Sustainable materials and processes design: the case study of Poly-Paper

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ABSTRACT

Polymeric materials used for packaging have unsolved environmental sustainability problems. The ideal packaging must be recyclable and/or compostable, with a unique recycling channel. This is focused on the development of an innovative eco-composite material based on water-soluble polymeric matrix reinforced with cellulose fibers up to 60 % w/w: Poly-Paper. The material can be processed by conventional processes of thermoplastics (extrusion, thermoforming, injection moulding, 3D printing) and can be recycled together with paper-based materials. This work describes the innovative expanded version of Poly-Paper and the developed forming technology: Water Shaping, able to give the material a great impact and energy absorption capacity in packaging applications.
Key Words: Cellulose fibers, eco-composite, recyclable, sustainable processes

1. INTRODUCTION

The classes of packaging materials are numerous, since there are many functions that packaging materials must fulfil. In many industrial case studies, different classes of materials are usually combined in a single package: corrugated cardboard, printed-paper, polystyrene foam, low-density polyethylene and various other plastic components, etc..(Del Curto et al., 2016). Each of these materials follows a different path for collection, disposal and recycling. If used jointly, in a single package, it can happen that they are not distinguished from the consumer and conferred jointly to the same waste stream. In a sustainable design process, the different packaging materials must be separable, separated by the consumer, and collected in a specific recycling channel. The cellulosic packaging is a perfect example of circularity: the cellulosic recycling channel represents the most active and constantly growing recycling channel; in fact, the high percentage of recycling rate from the ISPRA, Urban Waste Report was calculated at 80% in the 2016[1]. Nowadays, designers are asked to select, but also to combine and develop responsible packaging materials: the sustainable materials market is a growing trend for the years to come, as reported by European Bioplastics, nova-Institute, 2016[2].

2. BACKGROUND

In the last 20 years, economic, environmental, social and legislative assets have been the main drivers for the development of sustainable packaging materials. The use of natural fibers in bioplastic matrices has made possible the development of sustainable composites, enhanced in properties and performance. Natural fibers have multiple advantages over synthetic fibers as they are renewable, have low density and high mechanical strength and modulus and are not abrasive for machinery and plants (Cheng, Wang, Rials, & Lee, 2007). In particular, the advantages of cellulose fibers used as fillers in sustainable composites are hardness, flexibility, ease of processing, recyclability and eco-compatibility. This green reinforcement has been applied to various researches in the last decades (Thakur, 2013). These applications exploit different characteristics of cellulose fibers as biodegradability, mechanical performances, thermal or acoustic insulation in various fields of application (Curvelo, De Carvalho, & Agnelli, 2001; Graupner, Herrmann, & Müssig, 2009; Huda, Mohanty, Drzal, Schut, & Misra, 2005).
The aim of this work is the development of a sustainable composite material for packaging based on renewable fibers in a water-soluble and environmentally friendly polymer matrix: Poly-Paper.
The name Poly-Paper was born from the idea of combining the processes of thermoplastic polymers (e.g., extrusion) and the virtuous end of life of paper materials. The basic idea is designing a material suitable for carrying out all the functions of packaging: contain, protect, transport. In fact, Poly-Paper has been developed to be processed by extrusion, thermoforming, injection moulding to realize all those components that need high technical details but also in the "expanded" version for high performance in protection and transport.
In addition to traditional processes (extrusion, injection moulding, thermoforming, 3D printing), an innovative process has been presented that exploits the properties of the selected materials: water solubility. This type of technology, called "Water Shaping", allows to improve the resistance to shocks and the ability to absorb energy.

3. MATERIALS AND METHODS

The polymer selected as the matrix for the composite is an experimental formulation based on water-soluble polyvinyl alcohol (PVA), a fossil-based material selected to be used as a potential matrix for sustainable composites. This thanks to its biodegradability, biological and ecological compatibility, high tensile resistance tensile, excellent adhesive properties and chemical resistance and gas barrier properties (Chiellini, Corti, & Solaro, 1999; Mandal & Chakrabarty, 2014). The selected reinforcement is microcrystalline cellulose (MCC), environment friendly products, gained from replenishable raw materials.

3.1. Poly-Paper Formulations

Seven types of Poly-Paper composites were developed, increasing the MCC content from 0 to 60% w / w. For each considered percentage, filaments (ø 3mm) were obtained through the FSCM series twin-screw extruder (TSA Industriale, Cernobbio, CO, Italy). The filaments were subsequently pelletized to obtain granules which have been used as the main semi-finished products for subsequent processing.

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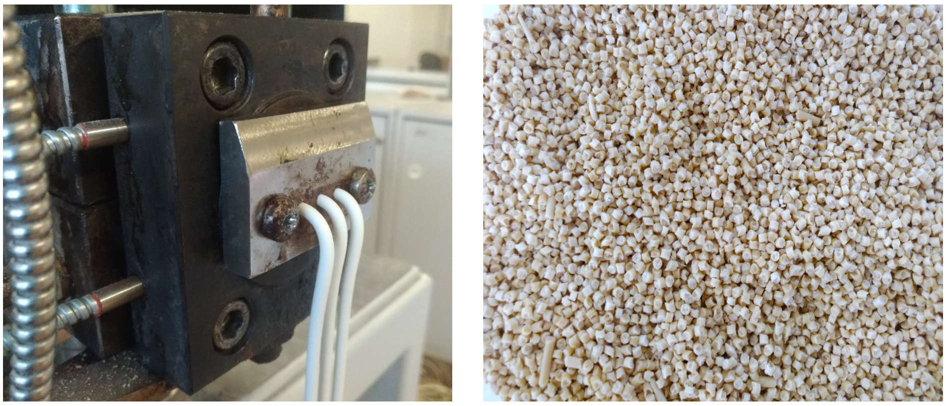
[1] Ispra, Urban Waste Report 2016, available at http://www.isprambiente.gov.it/it/pubblicazioni/rapporti/rapporto-rifiuti-urbani-edizione-2016

[2] European Bioplastics, 2018 Bioplastics – facts and figures, available at https://docs.european-bioplastics.org/publications/EUBP_Facts_and_figures.pdf

3.2. Poly-Paper expansion

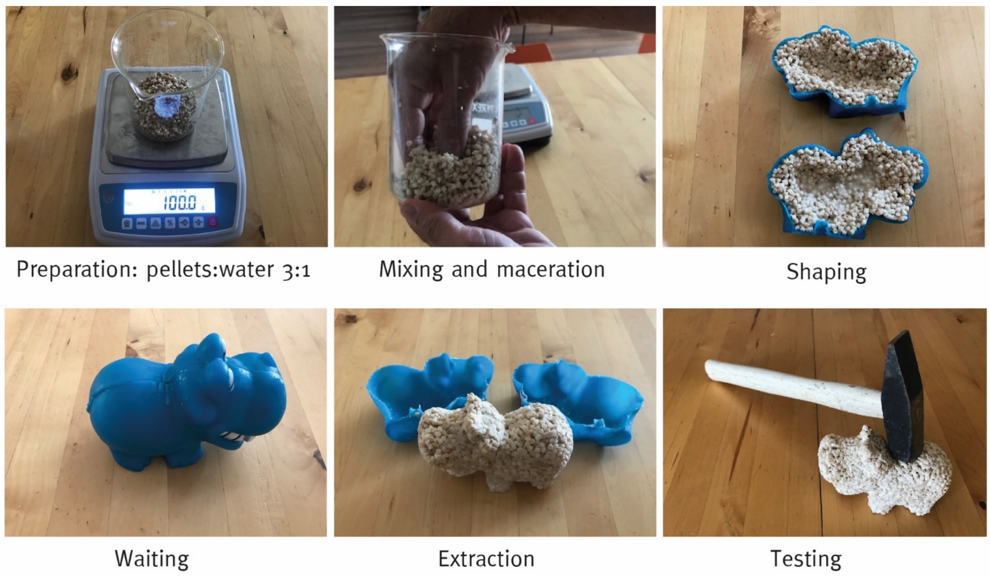
The expansion of the material has been obtained starting from the formulation of Poly-Paper and adding 2% w/w of expander agent. The composite expansion was obtained by the same extrusion process described above, without process variants. The PVA:MCC:EXPander mixtures were extruded into filaments with a nominal diameter of 3 mm and granulated as shown in Figure1.



[Figure 1] *Expanded Poly-Paper extrusion process (left) and obtained pellets (right)*

3.3. Water Shaping

A particular method for shaping has been developed using water, taking advantage of the fact that Poly-Paper matrix: PVA is a water-soluble polymer. The process has been called Water Shaping; a scheme of the steps of this process is proposed in Figure 2. A certain amount of Poly-Paper pellets is manually mixed with a third weight of water for 15 min. The obtained mixture is left to macerate for one hour at room temperature and then put into the desired mould. The granules, in this way, after the evaporation of the water excess through drying in the oven (60°C for 3 hours), are firmly bonded to each other and with materials such as paper, cardboard, wood and ceramics. The process has been performed using expanded and non-expanded Poly-Paper pellets.



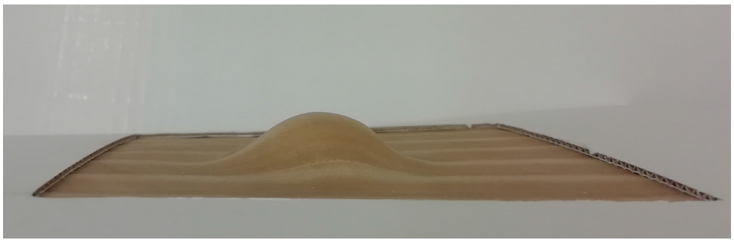
[Figure 2] *Water Shaping process stages*

3.4. Compression tests

Compression tests have been carried out on a large series of disks (diameter = 15 mm, height = 10 mm). The disks were subjected to compression tests with a crosshead speed of -0.15 mm/s, until a 90% deformation is achieved, i.e., the specimens reached a final thickness of 1.0 mm.

4. RESULTS

Poly-Paper[3] can be processed using traditional processes of thermoplastic materials. During the development of the material, in fact, the possibilities of thermoforming, 3D printing as shown in Figure 3, injection molding as well as extrusion have been tested with good results. These production versatility allows the material to open up multiple application scenarios (Testa, 2018).

[Figure 3] *Poly-Paper processed by 3D printing (left) and thermoforming (right).*

4.1. Poly-Paper expansion

The Poly-Paper mixture (PVA: MCC) was developed up to 60% of MCC w/w and results was taken as a reference for the expansion attempt (Santi et al., 2018). Various types of PVA:MCC ratios: 100:0, 70:30, 60:40 were extruded with a factor of 2% of expanding agent. Table 1 shows the different types of granules produced for the subsequent experimentation. As can be observed, the expanding agent has little effect on the pure PVA, while it has an important effect in the presence of MCC content of 30 and 40% w/w. By expansion, it is possible to reach an apparent density of about 0.45 g/cm³ with an expansion of 40% compared to the same mixture without expander agent. Furthermore, the addition of an expander causes the granules to change color, which turns from light brown to whitish as shown in Figure 4.

[Table 1] *Expansion factor determination of 100:0, 70:30, 60:40 PVA:MCC ratios compared to the non-expanded Poly-Paper formulations*

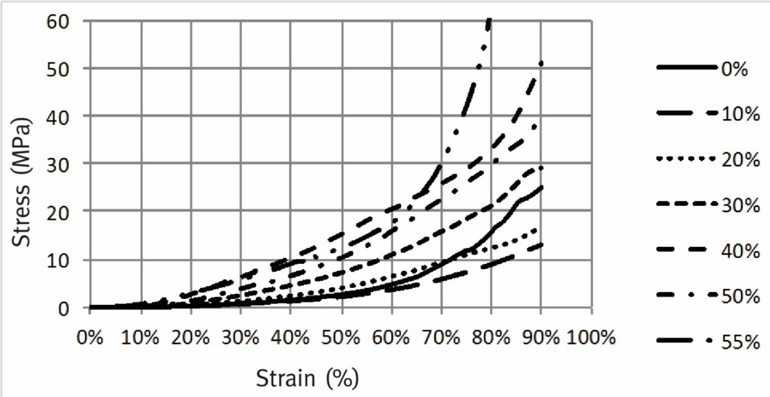
PVA:MCC ratio (%) w/w	Expanding agent (%) w/w	Volumetric mass density (g/cm³)	Expansion factor (%)	Color
100:0	0%	0,74	0%	Translucent brown
100:0	2%	0,73	1,3%	Translucent white
70:30	0%	0,70	0%	Light brown
70:30	2%	0,44	39,7%	Whitish
60:40	0%	0,70	0%	Brown
60:40	2%	0,46	34,3%	Whitish


[Figure 4] *Discs obtained by Water Shaping process (diameter =101 mm, height = 36 mm). On the left: 70:30 PVA:MCC disk without expander; on the right: a disk obtained with the same PVA:MCC ratio, additioned with the expanding agent.*

4.1. Compression tests

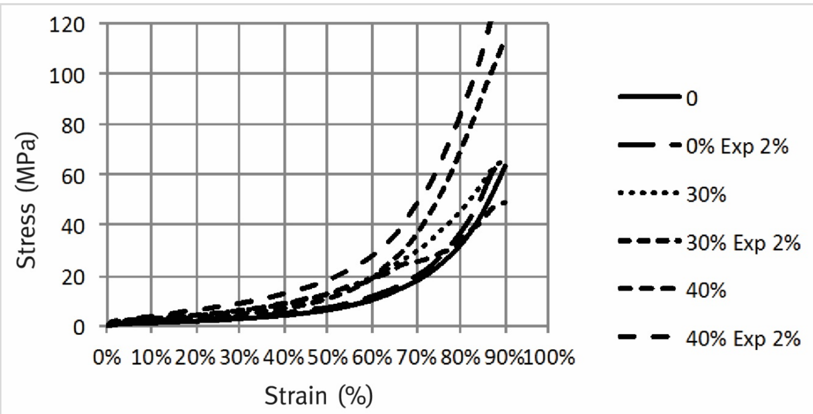
[3] Italian patent n. 102015000028276 "Materiale composito ad alta sostenibilità ambientale", filed 26/6/2015 - granted 30/11/2017.
PCT Application (UE, USA) n. IB2016/053777 "Highly environmentally sustainable composite material" filed 24/6/2016 – published EP 16745832.2 4/4/2018

As can be observed in Figure 3, the macrostructure of the material obtained Water Shaping appears similar to that of the expanded polystyrene, a material notoriously capable of absorbing shocks. The porous structure of the expanded foams is in fact able to absorb a high amount of energy during the compression deformation due to a "peak load" effect connected to the expanded structure and to the granular shape. It can be assumed that the structure of the Poly-Paper disks obtained by Water Shaping process may have the mechanical behaviour of the expanded foams. To evaluate these hypotheses, compression test were carried out. The tests results can be analysed in relation to the amount of MCC content or in relation with the expansion factor. To verify the effect of the cellulose percentage, samples with different percentages of MCC content have been obtained, ranging between 0 and 55% w/w. The increase in the percentage of cellulose causes an improvement in the mechanical compression properties, in particular the stiffness showed an increase of about five times in the slope in the first part of the stress-strain curves (Figure 5).



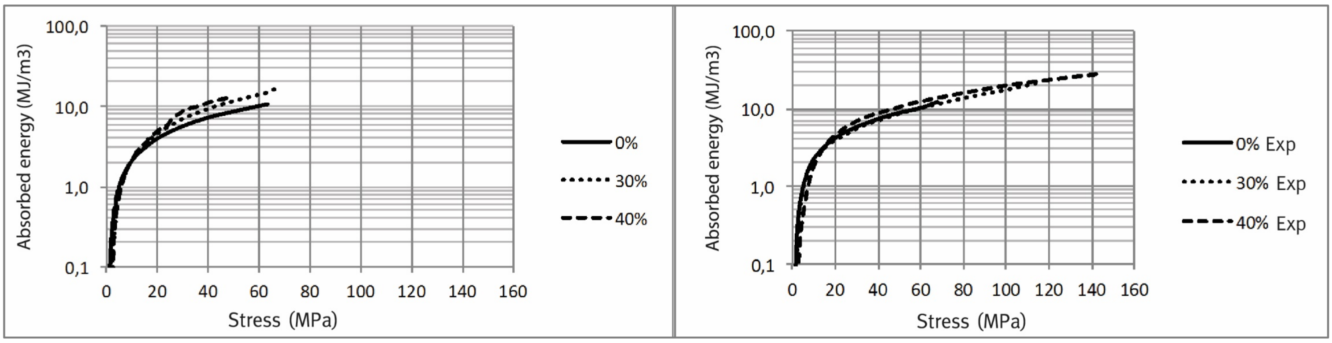
[Figure 5] *Representative stress–strain curves obtained by mechanical compression tests performed on the Water shaped disks made with different PVA:MCC ratios.*

As previously mentioned, the possibility of expanding the granules has been verified by adding an optimized percentage of 2% w/w of expanding agent into the starting dry PVA/MCC blend. As reported in Figure 6, the addition of an expanding agent while decreasing the apparent density of the material (Table 1) causes an increase in the mechanical properties.



[Figure 6] *Representative stress–strain curves obtained by mechanical compression test performed on the Water shaped disks made with different PVA:MCC Poly-Paper ratios expanded and non-expanded.*

The Stress-Strain curves can be integrated to calculate the absorbed energy as a function of the applied stress. This data was obtained for samples with 0, 30 and 40% w/w MCC content, without and with expanding agent (2% w/w). Results are shown in Figure 7 in semi-logarithmic scale. It can be clearly observed that with 30 and 40% w/w MCC the energy absorbing capacity of the expanded material is greater than that of similar unexpanded Poly-Paper.



[Figure 7] *Representative stress-absorbed energy of the Water shaped disks made with different PVA:MCC Poly-Paper ratios non-expanded (left) and expanded (right).*

5. CONCLUSIONS

Poly-Paper, due to the use of a water-soluble (PVA) matrix reinforced with cellulose fibers, has great advantages in terms of eco-sustainability:

- it is produced using cellulose fibers obtained from recycled paper, in a circular economy model;
- It can be selected for the production of "mono-material" packaging and can be recycled together with corrugated cardboard, overcoming the problems associated with the use of expanded polystyrene.

The developed expanded Poly-Paper version allows the production of low-density packaging products for protection and transportation. By Water Shaping process it's possible to obtain complex shapes into simple molds, ideally made of cardboard with hydrophobic treatment, without the need for complex molds or machining operations, and without disposal problems.
Important advantages over actual packaging components have been achieved in terms of ability to absorb energy, lower density and cost per unit volume. Using industrial technologies, the material can certainly be brought to higher levels of expansion.

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