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SUSTAINABLE FURNITURE PANEL COMPOSITES FROM FORESTRY AND FOOD INDUSTRY BY-PRODUCTS IN AUSTRALIA.

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Abstract:

This paper presents results of research into polypropylene based wood plastic composites reinforced with food industry and forestry by-products, identified as being particularly abundant in Australia but underutilised, viz. macadamia shells, pine cones and eucalyptus capsules. The present study considers and explores the suitability of these materials for high-moisture environment furniture panel applications. Results are presented for the relevant physical and mechanical properties and are compared with a conventional wood plastic composite utilising radiata pine as the filler.

The water absorption and swelling were generally lower in the forestry and food industry by-product composites than in the conventional radiata pine composite with the best results being obtained for the macadamia nut shell composite. The mechanical properties were however poorer than those of the conventional wood plastic composite. Nonetheless, it is considered that the forestry and food industry by-product composites do provide a viable material and have the potential to become a sustainable replacement option for high-humidity environment furniture panel composites. This would provide much better utilisation of these currently undervalued agricultural waste resources.

Key words: Recycled polypropylene, sustainable composites, macadamia shells, pine cones, eucalyptus capsules.

1 INTRODUCTION

In 1987, in the Brundtland report, the World Commission on Environment and Development [WCEAD] ("'Brundtland Report" Our Common Future: Report of the World Comission on Environment and Development ", 1987), defined sustainable development as “development that meets the needs of the present without compromising the ability of future generations to meet their own needs”. Subsequently, in the 1990s, the “triple-bottom-line” principle emerged, focussing on the equal importance of profits, planet and people, resulting in truly “sustainable design” (Elkington, 1998).

A design approach aligning well with the triple-bottom-line approach is the “product life cycle approach” which particularly focuses on the “planet”, or environmental sustainability aspect of a product. Its intention is to minimize ecological harm at every stage of the product’s life from “cradle to grave”. Even better is the “cradle to cradle” [C2C] approach (McDonough & Braungart, 2003). True application of cradle to cradle principles includes reusing all product material components within the product life-cycle, including the product itself at end-of-life.

Another consequence of the increased awareness of environmental sustainability issues has been the introduction of a number of schemes to help the general public to discern the environmental sustainability of particular products. For example ecolabeling schemes have been introduced in many countries (www.ecolabelindex.com) around the globe. Most ecolabeling programs tend to be voluntary, although this is likely to change in the future. The purpose of an ecolabel is to identify for the consumer the overall environmental performance of a product (i.e. goods or services) within a product category based on life cycle considerations (Eiderstrom, 1988).

The generation of waste from human production activities is a not insignificant burden on the environment. For example, most food and forestry industry activities result in large amounts of by-products that are often treated as waste and sent to landfill. In Australia, the macadamia nut industry generates as much as 25000 tonnes of macadamia shells annually (*Industry information & news: Statistics*, 2010) while just one operator in the pine tree silviculture industry (SeedEnergy Pty Ltd) produces 150 cubic meters of empty pinecones (Andy Cameron, Manager Pine Seed Production, SeedEnergy Pty Ltd, personal communication, October 19, 2009) annually. Currently, these by-products are used in applications such as garden mulch, in ground form as animal filler, or incinerated to fuel boilers. While this avoids their disposal in landfill, which is prohibitive due to their sheer volume, it is none the less poor utilisation of a potentially valuable resource.

In keeping with C2C principles, one approach to minimizing or even eliminating the waste burden is to find alternative uses for such by-products. These by-products are perfectly suited to the manufacture of wood plastic composites, as they come clean and dry after processing, and have excellent physical properties when exposed to high humidity environments, particularly when compared to softwood. This makes them suited to applications such as panel furniture in high moisture environments, including kitchen and bathroom sink countertops or drawers where dimensional stability and swelling problems are often an issue.

To this end, this paper examines the development of sustainable composite materials using readily available agricultural by-products as filler materials with a recycled polypropylene matrix, for use in furniture in high-moisture environments. Three sustainable composite materials were produced, using macadamia shell, pine cone and eucalyptus capsule fillers, respectively. These materials were compared with standard pine-wood-filler based particle board and unfilled polypropylene. The results show that the new fillers provide panels with performance comparable with commercially available panel materials.

2 SUSTAINABLE COMPOSITE MATERIALS

The term “composite material” implies a combination of two of more materials differing in form to achieve a particular function. One of the most important qualities of a composite material is that it can obtain a value of a given property which is not obtainable by either of the combined components on their own. As the term indicates, a composite material is one which is different from common homogeneous materials; e.g. metal, plastic. A composite material is instead heterogeneous (Kim, 2006); e.g. wood plastic composites. There are several viewpoints to define *sustainable* composite materials, and they mainly depend on the final application or treatment that the material will have. Some consider sustainable materials biodegradable, others think of them as consuming less energy to produce, while yet others think of them as recyclable or upcyclable material.

Consideration of materials should begin at the earliest stage of the design process, with selection made in the context of how the product will be used, and what performance characteristics are required (Baillie, 2004). Background on wood-plastic composite materials and on each of the fillers used in this study is presented below.

2.1 Wood Plastic Composites

Growing demand for wood plastic composites has led to continuous efforts to find alternative resources to wood. With increasing global population, the sustainable utilization of forest resources has been adversely influenced. Wood plastic composites have been used for several decades, but it has been only over the last decade that the use of plant-based fibres as an additive to plastics has accelerated, primarily due to improvements in process technology and economic factors (Ayrilmis, Buyuksari & Dundar, 2010; Dundar, Ayrilmis & Buyuksari, 2010).

Natural fibre fillers can be defined as cellulosic materials. They are the most abundant form of biomass and the form most likely to be used as reinforcement fibres, not only for ecological and economical reasons, but also because of their high mechanical and thermal performance (Baillie, 2004). They can be divided into four groups: leaf, bast, fruit and seed. Leaf and bast fibres are seeing increased usage as the reinforcement in composite materials, but seed and fruit production wastes are usually underused, mentioned by the Engineered Wood Products Association of Australasia [EWPAA] ("Facts About Particleboard and MDF," 2008). This study considers macadamia shells, pine cones and eucalyptus capsules.

2.1.1 Macadamia Nut Shells

Macadamia nuts come from plants belonging to the family of Proteaceae and are native to Australia. Macadamia nut trees grow best in sub-tropical climates where rainfall is gentle and plentiful during the spring flowering season and early autumn just prior to harvest (Cochrane, 2007; Fraser, 2007). Presently, Australia is the main commercial world producer of macadamia nuts, producing around 40,000 tonnes of macadamia nuts a year, out of a total global production of 100,000 tonnes (www.macadamias.org). They are also commercially produced in Latin American countries such as Brazil, Costa Rica and Bolivia, as well as in Hawaii and New Zealand.

The shells and other waste comprise almost 70% by weight of the macadamia nuts. They can be burned as a wood substitute in coffee roasting, ground to produce organic waste for gardening, used for mulch in the nut tree orchards, or used for chicken litter that, after use, returns to the orchard for use as fertilizer. They can also be used as a source of fuel (having a calorific value of 5,500 kcal/kg). It has also been reported that Macadamia shell powder can be used as a filler in the plastics industry (Woodroof, 1967).

The shell of the macadamia nut is hard and brittle. As a biological structure, the nut shell is highly optimized and efficient in terms of strength and toughness due to ecological evolution and selection (Wang, Zhang & Mai, 1995). Nut shells are known to have approximately the same fracture toughness as common ceramics and glass, and when compared on the basis of specific strength or modulus (strength or modulus divided by density), macadamia nuts outperform these materials due to their low density. The macadamia nut shell is a cellular solid with relatively low density and high strength, but their structure is reasonably isotropic and uniform, very different from that of trees.

The strength of the macadamia nut shell is naturally a compromise between attaining high strength and achieving isotropy as required by the need to provide all-round protection to the seed (Wang et al., 1995). The main components of the shell are lignin (47%), cellulose (25%), hemicelluloses (11%) and ash (0-2%). The shells have a bulk density of 680 kg/m3, and a moisture content of around 10% (Toles, Marshall & Johns, 1998).

Only very limited work has been carried out on the use of macadamia shells in composites materials (Ramirez & Wechsler, 2009), with the only product identified from the literature being Husque, a composite based on pulverized macadamia shells bonded with resin (Cochrane, 2007; Fraser, 2007)

The macadamia shells used in this study were supplied by Macadamia Processing Co. Limited (www.mpcmacs.com.au), located in Lismore, NSW, Australia.

2.1.2 Radiata Pine Cones

Australia has 122,400,000 planted hectares of radiata pine trees (www.fao.org). This species, native from California, has been successfully introduced in different areas worldwide, including Chile and Australia. It constitutes one of the most important potential raw material resources for the production of pulp, paper, and building materials. In Australia, the silviculture company, Energy Seeds, produces 100 to 150 cubic metres of empty cones per year after seed extraction (Andy Cameron, Manager Pine Seed Production, SeedEnergy Pty Ltd, personal communication, October 19, 2009). There are also at least four other producers in Australia.

Pine cones, are not utilised to their full potential. Vast quantities of cones are produced annually throughout the world. The principal components of pine cones are cellulose (32.7%), hemicelluloses (37.6%) and lignin (24.9%), as well as phenol extractives (4.8%), phenol, and condensed tannins (Ayrilmis, Buyuksari, Avci & Koc, 2009; Buyuksari, Ayrilmis, Avci & Koc; Duman, Onal, Okutucu, Onenc & Yanik, 2009). The cones are collected, dried to facilitate seed release, and generally discarded, burned, or sold to make decorations. Attempts have been made to use the cones for compost, but their decomposition is slow and they are inferior in this application to bark.

Existing research on the use of pine cones includes the developments of panel boards from pine cones using phenol formaldehyde adhesive and polypropylene as matrices, respectively (Ayrilmis et al., 2009; Ayrilmis et al., 2010; Buyuksari et al.; Dundar et al., 2010).

2.1.3 Eucalyptus capsules

Australia is the main eucalyptus producer in the world, with 127,024,000 hectares planted. Eucalyptus trees have been introduced to many other parts of the world with Brazil leading with 5,000,000 hectares planted, and Chile with 160,000 hectares planted. They have also been introduced into Ecuador and Colombia. They provide the raw for very substantial industries, such as sawmilling, pulp, charcoal and others (Brooker, Slee & Connors, 2002).

Seed capsules are produced in large quantities from seed production industries. After the seeds are extracted, the capsules are used mainly as mulch and fuel, due to their high lignin and cellulose content. Their reuse in material applications has not yet seen significant development.

Each of the three fillers discussed above are considered to be potentially suitable as alternatives to wood filler in wood plastic composite panel production. This study examines their physical and mechanical properties and compares these with those of a wood plastic composite produced with radiata pine wood filler, and also with unfilled recycled polypropylene.

3 MATERIAL PREPARATION AND PROCESSING

3.1 Materials

Macadamia shells were obtained from Macadamia Processing Co. Limited (www.mpcmacs.com.au), Lismore, NSW, Australia. The shells were supplied dried and cracked. The pine cones and eucalyptus capsules were obtained from plantations in Concepcion, Chile. The wood fibre used to manufacture a comparison panel was a by-product from the door manufacturing industry. Recycled polypropylene was used as the matrix material. The material was a polypropylene homopolymer obtained from recycled sacks. It was provided as pellets and is normally used for raffia extrusion.

To prepare the filler materials for composite material mixing, the pine wood, macadamia shells, pine cones, and eucalyptus capsules were ground and sieved through a 40 mesh screen and dried for 15 hours at 100°C (Ayrilmis et al., 2010; Ballerini, Goycoolea, Medina, Núñez & Wechsler, 2005; Ballerini, Reyes, Núñez & Wechsler, 2005; Dundar et al., 2010; Wolcott, 2001). This was sufficient to reduce the moisture content to one percent. Fig. 01 shows ground and dried macadamia shells, prior to mixing.

FIGURE 01: Macadamia shells ground at 40 mesh and dried overnight at 100°C

3.2 Composite Material Manufacturing Process

The fibres and the polypropylene were mixed in a Haacke Polydrive internal rotating mixer. First, the polypropylene was put into the mixer at 75 rpm, at a temperature of 165°C for two minutes. This was followed by adding the fibres and rotating them for another 3 minutes, for a total mixing time of 5 minutes. Fig. 02 shows the mixer used for material manufacture.

FIGURE 02: Laboratory mixer for plastic materials

The mixed samples were then pressed in a water cooled hot platen press with a 20 cm by 20 cm platen capacity. Each batch was pressed using a temperature of 170 Celsius degrees [°C] for 15 minutes, and then cooled while the samples were still under compression. The average target thickness of the samples was 3 millimetres [m]. The densities of the panels varied from 891 to 1053 kilograms per cubic metre [kg/m^3]. Fig. 03 shows the hot platen press with cooling system used for pressing, heating and cooling the panels.

FIGURE 03: Hot plate press with cooling system

3.3 Composite Material Formulations

Three different panel material combinations were made: recycled polypropylene matrix with macadamia shells, pine cones and eucalyptus capsules filler, respectively. The filler ratio chosen in the present study is 60%. Extended work performed by Klysov and Wolcott (Klysov, 2007; Wolcott, 2001); used filler ratio of 60% which has been successfully applied in Wood plastic composites available in the market. Properties of each were compared with a panel manufactured from recycled polypropylene with pine wood filler and another which was 100% recycled polypropylene. TABLE 01 shows the make-up of each of the five panels prepared.

TABLE 01: Ratio of raw material and recycled polypropylene (PP) used for panel manufacture.

Panel type	Recycled polypropylene based panels		
	Filler	*Filler ratio (*wt %)*	*PP ratio (*wt %)*
A	Macadamia shells	60	40
B	Pine cones	60	40
C	Eucalyptus capsules	60	40
D	Radiata pine wood	60	40
E	N/A	0	100

*Weight Percent [wt%]

3.4 Physical and Mechanical Testing

Tests were carried out on each of the composites to determine relevant physical and mechanical properties. Three to five samples were used per composite for each property evaluation test. Each test was carried out to the relevant American Society for Testings and Materials International [ASTM] standards (Ballerini, Bustos, Núñez & Wechsler, 2008).

The physical properties examined were density, moisture content, thickness swelling (TS) and water absorption (WA). These tests were carried out according to the ASTM 1037 specifications, with test sample sizes of 5 cm × 5 cm ("ASTM D 1037 - 99, Standard Test Method for Evaluating Properties of Wood-Base Fiber and Particle Panel Materials," 1999).

The density was measured using a Ludlum 4417 density profilometer. The moisture content was determined from the measured weight loss after drying the samples for 24 hours at 100°C. Thickness swelling was determined from the average of the measured change in thickness at four locations on each sample after immersion in water for 24 hours. Water absorption was determined from the measured weight gain of the panels during the 24 hours immersion period ("ASTM D 1037 - 99, Standard Test Method for Evaluating Properties of Wood-Base Fiber and Particle Panel Materials," 1999).

Flexural testing was conducted using the three point bend test according to ASTM 638 to determine the modulus of elasticity (MOE) and modulus of rupture (MOR). Tensile testing was carried out according to ASTM 246 using a dog-bone sample to determine the tensile strength. Both the flexural and tensile testing were conducted using an Instron 4468 universal testing machine. Impact testing was conducted per ASTM 256 using the IZOD impact strength testing method. Five replicate samples of each composite were used for each test.

4 TEST RESULTS AND DISCUSSION

4.1 Physical Properties

The results for the density, moisture content, thickness swelling and water absorption of the composite panels and also the unfilled polyethylene are presented in Tab. 02. The results for moisture content, thickness swelling and water absorption are also shown in Fig. 04.

TABLE 02: Density, moisture content, thickness swelling and water absorption of recycled polypropylene based panels

Panel type	Property			
	Density (kg/m3)	*Moisture content (*wt %)*	*Thickness swelling 24 hours (%)*	*Water absorption 24 hours (*wt %)*
A	1022	3.4	5.3	4.4
B	990	3.6	7.5	11.9
C	1053	2.8	7.0	6.6
D	1049	3.0	7.8	8.4
E	891	0.22	1.0	0.5

*Weight Percent [wt%]

The densities of the composites with the four different fillers are 10-20% higher than the density of the matrix polymer and are typical of commercial wood plastic composites (Klysov, 2007). Only minimal differences are evident amongst the four composites. The moisture content of the composites ranges from 2.9-3.6 wt% compared with 0.2% for the unfilled recycled polypropylene matrix material (Panel E).

The thickness swelling results for the composites are also very different to that for the unfilled recycled polypropylene, with values ranging from 1% for the unfilled matrix material compared with 5-8% for the composites. The values for the composites are all in the expected range for wood plastic composites (Klysov, 2007). Of the composites, the macadamia shell filled material (Panel A) had the lowest swelling of 5.3%. The remainder gave values of 7-8% but both the pine cone and eucalyptus composites (Panels B and C) showed slightly lower swelling than the standard radiata pine wood panel (Panel D).

The water absorption of the composites was 4-12 wt% compared with 0.5 wt% for the unfilled matrix material. Amongst the composites, the macadamia shell filled composite had the lowest water absorption of 4.4 wt%, which was approximately only half the value obtained for the standard radiata pine wood composite of 8.4 wt%. The water absorption for the eucalyptus capsules composite of 6.6 wt% was also substantially lower than for the standard radiata pine wood composite, but the value for the pine cones composite of 11.9 % was 40% higher than for the standard radiata pine wood composite Generally an absorption rate below 10% is considered to be low (Klysov, 2007) and both the macadamia shell and eucalyptus capsule composites are well below this limit.

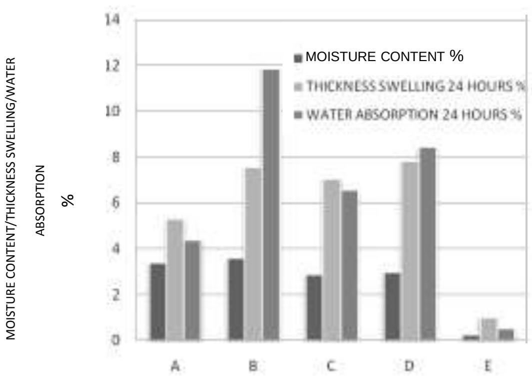

FIGURE 04: Moisture content, thickness swelling and water absorption of the samples

4.2 Mechanical Properties

The mechanical properties test results are given in Tab. 03. The MOR and tensile strength of the composites were 19-40 Mega Pascal [MPa] and 6-9 MPa, respectively, and were substantially lower than the values obtained for the unfilled polypropylene of 61 MPa and 26 MPa, respectively. The values for the composites made from macadamia shells, pine cones and eucalyptus capsules were also lower than those for the standard radiata pine wood composite, with the latter having values of 40 MPa and 9 MPa, respectively. Of the forestry and food industry by-product composites, the macadamia shell filled material had the lowest MOR (19 MPa) but the highest tensile strength (7.5 MPa) while the eucalyptus capsule material had the highest modulus of rupture with the pine cone material having the lowest tensile strength.

The modulus of elasticity was generally higher for the composites than for the unfilled polypropylene for which a value of 1.6 Giga Pascal [GPa] was obtained. The standard radiata pine wood composite had the highest modulus (2.8 GPa). The values for the eucalyptus capsule (2.0 GPa) and macadamia shell (1.9 GPa) composites were approximately 30% lower while the modulus for the pine cone composite was the same as that of the unfilled polypropylene.

The impact values for the forestry and food industry by-product composites were approximately 40-50% lower than the values obtained for the unreinforced polypropylene (1.5, 1.5 and 1.8 Kilo Joule per square meter [kJ/m^2] for the macadamia shell, eucalyptus capsule and pine cone composites respectively compared with 2.9 kJ/m^2)for unfilled polypropylene). However, no reduction in impact strength was observed for the standard radiata pine wood composite for which a value of 2.9 kJ/m^2 was obtained. The result for the radiata pine wood panel is surprisingly high; however the results for the other composites are as expected since the additional of filler should reduce impact strength.

TABLE 03 Modulus of rupture, modulus of elasticity, tensile strength and impact test results of samples

Panel type	Property			
	Modulus of Rupture (MOR) (MPa)	***Modulus of Elasticity (MOE) (GPa)***	***Tensile Strength (MPa)***	***Impact (kJ/m^2)***
A	19.4	1.9	7.5	1.5
B	21.1	1.6	5.9	1.5
C	27.7	2.0	6.4	1.8
D	39.5	2.9	9.3	2.9
E	60.9	1.6	26.0	2.9

Comparing the mechanical performance of the composite materials, it is possible to conclude that radiata pine wood filler has the best performance in a polypropylene matrix, followed by macadamia shells, eucalyptus capsules, and then pine cones. This can be associated with the shape of the fibre, as is discussed in the following section, as well as the amount of extractives in each filler. The extractives are non structural organic and inorganic materials present in the fillers, which modify their colour, smell, and calorific power. Extractives are a heterogeneous group of composites that can be found inside the cells, between the cells, or even in the interior of the cell walls. The content of the extractives influence density of the fillers (Gutierrez & Baonza, 2009).

Pine cones and eucalyptus fibres are much more porous than macadamia shells. However, they are less brittle, presenting better elasticity properties. Even though macadamia shells have higher amounts of lignin, which has been recognized as a binder, they have much lower amounts of cellulose than pine cones, as mentioned by Ayrilmis, Buyuksar, and Dundar (2009). In previous studies, it has been found that extractives in wood flour had a detrimental effect on the flexural properties of PP composites reinforced with wood flour, and as pine cones have higher amounts of extractives than wood, the pine cone composites would be expected to have a lower flexural modulus (Ayrilmis et al., 2009; Ayrilmis et al., 2010; Buyuksari et al.).

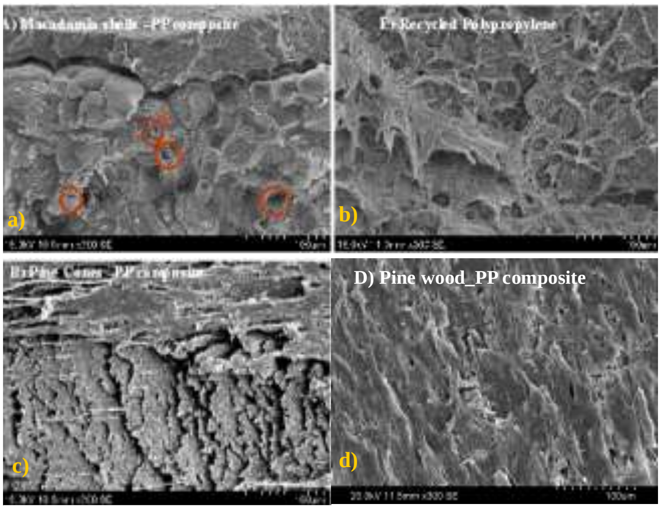

FIGURE 05 Morphological characterization of the a) macadamia shells-recycled PP plastic, b) recycled PP, c) pine cone-recycled PP and d) pine wood-PP composites.

4.3 Morphological Characteristics

The fibre-plastic interaction was examined on the surface of the composites using a Hitachi S3400I scanning electron microscope (SEM). SEM micrographs of the samples are shown in Fig. 05. As can be seen, the recycled polypropylene plastic seems to have a fluid and plasticized appearance (Fig. 05b) which can be clearly observed in the composite boards interacting with the agricultural by-products.

The macadamia shell filler panel gave the lowest MOR, which can be attributed to the shape and hardness of the shell fragments after milling, and their isotropic nature, as opposed to the nature of the fragments of the other fillers. This could affect the interconnections between the fibre and the matrix.

The pine cone and pine wood fibres are completely surrounded by the recycled plastic (see Fig. 05c and d). However, the direction of the fibres reveals an anisotropic orientation which affects the mechanical properties of the panel.

5 CONCLUSIONS

Truly sustainable panel materials are still in the development stages. This study has considered the use of agricultural crop by-products that are under utilised and therefore present the potential to be used in composite material panel fabrication. We have shown that the three new fillers considered in this paper, macadamia shells, pine cones and eucalyptus capsules all have acceptable properties for use as fillers.

The materials proposed and evaluated in this research present a new opportunity for panel furniture application. As they are composites, their mechanical properties can be improved and analysed by further research. Their physical properties showed that the newly proposed fillers are applicable in panel furniture for high humidity areas.

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