



# Sustainability Newsletter

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## Using Nature's Bounty

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Fundamentals of Plastics for a More Sustainable World

# Chairman's Letter

By **A. George Staniulis, Chair, Sustainability Division, Society of Plastics Engineers**

As that famous American philosopher Yogi Berra once said, "it's like déjà-vu all over again."

Once again, the plastic industry is in the environmental spotlight, and again, not in a good way. The primary culprits this time are discarded single-use plastics packaging polluting our oceans and landscape.

Perhaps we can recall a little bit of history. Back in 1987, the infamous Long Island garbage barge, the Mobro 4000, was traveling from port to port looking for a friendly locale to unload its cargo of 3,200 tons of municipal waste, much of it plastic.

What sparked this episode was that landfill space was at a premium in the north-east, thus expensive, and municipalities were exporting their trash to lower-cost destinations. First the barge went to North Carolina. After it was turned away, the barge went to Mexico with the same results, then to Belize, then back to Long Island where the garbage was finally incinerated.

Naturally this activity generated a lot of press, and the cry was raised that we were running out of landfill capacity. Landfills were reaching their maximum capacity and closing faster than new facilities were being commissioned. What was needed was to reduce the amount of waste going into landfills. Not a bad thing under any circumstance.

What became the bad boy in all of this was plastic. The poster child for this problem was single use EPS clam-shell containers utilized by the fast food industry. Sound familiar?

The plastics industry jumped in to protect itself by presenting loads of data showing that plastics packaging was the best packaging material environmentally, technically, and in every other way. However, once a public issue gets to this point, facts matter little. The populace just wants the problem to go away.

From the interactions that I had with a major packaging

manufacturer, they thought that the economic and technical benefits of the clam-shell container, notably a longer shelf life of the hamburgers, a cleaner product, it kept the food hot for take outs, etc. would carry the day with their customers. Boy, were they wrong.

As the landfill crises festered, loud voices were raised vilifying plastic's role in this situation. The hamburger sellers became concerned that the clam-shell was negatively impacting their ability to sell their product. It wasn't long before the clam-shell was discontinued irrespective of its economic and technological benefits.

One of the positive aspects of this situation was that it gave plastic recycling a big boost. Unfortunately this was not enough to save the clam-shell, which was a better product than what they ended up with, coated paper.

Now, 30 years later, we have a world-wide plastic pollution problem. The culprit again is single use packaging. Even though we went through this problem before, it seems to me that we are repeating the same mistakes in how we are addressing this issue: 1) by focusing too much on technological arguments favoring plastic packaging; and 2) by trying to deflect blame by positioning this issue as a disposal and littering problem. In my view, since the material we are talking about is plastic, it is our problem.

We need to make the problem go away - and quickly - otherwise many of our packaging products might end up like the clam-shell: obsolete. Developing collection and sortation technologies takes time. Converting the mindset of the populace to stop littering will take generations. As I am blessed with 20/20 hindsight, we should have considered these back-end issues when we started making these products. But then, who would have listened?

## Looking Forward

What can we do now? One way is to ban single-use plastic packaging - period. This will be effective, but it would not be my first option. Last year in Kenya, however, a law was enacted banning single use plastics bags with penalties of 4 years in jail and up to \$31,000 in fines for manufacturing, selling, or even using this product. This draconian approach seems to be working with a noticeable reduction of plastic pollution, thus encouraging a number of other African countries to consider similar legislation.

The European Union is also making noises to ban single-use packaging products.

Another option is to expand the bottle deposit programs to require a deposit on all plastic bottles, soda, water, milk, detergent, mouth wash, etc. The bottles, and, if need be, other plastic recyclables, would be returned to satellite redemption centers operated under the auspices of the resin and packaging companies. After all, following the cradle-to-grave model, this is their product and their responsibility. This approach would go a long way to address the collection, sortation, challenges. A little cumbersome, but very doable.

Finally, the packaging manufacturers themselves can mitigate this pollution problem by utilizing recycled plastics in their products or elsewhere in their operations. They can establish standards for cleanliness and consistency, and convey to the recyclers that if they meet these standards, they will buy their material. In doing so, they will create value for a problematic waste material.

We all know that nothing happens until someone sells something. I have confidence in our economic system that if there is a well-defined market for recycled plastic packaging, some bright entrepreneur will solve the problems necessary to satisfy that demand.

One last thought for the packaging companies: stating that your goal is make your packages recyclable, reusable, or compostable is not enough. No matter how recyclable you make your package, if it is not being recycled, it is not recyclable. The best thing that you can do to drive recycling is to buy recycled resin to manufacture your products where ever you can. |

### **Cover Image:**

“Tree-Based Console Substrate”. From the SPE Global Parts Competition, submitted by Ford Motor Company. The hybridization of cellulose fiber with long glass fibers in PP composites reduced the use of inorganic fibers in console substrate, leading to robust properties and significant weight savings.

*Photo courtesy of SPE.*

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# Why Not?



# Using Coconut Fiber and Shell as Functional Fillers in Polyolefin to Enhance Properties and Reduce Costs

By **Walter L. Bradley, Ph.D., P.E. & Steven W. Bradley, Ph.D., Distinguished Professor Emeritus, Mechanical Engineering & Professor of Entrepreneurship, Texas**

## Abstract

The goal of this research is to utilize the abundant agricultural waste from the 50 billion coconuts harvested each year as functional filler in polyolefin to enhance their mechanical properties, reduce their costs and create more environmentally friendly products.

## Introduction

Coconuts are an abundant renewable resource. Coconut trees that bear fruit (coconuts) are found on land that is within 20 degrees of the equator. Fifty billion coconuts are harvested globally each year. Most of the coconuts are cash crops for 11 million poor coconut farmers who each own several acres of land from which they harvest about 5000 coconuts per year that sell for ten cents each. The farmer opens the coconut husks to reach the coconut, which is a large nut (seed). The mature coconut's constituent parts illustrated in Figure 1 are: the fibrous husk on the outside, the hard coconut shell inside the husk, and a nutrient rich soft solid inside the shell called copra.

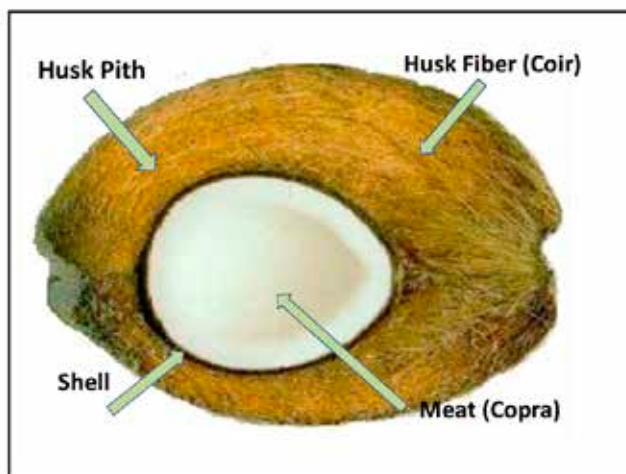


Figure 1. Constituent parts of coconut

The farmer opens the coconuts to remove the white paste called copra, which is a mixture of coconut milk and

coconut meal (fine particles that are carbohydrates and protein). The coconut milk is an amalgam of coconut oil and water. Currently, the highest value-added production from coconuts is primarily in the separation of coconut oil from the coconut milk. Alternatively, the coconut shell and husk have found limited commercial value and are, for the most part, considered a difficult-to-dispose-of waste product (Figure 2). The limited use of coconut shells from the 50 billion coconuts harvested each year is as a material for activated carbon filters or charcoal. Likewise, there is a limited market for coir (fibers) from the coconut husks in mattresses or seat cushions.



Figure 2. Pile of discarded coconut husk as waste.

The goal of this research is to (1) create polymeric composite materials that utilize coir fiber from coconut husks and (2) use fine powders made from coconut shells as functional filler in polyolefin. In doing so, we can enhance current materials' mechanical properties, reduce costs, and produce consumer goods that are more environmentally friendly such as decking, fencing, automobile parts and building materials.

## Physical and Mechanical Properties of Fibers (Coir) from Coconut Husks

The coconut husk's (see Figure 6) primary function in nature is to protect the coconut at the end of its 20m-25m fall from the coconut tree. A summary of the other functions in nature and properties of the coconut husk fiber (coir) that enable these functions are summarized in Figure 3. The husk is comprised of two constituents; namely, fibers (called "coir") that gives structural strength and 'pith' particles that act as a binder to hold the fibers together.

### Husk's function in nature

- Help nut survive impact after 60-80 ft drop
- Help nut avoid microbial attack
- Help nut survive forest fires
- High lignin (~40%) content is key

### Physical Properties of Coir Fiber

- Naturally burn resistance (high lignin)
- Excellent ductility (~25%) and formability
- Density ~ very low density (shell-high)
- Large diameter fibers (150-250  $\mu\text{m}$ )
- Excellent bending stiffness (EI)
- Durable in wet environments
  - Resistance to mold and microbial attack
- No problems with odor
- Moderate tensile strength and stiffness

Figure 3. Coconut husk's properties and function in nature.

Coir fibers are comprised of cellulose, hemi-cellulose, and lignin, all three constituents being common in woody materials. Coconut fibers have four relatively unique and very useful properties for end products.

Coconut fibers (and coconut shells as well) have a very high lignin content of ~40%. Most woody materials have a lignin content of 5-20%. The high lignin content of coir provides two benefits to the fiber (and therefore the husk). First, lignin is much more difficult to burn than cellulose or hemi-cellulose, making it more likely that a coconut can survive forest fires. This property of coconut husk fibers is also essential for safety for such applications as mattress filler or siding for houses. Second, lignin is resistant to microbial attack because the microbes cannot digest lignin. This is important in many applications because microbial attacks reduce durability and create disgusting odor (i.e., think composting of typical organic waste material from your yard, which is low in lignin and therefore susceptible to microbial attack).

A third critical property of the coir fiber in the husk is its low density and high ductility (~20-25%), allowing it to protect coconuts from fracturing on impact after their descent of 20-25m from their high "nests" in the coconut tree. This excellent ductility gives the coconut husk the capacity to absorb a large amount of energy on impact, protecting

the coconut inside the husk from breaking.. It also means that composite materials that use coir fiber will have good formability as well as good impact strength.

A fourth unique property of the coconut husk fiber is its microstructure, as seen in a scanning electron microscope (SEM) in Figure 4. The coconut fiber (coir) has an irregular honeycomb-like structure that gives the fibers a very high specific stiffness ( $E/\rho$ ) in bending.

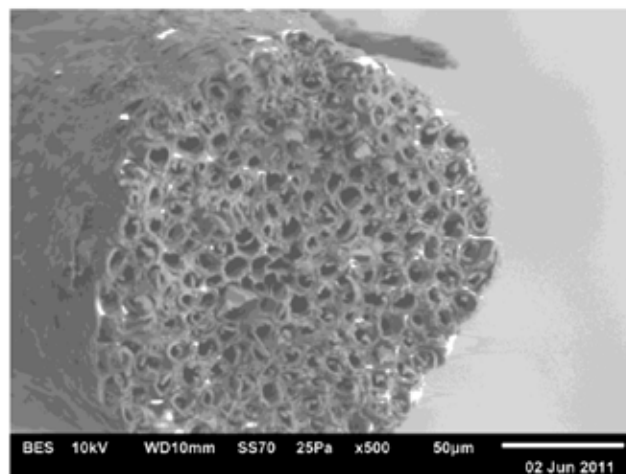


Figure 4. Scanning electron micrograph of cross-section of coconut husk fiber.

### Pathways from Husks to Finished Parts Made with Coir Fiber

Coir fibers from coconuts are blended with polypropylene fibers using a process called "air-laid, carding and needle punching" to produce a non-woven, flexible fabric composite felt. The coir fiber/PP mixture and resulting fabric is seen in Figure 5 along with fabric produced with the common PET/PE blend for comparison. Figure 6 shows coir and PP fibers before being mixed and after being air laid and needle punched in the SEM photo. The larger diameter fibers are the coir fibers while the smaller diameter fibers are the polypropylene fibers (PP), which melt and flow during compression molding at an elevated temperature and subsequently cooled to give a rigid part that has the shape of the mold. The production output from carding and needle punching is a flexible felt roll measuring 2 meters wide and 40 meters long as seen in Figure 7. The felt roll is a stage in the whole process which consists of: extracting coir (fibers) from coconut husks, bailing and shipping them, blending the coir fiber with



Figure 5. Comparison of tufts of coir (fiber) and propylene fiber (top) and tufts of PET and PE binder fiber (bottom). These are carded and needle punched for felt used to compression mold into trim parts.



Figure 7. (top) Tufts of coir fiber and PP made into a felt 2-meter wide by 30 meters long. (bottom) PP+PET felted and compression molded into a trunk lid cover.



Figure 6. Inside of coconut husk, tufts of coir fiber from husk and PP binder fiber, seen in scanning electron microscope (SEM) after being carded and needle punched.

polypropylene fibers to make a flexible piece of felt, die cutting pieces from the felt roll, and finally compression molding the die cut felt pieces into an interior panel part. An example of a compression molded trunk lid “trim” piece for an automobile is shown in Figure 7.

An advantage of the coir fiber/PP blend fabric, trade-named COIRFORM, is that the coir fibers are much stiffer in bending than PET fibers. The flexural modulus of COIRFORM increases with the increasing fraction of coir fiber used in the COIRFORM. The density can also be varied by varying the pressure applied in the compression molding process since the free space in any non-woven fabric composite decreases with increasing processing pressure and/or increasing pressing temperature. Because the coir fibers are both stiffer in bending and have a lower density (see Figure 4) than the PET fibers, the COIRFORM

will be much stiffer in bending (Figure 8a) at a comparable density.

The dramatic difference in flexural rigidity between a non-woven with PET and a non-woven with coir is due to the much larger diameter and “ragged” honeycomb structure. This difference can also be shown by comparing rigidity versus density for PET/PP non-woven fabric composite to the COIRFORM non-woven fabric composite in Figure 8b. The coir fibers significantly outperform the PET fibers in non-woven fabric composites, even at significantly lower densities. It is also worth noting that the coir fibers are much less expensive per pound than PET or other synthetic fibers and are obviously more environmentally friendly.

Products and parts can be formed from COIRFORM using thermoforming as well as compression molding. A small part that was made by thermoforming COIRFORM is seen in Figure 9. The interior panel for a truck cab in Figure 10 demonstrates the excellent formability that COIRFORM

has in compression molding.

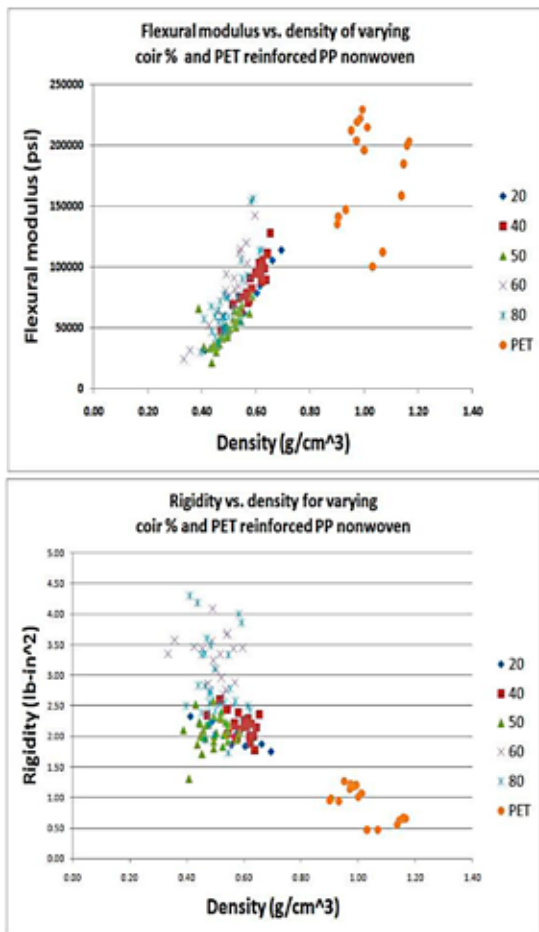


Figure 8. Flexure modulus (a) and rigidity (b) as a function of density for coir/PP vs. PET/PP.

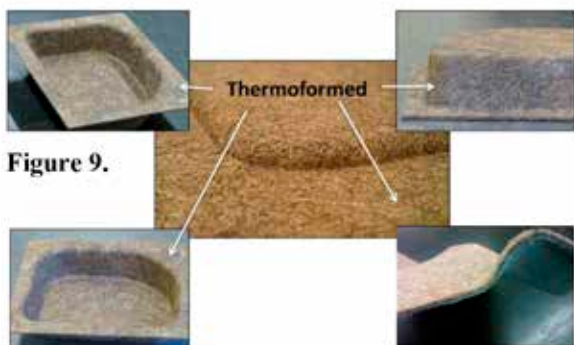


Figure 9.

Thermoformed part from felt with coir fiber and polypropylene.

### Successful Partnerships and Supply Challenges

We have greatly benefited from working with several major companies who were interested in using coir fiber

or coconut shell powder in the production of parts for their products. Working in partnership with Ford Motor Corporation, we demonstrated that specific car parts could indeed be made using a non-woven fabric composite comprised of coir fiber and polypropylene. The Society of Plastics Engineering recognized this "invention" of a very light but very stiff load floor panel in their electric car with a Materials Innovation award in 2012

The results of these collaborative development projects created some excellent opportunities that we were unable to actualize because we could not find suppliers capable of providing larger orders (i.e., 15,000 lbs.) with the same quality of cleanliness for husk fibers that we had received in small samples of 50-100 lbs that had been hand cleaned. For example, a major automotive company was able to make some trial door panels using coir fiber and polypropylene. The panels passed all of their tests and were their first choice for materials specification for this part. Unfortunately, this opportunity was missed because of the lack of an adequate supply chain for clean coir fibers at the time. Today, Dignity can provide a large supply of clean coir fiber as a byproduct to their coconut oil business.

We were invited to do a joint research project with one of the largest manufacturers of patio-furniture. They wanted to use natural fibers to make their cushions for their patio chairs. They did a whole battery of tests with

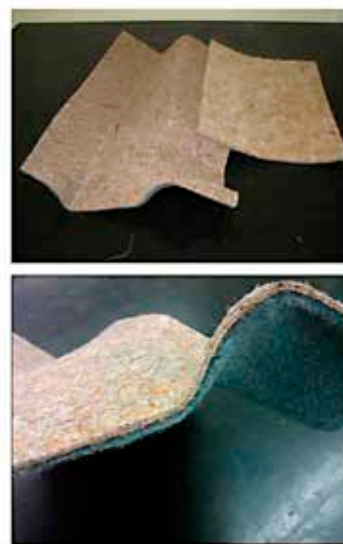


Figure 10. Truck cabin part for 18-wheeler using compression molding felt of coir fiber and polypropylene.

sample cushions made with coir fiber filler. The testing included burn tests and resistance to microbial attack with repeated moisture exposure. The manufacturer also ran compression fatigue tests of the cushions filled with coir fiber to see if there was any compacting of the cushions (permanent deformation) at 100,000 cycles. Coir fiber easily passed all the required tests. The furniture manufacturer had successful production runs to create cushions filled with coir fiber at two of their six plants spread around the United States. Unfortunately, the four older plants had air laid equipment that was unable to process coir fiber successfully due to the greater fiber stiffness than the fibers that they had been using. The company decided not to move forward as the upgrade of equipment at the four plants would have been cost-prohibitive and they did not want to have two different cushions for sale.

To summarize for coir fiber, the material has demonstrated valuable properties for finished products, but supply of the quality and quantity needed. We now have our own fully functional production facilities for coir fiber and coconut shell powder in the Philippines. We now turn to look at the exciting opportunities that coconut shell powder provides to enhance the mechanical properties of polypropylene and polyethylene in the next section.

### Pathways to Finished Parts Made With Coconut Shell Powder as Functional Filler in PE or PP

What are the unique physical and mechanical properties of the coconut shell (Figure 11) that provide interesting opportunities for its use as functional filler in polymers?

- High lignin content that makes it burn resistant and pest (and odor) resistance;



Figure 11. What is unique about the mechanical properties of coconut shell powder (CSP)?

- High density (1.2-1.3 g/cc) compared to 0.6 g/cc for hardwoods that are native (to the U.S.);
- High hardness that is the consequence of high density and possibly high lignin content.

The challenges that must be overcome in utilizing the outstanding properties of coconut shell powder as a functional filler in PE and PP are addressed along with the status in resolving each challenge.

*Filler size.* Could coconut shell with its excellent physical properties be produced in small enough diameters to meet common polymer filler standards? Yes, very fine coconut shell powder (20-200 microns diameter) can now be produced using a proprietary process that has already been successfully developed!

*Coating and bonding.* We needed to identify and create a chemical coating for coconut shell powder. The requirements were ease of application that bonds well with the coconut shell powder and with polyethylene, polypropylene and other engineering plastics to allow the hardness, stiffness and strength of the coconut shell powder particles to be effectively transmitted to the plastic matrix. Such a proprietary interfacial bonding agent has already been developed!

*Processing.* We developed processing parameters for a uniform mixture of coconut shell powder particles in pellets of PE and PP. Now, incorporating coconut shell powder into polyethylene, polypropylene, or other engineering plastic is easily accomplished with a twin-screw extruder.

*Environmental exposure.* We have examined the resistance to environmental factors such as UV radiation and moisture. A major toy manufacturer and potential partner determined for us that coconut shell powder acts as an UV inhibitor (as well as a mechanical property enhancer) reducing or eliminating the need for additional UV inhibitors depending on the projected life-time exposure.

*Microbes and odor.* The resistance to microbial attack and associated odor issues is established by the high lignin content organic material and the coconut shell powder has been confirmed for the use of in PE or PP.

*Product applications.* There are a wide range of products



(see Figure 12) that can be made by using PP or PE with 20-40 wt% coconut shell powder incorporated into the pellets prior to injection molding or extruding to consolidated PP or PE pellets with coconut shell powder. Some products such as children's toys need to have bright colors rather than the natural brown color that results when PP or PE have 20 wt% coconut shell powder incorporated in PP or PE pellets that will subsequently be injection molded or extruded. A company that makes toys paid to have various colored dyes incorporated into the pellets along with the coconut shell powder. These pellets were subsequently used for injection molding of pill cases of various colors, as seen in Figure 13. Clearly, various colored dyes can be incorporated in PE or PP pellets with coconut shell powder to give a wide range of color in the product.

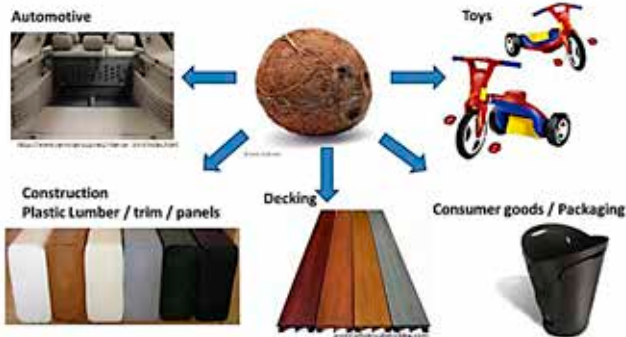


Figure 12. All the products seen in the figure have been made with polyethylene with 20wt% CSP.



Figure 13. Coconut shell powder can easily be dyed to give more pleasing colors than the natural brown.

**Mechanical properties.** Testing in our laboratory has established that there is high retention of ductility and Izod impact toughness in PE and PP with stiffness enhanced by the incorporation of coconut shell powder into engineering plastics.

Perhaps, the most interesting result of our experimental work has been to demonstrate the effectiveness of utilizing coconut shell powder as functional filler in PE and PP to

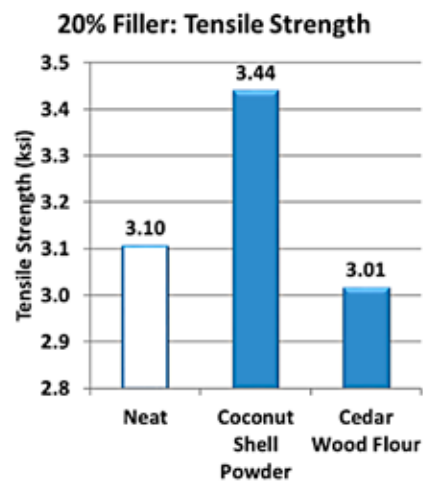
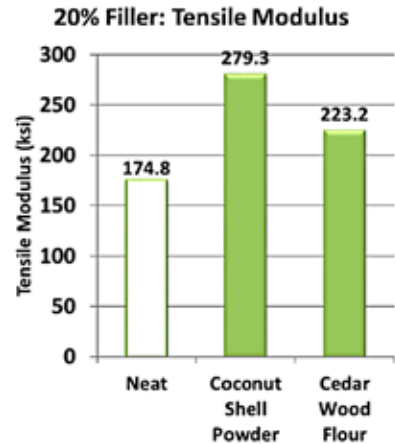


Figure 14. The addition of 20wt% coconut shell powder gives 56% increase in modulus and 10% increase in tensile strength of polyethylene and, much better than cedar wood flour.

increase the tensile strength and the tensile modulus. Our experiments compared (1) neat high density polyethylene to (2) high density polyethylene with 20 wt% cedar "wood flour" to (3) high density polypropylene with 20 wt% coconut shell powder added. The results are presented in Figure 14. The coconut shell powder increased the tensile modulus by 56% while the cedar "wood flour" increased the modulus by 32%. It is possible to add up to 40 wt% coconut shell powder to nearly double the tensile modulus in high density polyethylene. The tensile strength is also enhanced by the addition of 20 wt% coconut shell powder to high density polyethylene, but the increase in tensile strength is only 10% and the incorporation of cedar "wood flour" actually reduced the tensile strength by 3%.

### Summary

Coirform non-woven fabric composites made with coir

fiber and a polymeric binder fiber (i.e., PE or PP) offer: (1) substantial improvements in stiffness; (2) lower costs; and (3) reduced environmental impact. Coconut shell powder is functional filler that can: (1) significantly improve the modulus of elasticity of PE, PP and other engineering plastics; (2) reduce the cost of using pure PE or PP; and (3) reduce environmental impact.

**Acknowledgement**

The following students each made significant contributions to the results presented in this paper: Elisa Teipel, Stanton Greer, Sean Conroy, Matt Kirby, David Fait, Ben Peterson, David Hagen, and Ryan Vano. |

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# Life Cycle Assessment of Bio-Based Epoxies

By **Adhimoolam Bakthavachalam Kousaalya and Rakesh Krishnamoorthy Iyer, Department of Automotive Engineering and Clemson Composites Center, Clemson University, Greenville, SC; Srikanth Pilla, Department of Automotive Engineering, Clemson Composites Center, and Department of Materials Science and Engineering, Clemson University, Clemson, SC**

Editor's note: This paper was presented at ANTEC 2019.

## Abstract

The global demand for epoxy is increasing at a fast pace, with projections of the industry having a worth of \$11.5 billion by the year 2022. However, amidst growing concerns about eco-sustainability, the use of toxic and environmentally hazardous chemicals in conventional epoxies has triggered efforts among researchers on developing epoxies from various bio-sources. Yet, such efforts have not been accompanied by a thorough analysis of the environmental performance of such bio-based epoxies vis-à-vis their conventionally derived counterparts. This work aims at understanding the environmental performance of two different bio-based epoxies and compare with petroleum derived epoxy. It also highlights the impact of petroleum-based epoxies on human health and human carcinogen toxic categories. Lignin based epoxy performed poor on all the impact categories mainly due to use of excessive amount of chemicals during molecular breakdown of lignin to Vanillin.

## Introduction

Epoxies, a high-performance thermosetting resin containing the oxirane functional group, constitute the dominant share of global thermoset market (~ 70 %), primarily due to their versatility, high strength, good adhesion strength and excellent heat and electrical resistance. In the automotive sector, epoxies are mainly used as adhesives, under-the-hood electronics, and fiber-reinforced composites for body-in-white (BiW) applications [1]. Conventional petroleum-based epoxy resins (such as Diglycidyl Ether of Bisphenol-A or DGEBA) are synthesized through reaction between Bisphenol-A (BPA) and Epichlorohydrin (ECH) [2, 3]. However, both

chemicals (BPA and ECH) are highly toxic and are categorized as carcinogenic, mutagenic, and reprotoxic (CMR, a carcinogen of 1B category) [4]. This has driven efforts towards synthesis of epoxy from various biosourced precursors [5, 6] such as vegetable oil, liquefied wood/biomass, lignin, bark extractives, polyphenol, cardanol, tannin and rosin.

Among the above-mentioned bio-based alternatives, lignin and bark extractive-based epoxies exhibit properties similar to conventional DGEBA resins and possess the potential to be a green alternative to petroleum-based resins. However, to select the most environmentally benign epoxy for any application, understanding the environmental impacts is vital. Hence, this study aims to assess the environmental impacts of two bio-epoxies (Table 1) starting from raw material extraction till the manufacturing stage (cradle-to-factory gate). The advantages and disadvantages of these bio-epoxies are also provided in Table 1. Environmental impacts of both these bio-epoxies on 17 impact categories were compared with those of conventional DGEBA.

Table 1: Different types of epoxy based on their source.

| Epoxy nomenclature/ Source  | Advantage   | Disadvantage   | Ref |
|-----------------------------|---|--|-----|
| E-Epoxy/<br>Bark extractive | Easy synthesis and manufacturing  | Larger petroleum-based content process                                 | [7] |
| L-Epoxy/<br>Lignin          | Lignin a by-product of paper industry ensuring enhanced sustainability due to use of waste resource | Very high viscosity and molecular weight makes the process challenging | [8] |
| P-Epoxy/<br>Petroleum       | Conventionally used   | Presence of petroleum derived and carcinogenic chemicals               | [9] |

## Materials and Methods

### Goal Scope and Functional Unit

The goal of this life cycle assessment (LCA) study is to evaluate the ecological performance of bio-epoxies sourced from different biological sources and compare

their impacts with those of conventional petroleum-based epoxy. Hence, two bio-epoxies, namely, bark-based epoxy and vanillin-based epoxy (derived using lignin) have been considered. These have been compared with conventional petroleum-based epoxy. We have carried out a comparative cradle-to-factory gate LCA viz. sourcing of raw material and its subsequent extraction, processing and purification of epoxy, and the final manufacture of epoxy panel along with all chemicals/materials used, as per the methodology defined in the ISO 14040 and ISO 14044 standards [10, 11]. Both the use and end-of-life stages of the panel have been excluded from the scope of this LCA study (Figure 1). Use phase of the panel has not been considered in this study as it is assumed that there is no difference in the emission during the use phase.

Since any LCA should involve a like-for-like comparison, the functional unit of this study is defined as a rectangular panel that has length and width of 1 m each, but has variable thickness depending on the specific epoxy used to manufacture the panel. The reason for this variable thickness is because the panel is designed to withstand a uniform load of 250 N while undergoing a maximum deflection of only 1.5 mm. Hence, depending on the mechanical properties of the selected epoxy, its thickness

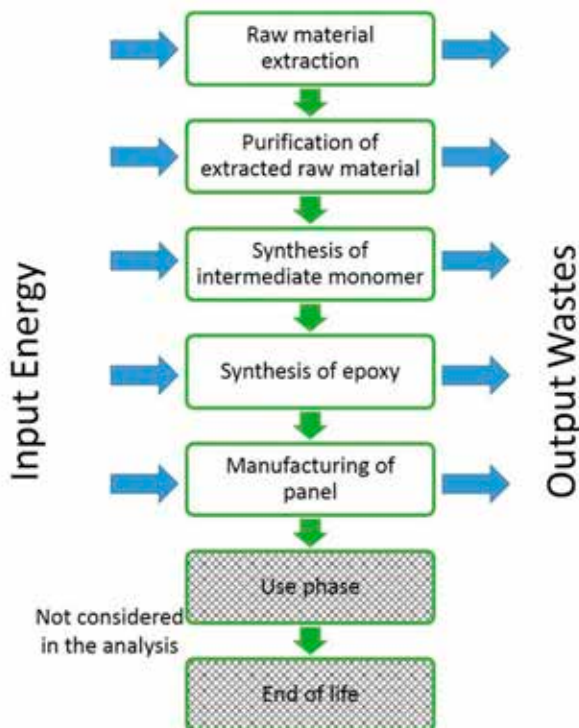


Figure 1: System boundary considered in this LCA study.

will be variable. Therefore, to design the panel and estimate the desired thickness values for each epoxy panel, its corresponding modulus values – as reported in existing literature – were considered and have been shown in Table 2. Here, each epoxy has been re-named as follows: bark-based epoxy is E-epoxy; vanillin-based epoxy is actually derived from lignin or it is renamed as L-epoxy; and conventional petroleum-based epoxy is P-epoxy.

Based on the mechanical properties (mentioned in Table), variable thickness of each epoxy panel can be calculated using Equation 1 and are provided in Table 2.

$$d^3 = \frac{15\omega l^4}{96Eb\Delta} \quad (1)$$

Where:

- $\omega$  - Load applied (N/m)
- $E$  - Young's Modulus (MPa)
- $l$  - Length of panel (m)
- $b$  - Width of panel (m)
- $d$  - Thickness of the panel (mm)
- $\Delta$  - Deflection (mm), 1.5 mm

Densities of each of the chosen epoxy systems was obtained from existing literature, based on which the weight of the epoxy panel was calculated and has been reported in Table 2. Finally, we assumed about 10 % of material wastage during processing, and thus calculated the input amount of epoxy required.

Table 2: Thickness of the panel for different epoxies and their mechanical properties.

| Epoxies Type | Thickness (mm) / Panel Weight (kg) | EEW (g/eq) | Tensile Strength / Modulus (MPa) | Ref |
|--------------|------------------------------------|------------|----------------------------------|-----|
| E            | 27 / 27.89                         | 250        | 63 / 1200                        | [7] |
| L            | 30 / 30.05                         | 250        | 45 / 959                         | [8] |
| P            | 24 / 23.87                         | 165        | 60 / 1913                        | [9] |

#### LCA Inventory and Impact Assessment

With regard to bark-based epoxy, the synthesis procedure followed by Kuo et. al [7] was considered. Initially bark chips were obtained after cutting of softwood, following which they were combined with aqueous sodium hydroxide solution and mixed thoroughly. The solution

was subsequently filtered, and later spray dried to enable the removal of water and sodium hydroxide as well as any impurities in bark chips. Following this, bark extractives were obtained through a two-step process, with the first step involving reaction with epichlorohydrin (added in excess) in the presence of aqueous sodium hydroxide, 1,4-dioxane and catalyst amidst stirring at higher temperature, and the second step of filtering and washing the bark-based solution to remove the aforementioned chemicals as well as any salt formed in the process. Finally, rotary evaporation was undertaken to remove any chemical present in bark epoxy, and that was subsequently mixed with petroleum-based epoxy and cured to obtain the final epoxy panel.

To produce panels from vanillin-based epoxy, lignin produced along with softwood pulp was considered [12], after which vanillin was derived from lignin [13]. This vanillin was later subjected to multiple processes, including treatment with chemicals (such as tetrahydrofuran, hydrochloric acid, hydrogen peroxide, sodium chloride, and sodium hydroxide) in order to first produce methoxyhydroquinone and subsequently, its diglycidyl ether. These two chemicals were later combined in a chemical reaction in the presence of a catalyst to produce an oligomer that was finally mixed with an amine hardener to obtain the desired vanillin-based epoxy panel. For the conventional epoxy, the appropriate resin in the Simapro software (Ecoinvent 3.4 database) was used.

Based on the inventory data, we quantified the environmental impacts of both bio-epoxy panels along with that of the conventional epoxy panel by applying the hierarchist perspective of ReCiPe midpoint method [14] on data provided using the Ecoinvent 3.4 database. Environmental impacts were investigated on all 17 midpoint impact categories, namely: GW (global warming); stratospheric ozone depletion (SOD); ionizing radiation (IR); ozone formation – human health (OHH); ozone formation – terrestrial ecosystems (OTS); fine particulate matter formation (FPM); terrestrial acidification (TA); freshwater eutrophication (FWE); terrestrial ecotoxicity (TE); freshwater ecotoxicity (FE); marine ecotoxicity (ME); human carcinogenic toxicity (HCT); human non-carcinogenic toxicity (HNT); land use (LU); mineral resource scarcity (MRS); fossil resource scarcity (FRS); and water consumption (WC). For each midpoint impact category, the

prominent contributing reasons were identified and have been briefly described. A termination criterion of 1 % was used in this work to determine the significant contributing reasons for each epoxy.

## Results and Discussion

Figure 2 shows a comparative performance of all three epoxy panels on all 17 midpoint impact categories. As can be seen, for all environmental impacts, vanillin-based epoxy panel shows the highest environmental impact – much higher than petroleum-based epoxy (conventional epoxy) and bark extractive-based epoxies on all impact categories. Table 3 shows the magnitude of the 17 midpoint environmental impacts caused by the vanillin panel. However, bark extractive-based epoxies exhibited comparable environmental performance in 7 impact categories and performed poorer than the petroleum based in 10 categories. This is a highly interesting finding, given that vanillin and Bark extractives are derived from biological resources, unlike conventional epoxy resins that are derived completely from petroleum resources. However, this is explained by a combination of three reasons. The first is that conventional epoxy resins inventory was selected from the Ecoinvent database that considers the production of epoxy on an industrial scale in large amounts from a completely optimized process conditions. While for vanillin and bark extractive-based epoxies, the inventory was developed based on laboratory-based experiments that were not highly optimized for an industrial process. Despite this difference, on impact categories such as ozone formation human health and human carcinogen toxicity, the toxic nature of conventional epoxies has been highlighted. And the second reason is that these epoxies are not 100 % bio-derived and only partially replaced with bio-based content resulting in less than 50 % bio-based content in the final epoxy molecule. The third reason is that the multi-step and energy-intensive nature of lignin extraction, purification and molecular breakdown along with the use of excessive amount of chemicals at each stage. While lignin was derived from bio-source, use of several other chemicals in the process path were petroleum derived. An additional factor behind this performance, especially on toxicity-related impacts, is the use of epichlorohydrin and triethylbenzylammonium chloride – both of which are well known for being toxic – and also cause ozone depletion.

While care was taken at our end with regard to energy savings and recycling of resources and materials used in the various steps for obtaining vanillin-based epoxy, there still remains the challenge of taking laboratory-scale inventory to commercial scale. Hence it is important to refine the synthesis process and optimize the production to engage in minimal consumption of resources and precursor materials while using environmentally benign chemicals at all stages. It is also important to synthesis epoxies that are free from toxic chemicals like epichlorohydrin.

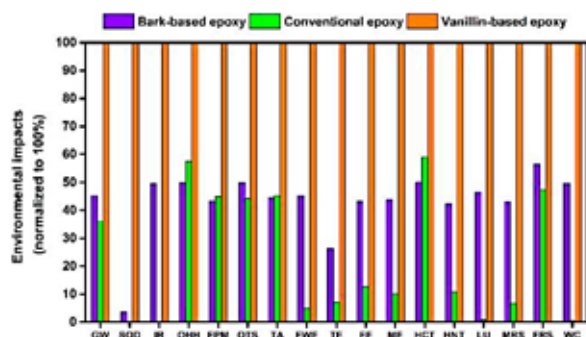


Figure 2: Environmental impacts of various epoxies on 17 impact categories

Table 3: Environmental impacts of vanillin-based epoxy

| Impact category                         | Unit                    | Value     |
|---|-------------------------|-----------|
| Global warming                          | kg CO <sub>2</sub> eq   | 515.2485  |
| Stratospheric ozone depletion           | kg CFC11 eq             | 0.0016    |
| Ionizing radiation                      | kBq Co-60 eq            | 110.0522  |
| Ozone formation, Human health           | kg NO <sub>x</sub> eq   | 1.6555    |
| Fine particulate matter formation       | kg PM <sub>2.5</sub> eq | 0.5474    |
| Ozone formation, Terrestrial ecosystems | kg NO <sub>x</sub> eq   | 2.1872    |
| Terrestrial acidification               | kg SO <sub>2</sub> eq   | 1.8514    |
| Freshwater eutrophication               | kg P eq                 | 0.1286    |
| Terrestrial ecotoxicity                 | kg 1,4-DCB eq           | 0.2037    |
| Freshwater ecotoxicity                  | kg 1,4-DCB eq           | 5.8822    |
| Marine ecotoxicity                      | kg 1,4-DBC eq           | 10.2666   |
| Human carcinogenic toxicity             | kg 1,4-DBC eq           | 11.6690   |
| Human non-carcinogenic toxicity         | kg 1,4-DBC eq           | 6183.2246 |

|                           |                          |          |
|---------------------------|--------------------------|----------|
| Land use                  | m <sup>2</sup> a crop eq | 20.2343  |
| Mineral resource scarcity | kg Cu eq                 | 0.5507   |
| Fossil resource scarcity  | kg oil eq                | 146.8513 |
| Water consumption         | m <sup>3</sup>           | 944.2213 |

## Conclusions

In sum, the aforementioned comparison sheds two insights with regard to making bio-based epoxies a more environment-friendly alternative to petroleum-based epoxies. First, it establishes the need for collaborations between researchers and industries or commercial organizations that can produce such epoxies, so as to identify possibilities for optimized, highly-efficient production means and methods to enhance the recycling of chemicals used in producing epoxies, as well as reducing the energy-intensiveness of the overall procedure. Second, it also shows that more efforts need to be undertaken towards developing novel epoxies that can be produced while completely avoiding the need for use of toxic chemicals (such as epichlorohydrin) to produce these systems. Vegetable oils-based epoxies could be one such alternative, and therefore, could be one potential area of exploring for the future.

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