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Conference Paper

Impact Property of PLA/Flax Nonwoven Biocomposite

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Flax fibre reinforced polylactic acid (PLA) biocomposites were fabricated by using a new technique incorporating an air-laying nonwoven web forming process and compression moulding technologies. The relationship between the main process variables and the properties of the biocomposite was investigated. The results show that with the increasing of flax content, the notched Izod impact strength increased. The maximum value of 28.3 KJ/m^2 was achieved at 60% flax fibre content. As the moulding temperature and moulding time increased, the impact strength decreased. The physical properties of the biocomposites were also evaluated. As the flax fibre content increased, the void content of the biocomposites increased. This was further confirmed by the surface morphology of the composite material. The appropriate processing parameters for the biocomposites were established.

1. Introduction

Natural fibre-reinforced polymer composites are of great importance in the end-use applications [1–3]. The combined behaviour of the stiffness, elastic matrix, and strong fibrous reinforcement is achieved by these composites. The developments of fibre reinforced composites have made materials that are stiffer than steel and harder than aluminium available [4]. Natural fibres and their composites offer environmental advantages such as lower pollutant emissions and lower greenhouse gas emissions. During the last few years, many conventional materials are replaced by polymer based materials in various applications. The productivity, ease of processing, and cost reduction are the most significant advantages which the polymers offer over other traditional materials [5–7]. Biocomposites are widely used for automotive interior parts, structural parts, and interior and exterior decoration materials [8]. Biodegradable composites are becoming more popular due to their low cost and low density, and also because of the increase in oil price and recycling and environment necessities. In this project, PLA is used as a matrix, and flax fibre is used as a reinforcing material. PLA is a synthetic aliphatic polyester from renewable agriculture products; it is biodegradable and with properties comparable to some fossil-oil-based polymers [9]. Flax fibres exhibit some unique mechanical properties. Baley [10] and Charlet et al. [11] showed that flax fibres can have mechanical properties greater than those of E-glass fibres.

To make fibre reinforced composite materials, the film stacking [12, 13], injection moulding [14, 15], and compression moulding [9, 15, 16] are the most widely used manufacturing methods. In the present study, the flax fibres were blended with staple PLA fibres to form a homogenous fibre mixture. This enhances the delamination resistance of composites made from film stacking. The mixed fibres were converted to fibre webs using an air-laying nonwoven process. A unique feature of air-laid nonwoven process is to produce the webs with isotropic fibre orientation distribution [17], leading to isotropic composites. The fibre webs were thermally consolidated before finally converted to composites. This avoids any potential fibre damage caused by the widely used needle punching method for nonwoven composites [18, 19]. The

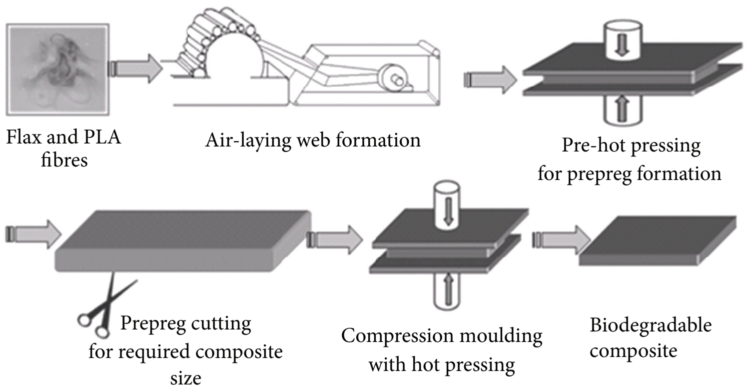


Figure 1: Schematic diagram of composite manufacturing steps.

nonwoven web was converted into biocomposites by the compression moulding process. The compression moulding method has the ability to mould large, fairly intricate parts and is cost effective.

The tensile, flexural, and thermal properties of the biocomposites manufactured by this project are reported in a previous paper [20]. The present report is mainly focused on the impact strength and physical properties of the biocomposites. The relationships between the main process variables and the performance of the composite material were investigated; the various factors that influence the performance of the composite were analyzed to determine the optimal parameters.

2. Experimental

2.1. Materials. Both PLA and flax fibres were supplied by Tilsatec Advanced Materials, Tilsatec Ltd., Wakefield, UK, in sliver form of commercial grade. The PLA was in staple fibre form with an average length of 75 mm, an average diameter of 28.46 μm, and a density of 1.25 g/cm^3. The flax fibre had an average length of 65 mm, an average diameter of 21.59 μm, and a density of 1.40 g/cm^3.

2.2. Composite Preparation. The PLA/Flax biocomposites were prepared by using the air laying web formation technique. The weights of PLA and flax fibres were measured so as to determine the weight percentage of fibre and matrix polymer of the resulting composite. The flax and PLA fibres were mixed during the web forming process. The nonwoven web was then folded to the required thickness. In order to facilitate handling, the folded web was then prepressed, cut to the required size, and finally hot pressed to form the composite material. Figure 1 shows a schematic diagram of the manufacturing steps of the biocomposites.

The biocomposites containing different weight percentages of fibres were produced. The thickness of the composites with one layer of nonwoven prepreg of 1065 g/m^2 and compression moulding pressure of 50 bar was found to be 1.00 ± 0.1 mm. Depending on the thickness of composite material required, a number of prepregs can be doubled to make the required thickness. The biocomposite samples were then cut to desired shape according to the standard for testing and evaluation. To ensure that all absorbed moisture was removed, the prepregs containing PLA and flax fibres were dried at 80°C under vacuum for 10 hours before final hot pressing. Fibre-matrix composition, the moulding temperature, and the moulding time were selected as process variables which are illustrated in Table 1.

Table 1: Process variables.

Levels	Factors			
	Fibre composition (%) PLA/Flax	Moulding temperature (°C)	Moulding time (min.)	Moulding pressure (bar)
1	60P/40F	180	05	50
2	50P/50F	190	10	50
3	40P/60F	200	15	50

2.3. Impact Strength. Izod impact tests were conducted on notched samples according to ISO 180: 2001 at room temperature using a pendulum of 4.2 J energy. The dimensions of the test specimen were 80 mm × 10 mm × 4 mm with 2 mm notched at the centre of the vertical edge. The direction of the blow in the Izod test was "edgewise parallel" with a striking velocity of 2.44 m/sec. Each value reported is the average of at least five tests, and the error bars correspond to plus or minus one standard deviation.

2.4. Fibre Volume Fraction and Void Content. Since the mechanical property of the biocomposites depends on their real fibre volume fractions, they were measured after the consolidation operation, in order to ensure the fibre-matrix ratio in the prepreg according to the initial selection of the fibre fraction by weight. Composites were dissolved in a solvent, dichloromethane (DCM), at room temperature by stirring to obtain complete dissolution of the PLA matrix. The fibre volume fraction and void content of the PLA/Flax biocomposites were determined using the digestion method

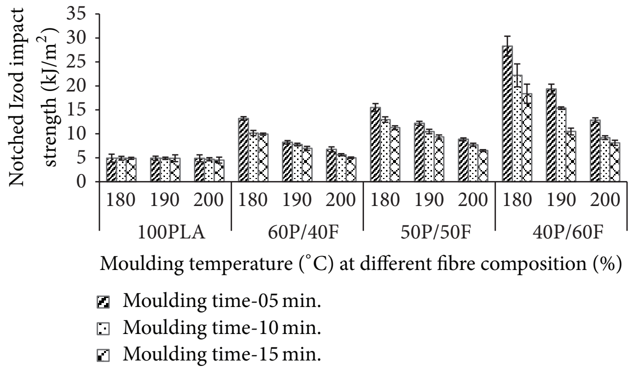


Figure 2: Effect of process variables on the notched Izod impact strength.

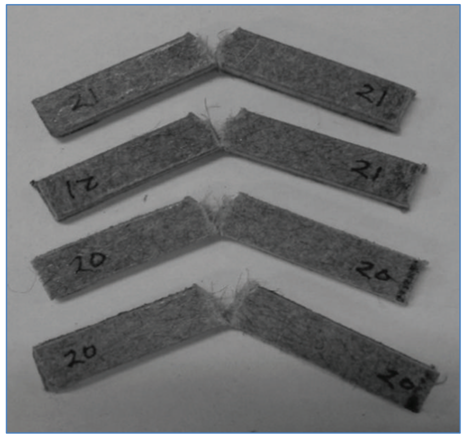


Figure 3: Photograph of the impact tested biocomposite samples with 60 wt% flax fibre.

in accordance with BS EN 2564: 1998. The void content by volume (V_o) was calculated according to

$$V_o = 100 - \left[W_f \times \frac{\rho_c}{\rho_f} + \left(100 - W_f\right) \times \frac{\rho_c}{\rho_r} \right], \quad (1)$$

where V_o is the void content as a percentage of the initial volume; W_f is the fibre content as a percentage of the initial mass; ρ_c is the specimen density, in grammes per cubic centimetre; ρ_f is the fibre density, in grammes per cubic centimetre; ρ_r is the density of the resin, in grammes per cubic centimetre.

2.5. Surface Morphology. Fractographic studies with scanning electron microscopy (SEM) were carried out in detail on the impact fracture surfaces and the surfaces of the PLA/Flax biocomposites. The samples were viewed perpendicular to the fractured surface. A SEM (Philips, XL 30) with field emission gun and accelerating voltage of 5.00 kV was used to obtain images for the biocomposite specimen. The nonconducting surface of the biocomposite was coated with carbon in Edwards coating system (E306A, USA) before being subjected to SEM in order to prevent electrical discharge.

3. Results and Discussion

3.1. Impact Strength. Impact strength of a composite is directly related to the toughness of the material. The fibres play an important role in the impact resistance of fibre reinforced biocomposites as they interact with the crack formation and act as stress-transferring medium. The notched Izod impact strength of the neat PLA and its biocomposites as the function of process variables is depicted in Figure 2.

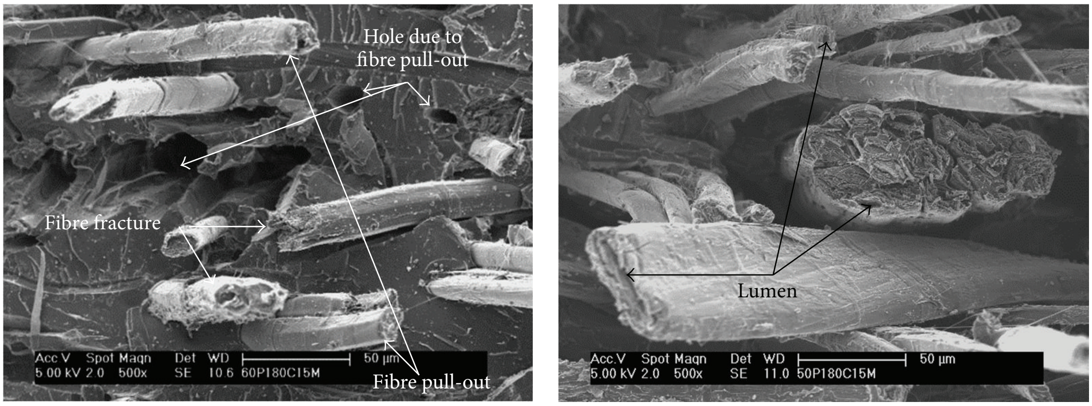
As can be seen, impact strength increased with increased flax fibre content for all moulding temperature and time. This was because as the fibre content increased, more interfaces exist on the crack path and more energy was consumed. In fact, the concentration of flax fibres would have increased with increased fibre content, which could lead to increased pull-out and also increased impact strength. From the thermal analysis, it was found that the crystallinity of the biocomposites decreased with increased fibre content [20]. In principle, the lower the crystallinity is, the lower the brittleness becomes. At the fracture surface, it can be seen that flax fibres were broken in the case of more brittle sample, whereas fibres were pulled out from the surface in the cases of less brittle samples.

The image of the impact tested biocomposites is illustrated in Figure 3, which shows that the samples were not completely separated into two pieces but flax fibres bridged the gap. This mode of failure was associated with high energy absorption [15]. In addition, examination of the impact fracture surfaces showed fibre pull-out due to the fracture of flax fibre during impact loading (Figure 4(a)). Good impact strength is mainly due to the energy absorption when the fibres are pulled out of the matrix. It is assumed that the weaker bonding leads to better impact strength than very strong bonding which can cause a sudden failure [21, 22]. This can also be explained by the voids in the biocomposites. Voids in the biocomposites increased with increased flax fibre content. Voids may be the cause for weaker bonding.

It was found that the notched Izod impact strength of the 60P/40F biocomposite was 13.2 KJ/m^2 at 180°C moulding temperature and 5-minute moulding time. The maximum impact strength 28.3 KJ/m^2 was obtained at 180°C moulding temperature and 5-minute moulding time with 40P/60F biocomposite. The neat PLA (100 PLA) shows very low impact strength (approximately 5 KJ/m^2), and it is not significantly affected by the moulding temperature and time. So it can be seen that addition of 40, 50, and 60% of flax fibre content increased the impact strength by about 170%, 216%, and 477%, respectively, at 180°C moulding temperature and 5-minute moulding time.

It was also observed that the impact strength decreased with increased moulding time and temperature. It might be that the longer period and higher temperature of moulding increased the crystallinity, that is, brittleness of the composites, resulting in reduced impact strength.

3.2. Fibre Volume Fraction and Void Content. The void contents of the biocomposites are illustrated in Figure 5. It is clear that the voids in the biocomposites increased as the



(a) (b)

Figure 4: SEM micrograph of the impact fracture surface of the biocomposite. (a) The fibre fracture and fibre pull-out. (b) The lumen in the flax fibre.

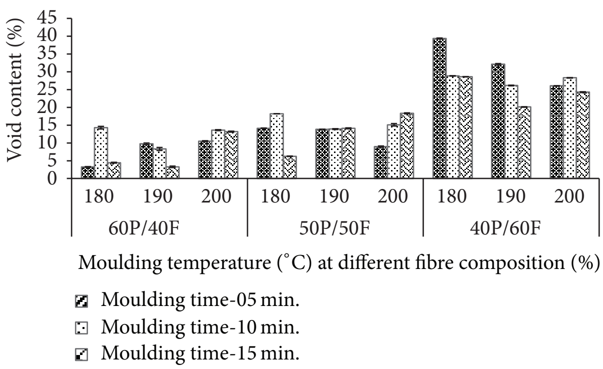


Figure 5: Effect of process variables on the void content of the biocomposites.

nominal fibre fraction by weight increased. In general, the voids are closely related to the processing conditions because they can be formed by gases which may be generated in the thermal process. Since this study adopted thermoplastic PLA and there are few possible sources of gas, the voids may be formed due to the discontinuous resin matrix inside the composites, resulting from the uneven distribution of the PLA fibres or their failure to form a continuous phase in the biocomposites.

The elementary flax fibres consist of a primary cell wall, a secondary cell wall, and a lumen. The lumen is an open channel in the centre of the fibre, and it can be as small as 1.5% of the cross-section of the fibres [23]. The size of the lumen mainly depends on the maturity of the fibres. Generally the lumen size (cross-section of the lumen) is larger for immature fibre and smaller for matured fibres. The lumens can be observed in the fracture surface of the biocomposites and are shown in Figure 4(b). These lumens also act as a void portion. Therefore, the flax fibres themselves are carrying the voids naturally. It might be the cause for increased voids with increasing flax fibre weight percentage. From Figure 5, it can be seen that there is no significant trend of void content with changing processing (moulding) temperature and time.

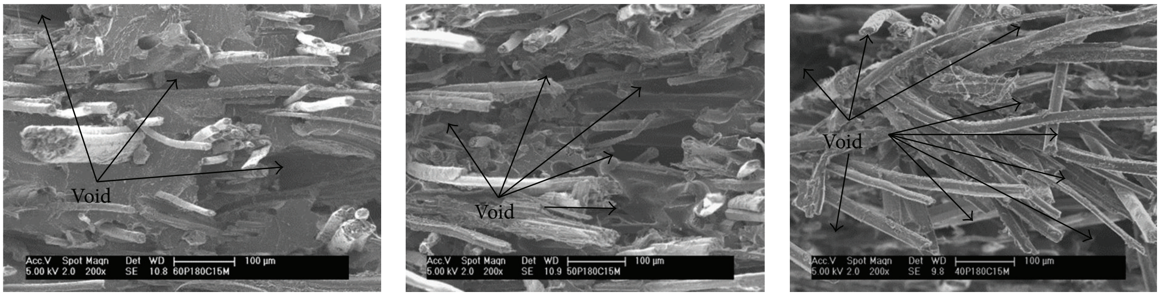
Figures 6(a)–6(c) represent the SEM micrographs of the impact fracture surfaces of the biocomposites. From the investigation, it can be seen that the amount of voids increases with increasing flax fibre content. The amount of voids is consistent with the quantitative analysis of the void content (Figure 5).

3.3. Surface Morphology. Figures 7(a)–7(c) show the SEM images of the surface of the PLA/Flax biocomposites with different flax fibre content. It can be seen that the number of pores gradually increases with increasing flax fibre content in the composites, and a large number of pores are clearly visible in the biocomposite with 60% flax fibres (40P/60F) (Figure 7(c)). In a previous study, it was found that the water absorption of the biocomposites increased with increased flax fibre content [24]. The increasing trend of water absorption may be because of increasing void content through which water may ingress into the materials.

It was also found that the water absorption of biocomposites increased with increased moulding temperature and time [24]. A large number of pores were created between the fibre and matrix interface due to the fibre degradation at higher moulding temperature and time. The pores act as passage for water into the biocomposites, and it may be the cause of higher water absorption. This indicates that it is very important to control the moulding temperature and time to decrease the water absorption of biocomposites when the fibre content is higher. The higher fibre content is desired in biocomposites to achieve good mechanical properties, and in this case, it is important to control the moulding temperature and time for decreasing water absorption. Therefore, the moulding temperature and time are recommended to be about 180°C and 5 minutes for manufacturing PLA/Flax biocomposites. These processing conditions are also recommended for the high tensile and flexural properties, reported in the previous paper [20].

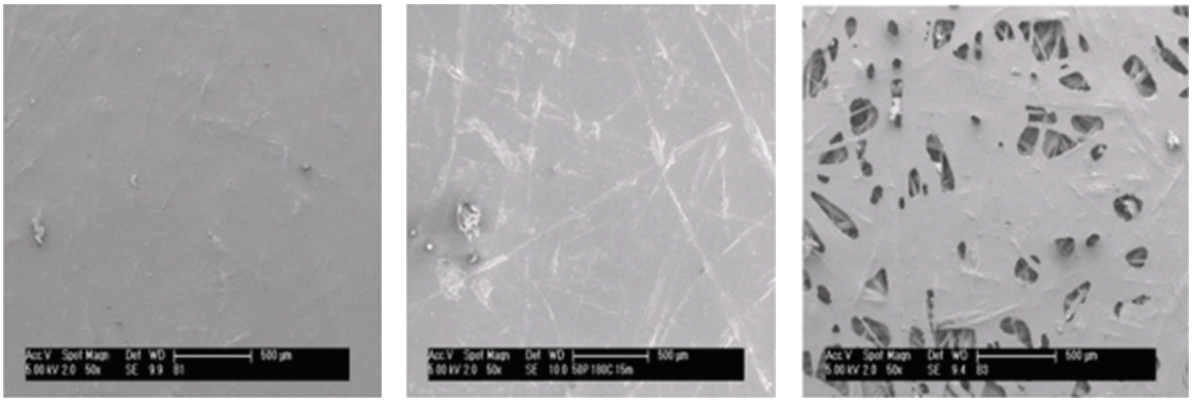
4. Conclusions

This paper reports the mechanical, physical, and morphological properties of flax fibre reinforced PLA biocomposites



(a) (b) (c)

Figure 6: SEM micrographs of impact fracture surface of 40% (a), 50% (b), and 60% (c) flax fibre reinforced composites.



(a) 60P/40F (b) 50P/50F (c) 40P/60F

Figure 7: SEM micrographs of the biocomposite surfaces with different flax fibre content.

based on air-laid nonwoven web forming method. Factors including flax fibre content, moulding temperature, and moulding time were investigated. Flax fibre content is the most significant factor influencing the physical and mechanical properties of the biocomposites. It is found that increasing flax fibre content in the biocomposites increases the mechanical properties, and the maximum notched Izod impact strength is 28.3 KJ/m^2. The notched Izod impact strength increased with increased flax fibre content, but it decreased with increased moulding temperature and time. The void content of the biocomposites also increases with increasing flax fibre content. The best processing conditions for the highest PLA/Flax biocomposite performance according to notched Izod impact strength was 60% flax fibre, 180°C moulding temperature, and 5-minute moulding time. 180°C moulding temperature and 5-minute moulding time are also recommended as the processing conditions for the highest PLA/Flax biocomposite performance according to the tensile and flexural properties reported in the previous paper [20].

Conflict of Interests

The authors of the paper have no direct financial relation or any other conflict of interests related to the paper with the company mentioned in this paper.

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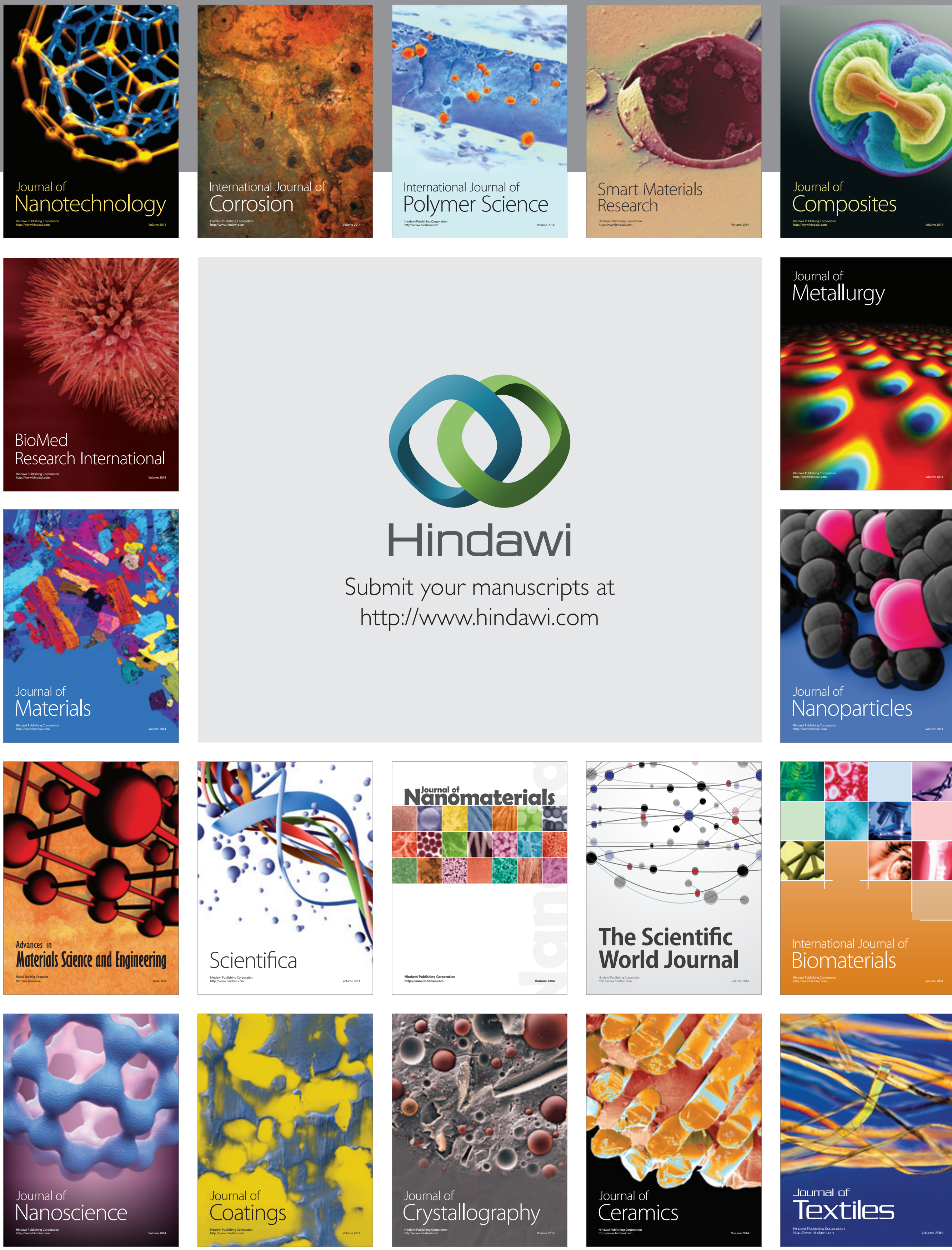
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