



## Original Research Paper

Deposition of nanostructures derived from electrostatically stabilised TiO<sub>2</sub> aqueous suspension onto a biocompositeM. Nazli Naim<sup>a,\*</sup>, Atiqah Raihan Jaafar<sup>a</sup>, Noor Fitrah Abu Bakar<sup>b</sup>, Azhari Samsu Baharuddin<sup>a</sup>, Roseliza Kadir Basha<sup>a</sup>, I. Wuled Lenggoro<sup>c,d</sup><sup>a</sup> Faculty of Engineering, Department of Process and Food Engineering, Universiti Putra Malaysia, Serdang, Selangor 43400, Malaysia<sup>b</sup> Faculty of Chemical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia<sup>c</sup> Graduate School of Bio-Applications and Systems Engineering, 2-24-16 Nakacho, Koganei, Tokyo 184-8588, Japan<sup>d</sup> Department of Chemical Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Nakacho, Koganei, Tokyo 184-8588, Japan

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## ABSTRACT

A nanostructure derived from TiO<sub>2</sub> particle deposition onto a biocomposite surface derived from coir dust (CD) was developed to control degradation using a spray dry technique. To stabilise and reduce the size of dispersed particles, the TiO<sub>2</sub> powder was prepared in deionised water at pH 10 and sonicated at 20 kHz and 400 W. The coir dust was obtained from coconut kernel waste and underwent drying treatment before it was mixed with polypropylene (PP) as the substrate. The suspension consisted of particles with an average size and zeta value of 285 nm and −19.2 mV, respectively. The suspension was spray dried onto a hot-pressed substrate (biocomposite) with a surface roughness between 0.23 and 1.57 μm at ambient temperature. Scanning electron microscopy image analysis and Fourier transform infrared spectroscopy analysis indicated that the TiO<sub>2</sub> particles were successfully deposited onto the substrate, shown by the existence of a carboxylic acid group (—COOH) in the CD matrix. Moreover, the weight of the deposited substrate increased exponentially with deposition time compared to pure PP substrate. However, the deposition rate of TiO<sub>2</sub> nanoparticles was limited by the ratio of the substrate surface roughness to particle diameter, as predicted by a previous study.

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## 1. Introduction

Waste from coconut shells, such as coir dust (CD), is normally used as an abundant, naturally occurring polymer that can be obtained from numerous resources in tropical regions. In 2010, Asia was the major producer of coconuts in the world with a total annual production of up to 46 million tons per annum [1]. Discarded CD normally becomes enormous piles of waste that are difficult to dispose of. CD is commonly used by locals as biofuel, which increases air pollution to the environment. Coir dust consists of lignocellulosic fibres organised into fibrils, which are surrounded by a matrix of lignin and hemicellulose. The fibre, when combined with certain polymers, could act as a reinforcing phase in polymeric matrix composites [2]. The applications of agro-fibre based components include railways, automotive, aircraft and furniture industries, irrigation systems and sports items [3]. Polymeric materials are soft, flexible and lightweight in comparison to fibres;

their combination provides a high strength-to-weight ratio to the resulting biocomposite. A major disadvantage of cellulose fibres is their high polarity in the natural form, which makes them incompatible with non-polar polymers. Biocomposites are used only to a limited extent in industrial practice due to the inherent polar and hydrophilic nature of polysaccharides that contribute to the attraction of contaminants such as dirt and microbes. Biocomposites also possess poor resistance to moisture absorption, which makes the use of natural fibres less attractive for exterior applications. In general, the properties of biocomposites depend on those of the individual components and on their interfacial compatibility.

Thin-film deposition of a photocatalytic material on a biocomposite surface provides a protective layer. The self-cleaning mechanism of TiO<sub>2</sub> is useful on the surface and increases the added value of the composite. Thin layer of TiO<sub>2</sub> will work as a photocatalyst layer, where its function is to decompose dirt, bacteria and other organic contamination with the aid of ultraviolet light [4]. For example, Minabe et al. studied the degradation of several liquid and solid organic compounds using thin TiO<sub>2</sub> films under UV

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illumination. They showed an exact agreement between the weight losses of the solid compounds octadecane and stearic acid and the weight of CO<sub>2</sub> produced during photocatalytic degradation [5]. Further work by Sunada et al. discovered that the TiO<sub>2</sub> photocatalyst had both bactericidal activity and decomposing activity against endotoxin (i.e., detoxifying activity) [6]. Due to the nature of biomaterial that always consist of pores and gaps, thin film coating with nanometer-order TiO<sub>2</sub> is preferable for the deposition purposes. Previously, we have demonstrated several recognised deposition techniques for nanometer-order film deposition from the liquid phase. The applied methods were electrophoretic and electro-spray deposition [7,8]. A few methods demonstrated by several workers also show potential deposition techniques such as sol-gel, electroless plating and impregnation [9]. Compared to the above mentioned methods, atomising tools using a spraying mechanism is the simplest process to deliver TiO<sub>2</sub> particles onto a biocomposite surface. There are four major controlled processes in the spray-drying or atomiser technique for thin layer formation: (i) the production of droplets; (ii) the shrinkage of charged droplets (due to solvent evaporation); (iii) droplet-to-particle conversion; and (iv) the deposition of particles (deposit) on the substrate [10,11]. Although the processes have been widely discussed in several reviews, investigation of the relationship between deposited droplets and the biocomposite surface conditions of the substrate, which may be different in terms of chemical and physical structure, is still a new research topic and is not well-studied.

In this work, a specific composition ratio consisting of coir dust and polypropylene were mixed together before being hot-pressed onto a plastic sheet and further abraded to vary the surface roughness. The suspension which consists of electrostatically stabilised titanium dioxide (TiO<sub>2</sub>) mixtures was spray dried onto the abraded sheets (substrate). This study focused on the morphology of the biocomposite surface because the relationship between the deposition of TiO<sub>2</sub> particles and the surface roughness exerts an effect on the deposition rate, as proposed previously by Browne [12]. These finding were also supported by Wood and Oron, as they found that particle deposition increased markedly by increasing the surface roughness [13,14]. Fan and Ahmadi conducted a study on the deposition of spherical particles from turbulent air streams in vertical ducts shows that instead of wall roughness, particle-to-fluid density ratio, the shear-induced lift force, the gravity direction and the flow Reynolds number had profound effects on the particle deposition rate [15]. The work is also support by Tian and Ahmadi, they also showed that when sufficient care was given to these parameters, the particle deposition rates could be predicted with reasonable accuracy [16].

Some parameters that concerned Browne and others, such as mean particle size, particle density, the Reynolds number and surface roughness ratios are also considered in this work. On the other hand, further analysis of the biocomposite components also confirmed that the lignocellulosic material derived from coir dust provides a functional group and increases the adhesion of TiO<sub>2</sub> particles to the biocomposite network on the surface.

## 2. Experiment method

### 2.1. Material and substrate preparation

#### 2.1.1. Material: Coir dust

Coir dust (CD) was collected from a plantation that produces coconuts. The plantation is located in Bagan Datoh, Perak, Malaysia. Coir dust is a sub-product of the coconut. Once the core and the fibre (coconut kernel) are separated, the coir dust is left over as a waste. The received CD was characterised by a high water content and various sources of microbes, fungi and insects. For

cleaning purposes, the CD was manually segregated, sterilised and dried in an oven at 120 °C for 48 h.

#### 2.1.2. Material: Polypropylene

A polypropylene pellets were used as the polymeric matrix, supplied by Polypropylene Malaysia Sdn. Bhd. The formula structure of polypropylene is (C<sub>3</sub>H<sub>6</sub>)<sub>n</sub> as a linear and crystalline polymer. Polypropylene was chosen for this study due to its lowest density of all major plastics, which is 0.9 g/cm<sup>3</sup>, and high tensile strength, 25–38 MPa [17].

There are three types of polypropylene: isotactic, syndiotactic and atactic. All these types of polypropylene have a different value for the melting point temperature ( $T_m$ ) and glass transition temperature ( $T_g$ ). For isotactic polypropylene, the value of  $T_m$  is 165 °C and  $T_g$  values range from –30 to 25 °C depending on the method of measurement and heat-annealing treatment. For atactic polypropylene, the value of  $T_g$  is between –12 and –15 °C and there is no definite melting point [18].

### 2.2. Substrate preparation

A grinder was used to reduce the size of the dried CD; the process was conducted for 1 h/kg. Prior to size reduction and moisture removal, CD was sieved using a 500 µm sieve and placed in the oven at 105 °C for 24 h. The dried CD was blended with a HAKKA mixer for 30 min. For PP, the granules were heated to 165 °C in an oven before being continuously left in the melted condition. CD and melted PP were mixed at a 1:1 ratio. During mixing, the CD powder was gradually inserted into the melted PP for 25 min until a well-mixed compound was observed. After the mixing process, the compound was left at ambient conditions for cooling until it turned into hard pieces of the biocomposite. The biocomposite was subjected to a compression process using a fixed rectangular mould. During compression, the biocomposite was pressed with two heated plates; the pressing process conditions of the biocomposite are shown in Table 1. The cooled pressed biocomposite was cut into small rectangular pieces which were used as the substrate. Each substrate was abraded using sandpaper to vary the surface roughness. The abrasion process also provided a contaminant-free surface and removed the undesired layer that prevents titanium dioxide from adhering onto the biocomposite matrix.

In this work, the abrasion process was conducted by treating the substrate surface with sandpaper (paper grit type 00). Prior to the uniform abrasion process, the sandpaper was placed on the tip of a metal cylinder (weight 0.45 kg) before it was applied to the substrate surface. The abrasion process for each sample was conducted for 2–4 min at 1 rpm.

### 2.3. Preparation of the electro-stabilised TiO<sub>2</sub> colloid

TiO<sub>2</sub> colloidal media in aqueous suspension was prepared using the as-received TiO<sub>2</sub> powders obtained from Merck Chemical, Sdn. Bhd. (Malaysia). The powder bulk density was 850 kg/m<sup>3</sup> and it consists of 95% Anatase with an average particles size of 27 nm. The colloidal TiO<sub>2</sub> was dispersed in deionised water using a stirrer followed by ultrasonication. The mixture was concentrated to

**Table 1**  
Processing conditions of the compounding machine.

Pressing parameter	Setting
Temperature	165 °C
Pressure	150 kg/cm <sup>2</sup>
Pre-heating	15 min
Venting	5 min
Pressing	1 min
Cooling	5 min

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