



# Synergetic effect of *Posidonia oceanica* fibres and deinking paper sludge on the thermo-mechanical properties of high density polyethylene composites



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## ABSTRACT

This study evaluated the reinforcement potential of *Posidonia oceanica* fibres (POF) and deinking paper sludge (DPS) on the thermo-mechanical properties of high density polyethylene (HDPE) binary and hybrid composites. The weight ratios of the fillers (POF and/or DPS) to the polymer were 0:100; 20:80; 30:70 and 40:60 (wt:wt). Maleated polyethylene (MAPE) was added at a proportion of 3% by weight. The chemical composition, structural and thermal properties and morphology of the fillers were investigated by Fourier transform infrared (FTIR), thermogravimetric analysis (TGA) and scanning electron microscopy (SEM). The mechanical (tensile and impact strength) and thermal properties (TGA and Differential Scanning Calorimetry (DSC)) of the binary and hybrid composites were also investigated. Binary and hybrid composite properties depend on the chemical composition of the fillers (POF and DPS), the filler/matrix interfacial adhesion and the POF: DPS ratio. The tensile modulus and strength of the binary and hybrid composites increased with increasing filler content (POF or/and DPS). A better interfacial adhesion between fillers and matrix was achieved in the presence of MAPE. HDPE/POF/MAPE composites achieved the highest tensile modulus and strength with 40% POF. But lower thermal stability, ductility and impact strength were found with the addition of the POF. However, the thermal stability, crystallinity, ductility and toughness improved in HDPE/POF/DPS/MAPE hybrid composites due to the addition of DPS or the substitution of POF by DPS.

## 1. Introduction

The use of natural fibres to process novel composites has attracted growing interest (Habibi et al., 2008) given their abundance, low cost, low density, non-toxicity, biodegradability, high specific properties (modulus and strength), ease of fibre surface modification and low abrasiveness (Jawaid et al., 2011). In fact, several industries, such as automotive (Bajpai et al., 2013; Faruk et al., 2014), construction (Zhu et al., 1994) and packaging (Khiari et al., 2010), have focused their attention on the development of new materials filled with natural fibres. However, given their increasing demand for traditional uses (paper, textile, cellulose derivatives) and their potential incorporation into new material families, new sources of lignocellulosic fibres must be identified. For this reason, several studies have investigated the feasibility of using various agricultural waste fillers such as almond (Essabir et al., 2013), walnut (Zahedi et al., 2013) and coconut (Bledzki et al., 2010) shells, barley husks (Bledzki et al., 2010), cotton stalks (Habibi et al., 2008) and rice straw (Habibi et al., 2008), as reinforcements in

polymer composites. Fibre waste derived from marine biomass such as *Posidonia oceanica* balls, which are extensively available on Tunisian beaches, has attracted interest. *Posidonia oceanica* (*P. oceanica*) is the dominant grass in the Mediterranean Sea, which appears as a raw material in the form of balls (egagropili). *P. oceanica* has been used as a reinforcement in composite materials with many polymeric matrixes such as BIOPLAST (Khiari et al., 2011), wheat gluten (Ferrero et al., 2013), high density polyethylene (HDPE) (Puglia et al., 2014), biobased HDPE (Ferrero et al., 2015) and polyester (Zannen et al., 2016). However, there is a need for constant production and management of *P. oceanica* fibres (POF) to produce composites for industrial applications.

Deinking paper sludge (DPS) is another type of waste. Important quantities of DPS are generated by the paper industry and are more easily available for industrial production compared to natural fibres. DPS is generally composed of short cellulose fibres and inorganic materials (predominantly calcite, kaolinite and talc). POF and DPS can offer several benefits as substitutes for conventional inorganic reinforcement fillers in the manufacturing of thermoplastic composites

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(Son et al., 2001). The use of DPS at high concentrations with HDPE could improve the thermo-mechanical properties of composites (Eloumi et al., 2018; Haddar et al., 2017). DPS improves composite stability, density and ductility and reduces cost. The hybridisation of fibres, which is the combination of two or more types of fibres in a matrix to produce hybrid composites, is gaining interest (Majeed et al., 2013; Zainudin et al., 2014). It combines the advantages of each type of fibre to produce composites with superior performance at a reduced cost. Recently, interesting results were found for hybrid composites combining two types of natural fibres (Jawaid et al., 2011; Pérez-Fonseca et al., 2014), glass fibres (Khalil et al., 2007; Kord, 2012), carbon fibres (Rattanasom et al., 2007; Kada et al., 2016) and inorganic fillers (Essabir et al., 2016). DPS has gained importance as a filler because of its high performance, high stability, low density, ease of processing, availability and lower cost. Its use could produce hybrid composites with interesting specific properties and costs. Hybrid composites with 10% wheat straw fibre and 30% paper mill sludge showed high bending strength (Khademieslam and Kalagar, 2016).

In this work, POF and DPS were used as reinforcement agents for composite materials. These fillers were added at different ratios to a thermoplastic matrix (HDPE) to produce various combinations of binary and hybrid composites. The objective of the present study is to produce composites with optimal thermal and mechanical performance through the hybridisation of POF natural fibres and DPS waste.

## 2. Materials and methods

### 2.1. Preparation of the samples: POF and DPS

The *Posidonia oceanica* balls (egagropili) (Fig. 1a) used in this study were collected from Monastir (Tunisia) in March 2014. They were opened manually and washed with tap and distilled water to eliminate

sand and other soil contaminants. They were subsequently dried at 80 °C in a hot air oven for 7 days. The fibre was reduced to powder by milling with a grinder (Retsch SK 100) and sieved to an approx. 1 mm size (Fig. 1b). It was then stored in plastic bags to protect it from moisture.

The DPS material used in this work was sampled from the deinking process of recycled paper at the TUNISIE OUATE waste treatment plant in Tunisia. The DPS used in this study (Fig. 1c) is described in a previous report (Haddar et al., 2017).

### 2.2. Characterisation of the raw samples

The chemical composition of the raw POF was determined by using several standard methods. The evaluation of extractive substances was carried out in different liquids: Ethanol-toluene (TAPPI method T204 cm-07, 2007), hot water (TAPPI method T207 cm-08, 2008) and 1% sodium hydroxide solution (TAPPI method T212 om-07, 2007). The ash content was determined according to the TAPPI method T211 om-93 (2000) standard method. The amount of lignin, holocellulose and cellulose were also assessed by using the Klason lignin content (TAPPI method T222 om-06, 2006), the Wise et al. (1946) and the Weende method using the Fibertec M6 system (Carrier et al., 2011), respectively. The morphology of the POF was investigated by scanning electron microscopy (SEM). The Fourier transform infrared (FTIR) spectrum of POF was measured with a Perkin Elmer Spectrum BX FTIR System using the KBr-pellet method as previously described in Haddar et al. (2017). The thermal stability of POF was performed using a PerkinElmer Pyris 6 TGA analyzer in a nitrogen atmosphere. The sample was heated from room temperature to 900 °C at a rate of 10 °C/min with a nitrogen gas flow rate of 40 mL/min. A derivative thermogravimetric (DTG) curve was calculated from the TGA data.



Fig. 1. Filler images ((a) *Posidonia oceanica* balls, (b) *Posidonia oceanica* fibres (POF) and (c) Raw DPS).

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