,51$	$248,84 \pm 20,48$
10 wt%	$7,95 \pm 0,44$	$46,12 \pm 3,47$	$103,26 \pm 28,69$
20 wt%	$8,79 \pm 0,52$	$79,75 \pm 10,72$	$70,17 \pm 8,68$
30 wt%	$9,21 \pm 0,10$	$127,24 \pm 11,42$	$42,18 \pm 7,24$

Table 1: Mechanical parameters from tensile characterization tests performed on PVA reinforced with different percentages of cellulose fibers (0 to 30 wt%).



Figure 1: Tensile modulus increase in relation to cellulose fibres content.

One formulation of the new eco-composite material (30 wt%) has been preliminary tested to check the possibility of belonging to the cellulosic recycling chain. Then water dissolution tests were carried out at temperature of 21°C and 40 °C, in static and turbulence water conditions. A correlation between water turbulence and temperature increase with the reduced dissolution time of the material was evidenced by the tests as shown in Figure 2.





#### **5** CONCLUSIONS

The eco-composite developed has been called Poly-paper and its specific application is related to packaging industry [1], [13]. Poly-paper is feasible as thermoplastic polymers, so it can replace various plastic components in the packaging. Poly-paper can be considered a partly renewable composite that can be easily integrated with paper and cardboard packaging, being recyclable with them thanks to the easy separation of the two components: the amount of PVA matrix is dissolved in water and the cellulose fibres extracted.

Therefore, Poly-paper combines the fundamental aspects of the two main packaging materials families: from the polymers the productive versatility, from the cellulosic materials the conscious recycling and the value of environmental responsibility.

The innovation of this project concerns a conceptual rethinking before technological and engineering one: the study of materials, processes and management of circularity through the enhancement of natural resources and the minimization of waste production. The management of the life of packaging is certainly the most important part for the economy and the environment.

Our findings resulted in the filing of a patent co-owned by the Milan Polytechnic and NextMaterials srl [14], [15].

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