

# SUSTAINABLE BIOCOMPOSITES FOR FOOD PACKAGING APPLICATIONS

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## ABSTRACT

Biocomposites produced from renewable biomass were investigated for suitability in food packaging applications. Poly(lactic acid) was compounded with 10 – 30 wt. % hemp hurd through extrusion and injection moulding to produce rigid plastic biocomposites. In addition to their cost-effectiveness and equivalent mechanical properties, the biocomposites were particularly effective as antimicrobial materials with the inclusion of silver nanoparticles. A 16% cost margin was achieved in the biocomposites with additional antimicrobial capabilities. Through further research, development, and supply chain management, economic and environmental sustainability can be achieved in production.

## KEYWORDS

Biodegradable, antibacterial, biocomposite, thermomechanical, injection moulding, electron microscopy.

## INTRODUCTION

Bio based economies are burgeoning throughout the world and with growing support from federal governments and international collaborations, a sustainable bio-based future can be envisioned. Programmes have been developed within North America, Europe and the Asia Pacific on greater usage of biomass and development of sustainable materials for commercial applications (Salentijn et al., 2015). With only 3% of biomass utilised worldwide, there is a huge opportunity for using lignocellulosic biomass as precursor for organic macromolecules, bioenergy, and for biofuel development.

However, the use of biomass has taken a back step since the industrial revolution and with the emergence of fossil fuels, the supply chains for biomass have not received attention for solving contemporary world problems of fuel, energy, food, and sustainability. To develop sustainable, well-designed supply chains, innovative business models are required, with strong government support and involvement of end users.

A key end use of biomass is for development of polymers and macromolecules and fillers in existing plastics, with food packaging as the end market. In packaging, the rising demand for safe, minimally processed materials that can resist foodborne pathogens such as *Escherichia coli*, *Salmonella* spp. and *Listeria monocytogenes* is a major driving factor for innovations in food quality, freshness, and safety (Khan et al., 2015, Khan et al., 2014, Appendini and Hotchkiss, 2002).

Antibacterial packaging for increasing the shelf life of minimally processed food is replacing traditional materials and production methods. Many recent innovations, primarily antimicrobial agents for microbiological safety of food (Su Cha and Chinnan, 2004) are revolutionising the advancements in the packaging materials market. The use of antimicrobial agents in packaging materials is a solution for preventing growth of microorganisms at the surface, and realising increased shelf life.

The key objective for food packaging is containing food in a cost-effective way that satisfies industry/consumer requirements, maintains food safety, and minimises environmental impact (Marsh and Bugusu, 2007). The market for rigid thermoplastics is currently rising at a rate of 6.4% per annum and projected to reach a value of US\$472 B in 2016, with 30% of this market in food packaging, primarily as polyolefins, polyesters, ionomers, styrenics, nitriles, fluoropolymers, and cellulose (Coles et al., 2003). Plastic packaging is often contaminated with foodstuff; hence recycling is not economical (Siracusa et al., 2008) because of the increased extraction

steps. Hence, plastic packaging is landfilled every year, raising environmental concerns, and biodegradability has emerged as a functional requirement for packaging today.

Many biodegradable polymers are derived from renewable materials, which are inherently compostable (Tharanathan, 2003). Biodegradable polymers in food-contact articles include disposable cutlery, drinking cups, salad cups, plates, overwrap and lamination film, straws, stirrers, lids and cups, plates and containers for commercial food establishments (Siracusa et al., 2008). The increase in the development of biodegradable polymers has not evidenced a concomitant decrease in retail price. Moreover, some biopolymers show a lack of thermo-mechanical stability. This shortcoming has led new research on combinations of natural fillers with biodegradable thermoplastics to provide lower cost and higher durability. The focus of this study is hemp hurd, which is derived from the hemp plant after the bast has been removed for textiles and technical fibres.

An antibacterial packaging material based on poly(lactic acid) (PLA) and hemp hurd (HH) can be hypothesised to be superior to existing antibacterial alternatives because of lower raw material cost, effective antibacterial activity, fewer regulatory concerns, biodegradability, and environmental friendliness. Findings of toxicological studies have shown that PLA is safe and generally recognized as safe for use in manufacturing food-contact packaging because the level of lactic acid migrating to food from packaging containers is lower than the amount of lactic acid used in common food ingredients themselves.

In HH filler, inherent antimicrobial activity is the result of the heavy metal absorption capability of industrial hemp plant. Heavy metals uptake by hemp plant is an effect of soil and plant factors (Angelova et al., 2004), including heavy metal content, absorptive capacity of soil, redox conditions, organic matter, and pH of the soil (Alloway, 1995). Plant genotype is considered the most critical plant factor affecting heavy metal uptake (Hocking and McLaughlin, 2000). Angelova et al (2004) suggested that flax and hemp production is suitable for industrially polluted regions, as they remove considerable amounts of heavy metals from the soil through their roots, and can be used as potential crops for cleansing heavy metals. In addition to increasing soil quality, heavy metals bear no influence on future crop development and productivity (Baraniecki et al., 2001). However, it can be an issue of health risk if food packaging contains a high level of heavy metals that migrate to the food in contact.

In food packaging plastics, the heavy metals act through termination of bacterial or microbial activity through gradual damage to the bacterial cell walls, preventing nutrient uptake and reducing survival. The unique porous microstructure of the HH is beneficial for light weighting and controlled release of additives such as antimicrobial additives and self-healing chemicals.

The current study is a continuing effort to create a value added HH application in the food packaging industry. The increased utilisation of PLA in food packaging with certain shortcomings in thermomechanical properties and high cost can be alleviated with the use of a multifunctional filler. HH is such a filler that provides benefits in property retention, but at the same time strengthens the case for economic and environmental sustainability. In this study, the focus is on the utilisation of novel value chains for industrial hemp products.

## MATERIALS AND METHODS

A commercial grade of PLA was used as the polymer for the biocomposite. The HH, which is the biomass filler in the biocomposite was received from Ecofibre Industrial Operations Pty Ltd, Maleny, QLD, Australia as powder of 40 µm mean size. Glicidyl methacrylate (GMA) tert butyl perbenzoate (TBPB) were used for grafting the PLA for interfacial compatibilization. The 2015 estimated price and composition of the biocomposites are shown in Table 1.

Table 1. Cost and Constituents of Biocomposites

Raw materials	Price (USD/kg)	Weight Fraction (wt. %)
PLA (4032D)	3.00 - 3.80	68.5-88.5
HH*	0.20 - 0.40	10.0-30.0
GMA	6.20 - 7.60	1.0
TBPB	3.80 - 4.50	0.5
AgNP-loaded HH*	0.50 - 0.80	10.0-30.0

\* Either HH or AgNP-loaded or as-received HH is used in different grades

The approximate cost of raw materials produced using processing techniques similar to that of PLA for the 10-30 wt. % AgNP-loaded HH/PLA biocomposite is on an average 16% lower than that of the neat PLA. At increased loading of the biomass filler, further cost savings can be surmised.

A proprietary method of loading silver nanoparticles (AgNP) was used. Neat PLA, HH/PLA, HH/GMA/PLA and AgNP-HH/GMA/PLA were compounded on a laboratory-scale twin screw extruder with a speed of 40 rpm at a nozzle temperature of 175°C, followed by injection moulding as standard coupons on an injection moulder. X-ray diffraction was conducted using Cu K $\alpha$  radiation ( $\lambda = 0.154\text{nm}$ ). A scanning electron microscope (SEM) was used to image the structure and morphology of the blends. The macro- and micro-structure of the hemp hurd featuring the porous structure is shown in Figure 1.

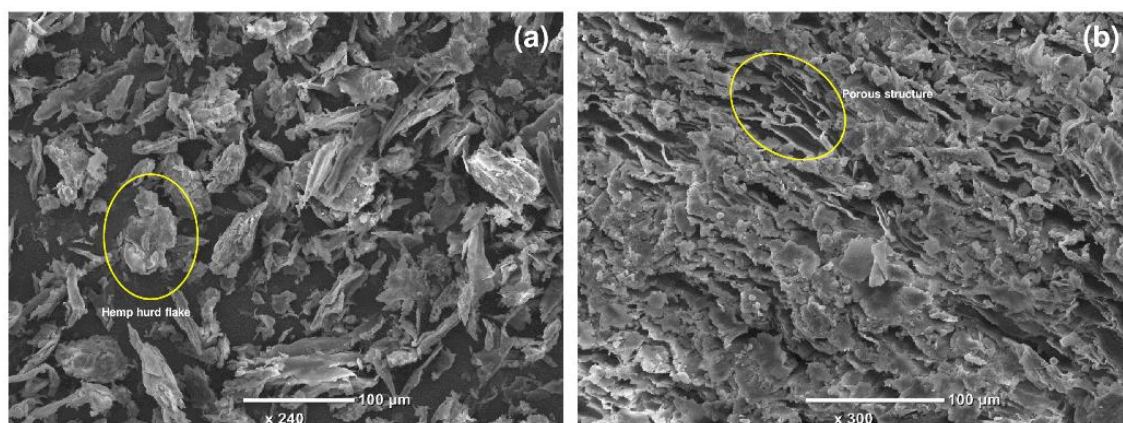


Figure 1. Hemp hurd as (a) ground, and (b) morphology.

Fourier transform infrared (FT-IR) was used as a spectroscopic method of chemical analysis. All tests were conducted with at least a duplicate and according to international testing standards. The antibacterial efficacy of the biocomposites was determined on *E. coli* (ATCC # 25922), through bacterial cultures maintained on nutrient agar slopes. The antibacterial performance of the HH powder was investigated according to ASTM E2149-10 standard. The migration testing for plastic materials was conducted according to guidelines of the European Council Directive 82/711/EEC with two food simulants, i.e., 3% (v/v) aqueous acetic acid and 95% (v/v) aqueous ethanol. This selection encompasses all alcoholic, aqueous, and acidic foods. The elemental migration from the biocomposites in the aforementioned food simulants was conducted at 40 °C and 70 °C in an oven at specific time intervals, followed by analysis on an inductively coupled plasma optical emission spectrometer (ICP-OES).

## RESULTS AND DISCUSSION

The AgNP loading on the HH was confirmed through XRD analysis on the AgNP-loaded HH and the PLA and unfilled HH counterparts. The deconvolution of Ag planes in the XRD patterns of AgNP-loaded HH supported the presence of Ag in the filler. The mechanical properties of the 10 – 30 wt. % AgNP-loaded HH are near equivalent to that of neat PLA, as confirmed from previous studies by the authors. The AgNP loading in HH showed no specific influence on mechanical behaviour, as the strength and modulus of the AgNP-loaded HH/PLA were nearly similar to those of the as-received HH/PLA biocomposites.

The addition of HH and GMA often affect the recrystallization of the base polymer, with the HH acting as a nucleating agent, accelerating recrystallization, a desirable effect for realising faster production cycles. The retention of the T<sub>g</sub> (~3.5% change), lowering of crystallisation temperature (~8 °C), and moderate change in melting temperature (~4.5 °C) with respect to the neat PLA preclude the need for heavy re-engineering of the processing and production of these biocomposites for commercial use.

The antibacterial activity was assessed against the gram negative *E. coli*, showing a distinct benefit of adding AgNP to the HH. The enhanced antibacterial activity was confirmed through a viable cell counting method of determining the colony forming units, and the AgNP-loaded HH/PLA composites showed a greater resistance to the formation and survival of *E. coli* at five successive concentrations, each increasing by an order of magnitude.

Table 2 shows the migrated amount of heavy metals from HH in food simulants at 70 °C for 48 hours. The migration values of heavy metals is practically negligible, and bears no health risk to food in contact. Thus, using HH, which is loaded with heavy metals in HH/PLA biocomposite does not have any effect on food safety because of high retention within the HH structure and consequent low migration to food.

Table 2. Migration of Heavy Metals in Food Simulants at 70°C after 2 Days

	Absorption (mg/kg)			
	Ni	Cd	Pb	Cr
Acetic Acid (3% v/v)	0.013	0.002	0.004	0.199
Ethanol (95% v/v)	0.012	0.002	0.003	0.147

Many heavy metals are capable of imparting antimicrobial activity in food packaging plastics. However, Ag as a nanoscale additive has emerged as an effective solution for a diverse range of plastics for this purpose. However, the main concern with the usage of nanoscale additive in plastics without encapsulation is the ready migration into foodstuffs, which causes adverse toxic effects on human physiology. The encapsulation of AgNP in HH specifically is a way of precluding excessive nanoparticle migration.

## CONCLUSIONS

Poly(lactic acid) was compounded with hemp hurd and injection moulded as a biocomposite. Silver nanoparticles were introduced into hemp hurd to achieve additional antimicrobial activity. The mechanical and thermomechanical properties of the biocomposite with and without the silver nanoparticle loading on hemp hurd were comparable. The heavy metal and silver migration from the biocomposites into food simulants tested at standard conditions was determined to be within the safe limits as prescribed by regulatory authorities. Overall, a cost-effective biocomposite was produced from a biomass-based renewable resource and combined with a biopolymer, will provide a commercially viable, biodegradable solution for food packaging.

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