Utilization of Natural Fibers in Plastic Composites: Problems and Opportunities

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Abstract

Results suggest that agro-based fibers are a viable alternative to inorganic/material based reinforcing fibers in commodity fiber-thermoplastic composite materials as long as the right processing conditions are used and for applications where higher water absorption may be so critical. These renewable fibers hav low densities and high specific properties and their non-abrasive nature permits a high volume of filling in the composite. Kenaf fivers, for example, have excellent specific properties and have potential to be outstanding reinforcing fillers in plastics. In our experiments, several types of natural fibers were blended with polyprolylene(PP) and then injection molded, with the fiber weight fractions varying to 60%. A compatibilizer or a coupling agent was used to improve the interaction and adhesion between the non-polar matrix and the polar lignocellulosic fibers. The specific tensile and flexural moduli of a 50% by weight (39% by volume) of kenaf-PP composites compares favorably with 40% by weight of glass fiber (19% by volume)-PP injection molded composites. Furthermore, prelimimary results sugget that natural fiber-PP composites can be regrounded and recycled.

Introduction

There is a greater awareness of the need for materials in an expanding world population and increasing affluence. It took all of recorded history for the world population to reach 1 billion by the year 1830. In 1930, it had doubled to 2 billion. While it took one hundred years to increase the population by 1 billion people, at the present rate of population adds 1 billion people every eleven years.

It is estimated that by the year 2000, twenty-five percent of the population of China will become "middle class". This represents more people than the entire population of the United States. If the desire for Materials in this growing segment of China equals the middle class of others countries, there will be a great need for new materials.

China is only one example of ;hte large new markets that will open up for new materials. Asia, Mexico, South America and Eastern Europe are also "emerging" as industrial consumers that will seek new materials.

It is possible to make completely new types of composite materials by combining different resources. It is possible to combine, blend, or alloy lignocellulosic or agro-based fiber with materials such as glass, metals, plastics and synthetics to produce new classes of composite materials. The objective will to be combine two or more resources in such a way that a synergism between the components results in a new material that is much better than the individual components.

One of the biggest new areas of research in this field is in combining natural fibers with thermoplastics [Sanadi et al. 1994 a,b,c]. Since prices for plastics have risen sharply over the past few years, adding a natural powder or fiber to plastics provides a cost reduction to the plastic industry (and in some cases increases performance as well) but to the agro-based industry, this represents an increased value for the agro-based component. Most of the research has concentrated on using a compatibililzer to make the hydrophobe (plastic) mix better with the hydrophil (lignocellulosic). The two components remain as separate phases, but if delimitation and/or void formation can be avoided, properties can be improved over those of either separate phase. These types of materials are usually referred to as natural fiber/thermoplastic blends.

Recent interest in reducing the environmental impact of materials is leading to the development of newer materials or composites that can reduce the stress to the development. In light of petroleum shortages and pressure for decreasing the dependence on petroleum products, there is an increasing interest in maximizing the use of renewable materials. The use of agricultural materials as source of raw materials to the industry not only provides a renewable source but could also generate a non-food source of economic deveopment for farming and rural areas.

Several billion pounds of fillers and reinforcements are used annually in the plastic industry. The use of additives in plastics is likely to grow with the introduction of improved compounding technology and new coupling agents that permit the use of high filler/reinforcement content [Katz and Milewski, 1987]. As suggest by Katz and Milewski, fillings up to 75 pph could be common in the future: this could have a tremendous impact in lowering the usage of petroleum based plastics. It would be particularly beneficial, both in terms of the environment and also in socio-economic terms, if a significant amount of the fillers were obtained from a renewable agricultural source. Ideally, of course, an agro-/bio-based renewable polymer reinforced with agro-based fibers would make the most environmental sense.

Advantages of Using Agro-Fibers in Plastics

The primary advantages of using annual growth lignocellulosic fibers as fillers/reinforcements in plastics are low densities, non abrasive, high filling levels possible resulting in high stiffness properties, high specific properties, easily recyclable, unlike britte fibers, the fibers will not fractured when proessing over sharp curvatures, biodegradable, wide variety of fibers available thoughout the world, would generate rural jobs increases non-food agricultural/farm based economy, low energy consumption and low cost.

Material cost savings due the incorporation of the relatively how cost agro-fibers and ghigher filling levels possible, coupled with the advantage of being non-abrasive to the mixing and the molding equipment are benifits that are not likely to be ignored by the plastics industry for use in the automotive, building, appliance and other applications.

Prior work on lignocellulosic fibers in thermoplastics has concentrated on wood based flour or fibers and significant advance have been made by a number of researches [Woodhams c at 1984, Klason and Kubat 1986a,b, Myers *et al.* 1993, Kokta *et al.* 1989, Yam *et al.* 1990, Bataille *et al.* 1989 and Sanadi *et al.* 1994d]. A recent study on the use of annual growth lignocellulosic fibers indicate that these fibers have the potential of being used as reinforcing fillers in thermoplastics [Sanadi *et al.* 1994b]. The use of annual growth agricultural crop fibers Such as kenaf has resulted in significant property advantages as compared to typical wood based fillers/fibers such as wood flour, wood fibers and recycled newspaper. Properties of compatibilized PP and kenaf has mechanical properties comparable with those of commerical PP composites [Sanadi *et al.* 1994b]

Limitations

The primary drawback of the use of agro-fibers is the lower processing temperature permissible due to the possibility of lignocellulosic degradation and/or the possibility of volatile emissions that could effect composite properties. The processing temperatures are thus limited to about 200°C although it is possible to use higher temperatures for short periods. This limits the type of thermoplastics that can be used with agro-fibers to commodity thermoplastics such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC) and polystyrene (PS). However, it is important to note that these lower priced plastics constitute about 70% of the total thermoplastics consumed by the plastics industry and subsequently, the use of fillers/reinforcement presently used in these plastics far outweigh the use in other higher cost plastics.

These second drawback is the high moisture absorption of the natural fibers. Moisture absorption can result in swelling of the fibers and concerns on the dimension stability of the agro-fiber composites cannot be ignored. The absorption of moisture by the fibers are minimized in the composite due to encapsulation by the polymer. It is difficult to entirely eliminate the absorption of moisture without using expensive surface barriers on the composite surface. If necessary, the moisture absorption of the fibers can be dramatically reduced through chemical modification of some of the hydroxyl groups present [Rowell *et al.* 1986] in the fiber but with some increase in the cost of the fiber. Good fiber-matrix bonding can also decrease the rate and amount of water absorbed by the composite.

It is important to keep these limitations in perspective when developing end use applicatioons. We believe that by understanding the limitation and benefits of these composites, these renewable fibers are not likely to be ignored by the plastics/composites industry for use in the automotive, building, appliance and other applications.

Fiber Supply

In any commercial development, there must be a long term guaranteed supply of resources. In order to insure a continous fiber supply, management of the agricultural producing land shoud be under a proactive system of land management whose goal is both sustainable agriculture and the promotion of healthy ecosystems. Ecosystem management is not an euphemism for preservation, which might imply benign neglect. Sustainable agriculture denotes a balance between conservation and utilization of agricultural lands to serve both social and economic needs, from local, national and global vantage points. Sustainable agriculture does not represent exploitation but rather is aimed toward meeting all the needs of the present generation without compromising the ability of future generations to meet their needs. It encompasses, in the present case, a continous production of fiber, considerations of multi-land use and conservation of the total ecosystem.

There is a wide variety of agro-based fibers to consider for utilization. All of them should be considered for composites to take advantage of unique fiber properties each plant type has to offer not just because we have a desire to promote one fiber over another. Unless one particular fiber has some advantage in the market, it will be replaced with whatever resource has the market advantage. That market advantage can be based on many elements such as availability, price, or performance. Desire does not drive markets! Producers and manufacturerrs of agro-fiber must explore common interests and, where possible, prepare an enterprise-driven long range strategic plan for development and promotion of an agro-fiber industry [Rowell 1994].

Table 1 shows the inventory of some of the larger sources of agricultural crop fiber that could be utilized for natural fiber/thermoplastic composites. The data for this table was extracted from several sources using estimates [Atchison 1991] and extrapolations for some of the numbers. For this reason, the data in Table 1 should only be considered to be a rough relative estimate of world fiber resources.

Table 1. Inventory of major potential world fiber sources.

Fiber Source	World (dry metric tons)
Wood	1,750,000,000
Straw (wheat, rice, oat, barley, rye, flax, grass)	1,145,000,000
Stalks (corn, sorghum, cotton)	970,000,000
Sugar cane bagasse	75,000,000
Reeds	30,000,000
Bamboo	30,000,000
Cotton staple	15,000,000
Core (jute, kenaf, hemp)	8,000,000
Papyrus	5,000,000
Bast (jute, kenaf, hemp)	2,900,000
Cotton linters	1,000,000
Esparto grass	500,000
Leaf (sisal, abaca, henequen)	480,000
Sabai grass	200,000
TOTAL	4,033,080,000

The traditional source of agro-based fiber has been wood and for many countries, this will continue to be the major source. Wood has a higher density than annual plants so there will be more bulk when using agricultural crop fiber. There are also concerns about the seasonality of annual crops which requires considerations of harvesting, separating, drying, storing, cleaning, handling and shipping. In the present system of using wood, storage costs can be reduced by letting the tree stand alive until needed. With any annual crop, harvesting must be done at a certain time and storage/drying/cleaning/separating will be required. This will almost certainly increase costs of using agro-based resources over wood depending on land and labor costs, however, in those countries where there is little or no wood resource left or where restrictions are in place to restrict the use of wood, alternate sources of fiber are needed if there is to be a natural fiber industry in those countries.

Other large sources of fiber can come from recycling agro-fiber based products such as paper and paper, waste wood and point source agricultural residues. Recycling paper products back into paper requires wet processing and removal of inks, inorganic and adhesives. Recycling these same products into composites can be done using dry processing (thus eliminates a waste water stream) and all co-existing resources can be incorporated into the composite. Point source fiber sources represent resources such as rice hulls from a rice processing plant., sun flower seed hulls from an oil processing unit and bagasse from a sugar mill.

Table 2 shows the chemical composition of many different types of agro-fibers, Table 3 shows fiber dimensions and Table 4 shows tensile strength [Atchinson 1983]. This type of data is critical in order to select a certain fiber for a specific use. While this type of data exists in the literature for some types of agro-fibers, the data is incomplete. There Needs to be a concerted effort to expand the data base to include all potential fiber sources.

Processing Considerations and Techniques

Separation of the fibers from the original plant source is an important step to ensure the high quality of fibers. The limiting processing temperatures when using lignocellulosic materials with thermoplastics is important in determining processing techniques. High processing temperatures (200°C) that reduces melt viscosity and facilitates good mixing cannot be used (except for short periods) and other routes are needed to facilitate mixing of the fibers and matrix in agro-fiber thermoplastics.

An excellent review by Milewski [1992] on short fiber composite technology covers a variety of reasons associated with composite properties falling short of their true reinforcing potential. The major factors that govern the properties of short fiber composites are fiber dispersion, fiber length distribution, fiber orientation and fiber-matrix adhesion. Mixing the polar and hydrophilic fibers with non-polar and hydrophobic matrix can result in difficulties in dispersing the fibers in the matrix. Clumping and agglomeration must be avoided to produce efficient compsites. The efficiency of the composite also depends on the amount of stress transferred from the matrix to the fibers. This can be maximized by improving the interaction and adhesion between the two phases and also by maximizing the length of the fibers retained in the final composite [Bigg *et al.* 1988]. Using long filaments during the compounding stage can result in higher distribution. However, long fibers sometimes increase the amount of clumping resulting in areas concentrated with fibers and areas with excessive matrix; this ultimate reduces the com-

Table 2. Chemical Composition of Some Common Fibers.

Type of Fiber	cellulose	Lignin Pentosan	Ash	Silica
Stalk fiber				
Straw				
Rice	28-36	12-16	23-28	15-20
Wheat	29-35	16-21	26-32	4.5-9
Barley	31-34	14-15	24-29	5-7
Oat	31-37	16-19	27-38	6-8
Rye	33-35	16-19	27-30	2-5
Cane fiber				
Sugar	32-44	19-24	27-32	1.5-5
Bamboo	26-43	21-31	15-26	1.7-5
Glass fiber				
Esparto	33-38	17-19	27-32	6-8
Sabai	22.0	23.9	6.0	—
Reed fiber				
Phragmites commnis	44.75	22.8	20.0	2.9
Bast fiber				
Seek flax	43-47	21-23	24-26	5
Kenaf	31-39	15-19	22-23	2-5
Jute	45-53	21-26	18-21	0.5-2
Core fiber				
Kenaf	31-44	15-21	—	2.2
Jute	41	24	22	0.8
Leaf fiber				
Abaca (Manila)	60.8	8.8	17.3	1.1
Sisal (agave)	43-56	7-9	21-24	0.5
Seed Hull fiber				
Cotton linter	80-85	—	0.8-2	—
Wood fiber				
Coniferous	40-45	26-34	7-14	—
Deciduous	38-49	23-30	19-26	—

Table 3. Dimensions of Some Common Lignocellulosic Fibers.

	Fiber Dimension (mm)		
Type of Fiber	Average Length	Length	Width
cotton	10-60	18	0,02
Flax	5-60	25-30	0.012-0.027
Hemp	5-55	20	0.025-0.050
Manila hemp	2.5-12	6	0.025-0.040
Bamboo	1.5-4	2.5	0.025-0.040
Esparto	0.5-2	1.5	0.013
Cereal straw	1-3.4	1.5	0.023
Jute	1.5-5	2	0.02
Deciduous wood	1-1.8	1.2	0.03
Coniferous wood	3.5-5	4.1	0.025

Table 4. Tensile strength of some agro-based fibers.

Fiber	Tensile Strength + (Gpa)
Kenaf	11.91
Hemp	8.95
Wood	7.48
Sisal	6.14
Cotton	3.54

+ all single fiber strength except sisal which is for fiber bundles.

posite efficiency. Uniform fiber dispersion cannot be compromised and a careful selection of processing techniques, initial fiber lengths, process conditions and processing aids are needed to obtain efficient composites. Several types of compounding equipment, both batch and continuous equipment, have been used for blending lignocellulosic fibers and plastics.

The ultimate fiber lengths present in the composite depends on the type of compounding and molding equipment used. Several factors contribute to the fiber attrition such as the shearing forces generated in the compounding equipment, residence time, temperature and viscosity of blends. An excellent study on the effect of processing and mastication of several types of short fibers in thermoplastics was conducted by Czarnecki and White [1980]. They concluded that the extend of break-age was most severe and rapid for glass fibers, less extensive for kevlar (aramid) fibers and the least for cellulose fibers. The level of fiber attrition depends on the type of compounding and molding equipment used, level of loading, temperature and viscosity of the blend [Czarnecki and White 1980].

The properties of the agro-based thermoplastic composites are very process dependent. Yam *et al.* [1908] at Michigan State University, studied the effect of twin screw blending of wood fibers and HDPE and concluded that the level of fiber attrition depended on the screw on the configuration and the processing temperature. Average fiber lengths decreased from about 1.26 mm prior to compounding to about 0.49 mm after extrusion. Modification of the screw configuration reduced fiber attrition to an average length of about 0.78 mm. Fiber weight percent up to 60% were reported to have been mixed. The tensile strength of the pure HDPE was higher than that of the wood fiber-HDPE, irrespective of the level of fiber filling. This was explained to be because of a lack of dispersion with fibers clumping in bundles and poor fiber-matrix bonding. Use of stearic acid in HDPE/wood fibers improved fiber dispersion and improved wetting between the fiber and matrix [Woodhams 1984] and resulted in significant improvement in mechanical properties. Work by Raj and Kokta [1989] indicate the importance of using surface modifiers to improve fiber dispersion in cellulose fibers/PP composites. Use of a small amount of stearic acid during the blending of cellulose fibers in polypropylene decreased both the size and number of fiber aggregates formed during blending in an internal mixer (Brabender roll mill).

Another technique that is gaining acceptance is the high intensity compounding using a turbine mixer (thermokinetic mixer). Woodhams *et al.* [1990] and Myers *et al.* [1992] found the technique effective in dispersing lignocellulosic fibers in thermoplastics. Addition of dispersion aids/coupling agents further improved the efficiency of mixing. The high shearing action development in the mixer decreased the lengths of fibers in the final composite. However the improved fiber dispersion resulted in improved composite properties. Recent work using a thermokinetic mixer to blend kenaf in PP [Sanadi *et al.* 1994b] has confirmed the usefulness of the compounding technique in effectively dispersing natural fibers in the thermoplastic matrix. An added advantage is that no pro-drying of the fibers is needed prioor to the blending stage in the mixer.

Properties of Natural Fiber-Thermoplastic Composites

Cellulosic fillers/fibers have been incorporated in a wide variety of thermoplastics such as polypropylene, polyethylene, polystyrene, polyvinyl chlordide, polyamides [Klason and Kubat 1986a,b]. In general, dispersing agents and/or coupling agents, are necessary for property enhancement when fibers are incorporated in thermoplastics. Grafting chemical species on to the fiber surface has also been reported to improve the interaction between the fibers and matrix. Although grafting can improve the properties of the composite to a significant extent, this process increases the material cost of system. The use of dispersing agents and/or coupling agents is a cheaper route to impmrove properties and makes more practical sense for high volume, low cost composite systems.

In general, cellulosic fillers or fibers have a higher Young's modulus as compared to commodity thermoplastics thereby contributing to the higher stiffness of the composites. The increase in the Young's modulus with the addition of cellulosics depends on many factors such as the amount of fibers used, the orientation of the fibers, the interaction and adhesion between the matrix, the ration of the fiber to matrix Young's modulus, etc. the Young's modulus of the composite can be crudely estimated through the simple rule of mixtures and other simple models if the Young's modulus of the filler/fiber is known [Hull 1981]. The use of dispersing or coupling agents can change the molecular morphology of the polymer chains both at the fiber-polymer interphase and also in the bulk matrix phase. Crystallites have much higher moduli as compared to the amorphous reghions and can increase the modulus contribution of the polymer matrix to the composite modulus. A good understanding of the effect of dispersing agents and coupling on transcrystallinity at the fiber-matrix interphase and the corresponding effect on the composite Young's modulus is nonexistent. Therefore the influence and contribution of the molecular morphology on estimating the composite modulus through simple models is lacking.

In order to use models to estimate composites properties, it is necessary to know the property of the fibers. In general, natural fibers such as kenaf and jute are in the form of filaments that consist of discrete individual fibers, generally 2 mm to 6 mm long, which are themselves composites of predominantly cellulose, lignin and hemicelluloses. Filament and individual fiber properties can vary widely depending on the source, age, separating techniques, moisture content, speed of testing history on the fiber, etc. The properties of the individual fibers are very difficult to measure. Earlier work on a natural bast filament, sun hemp *(Crotalaria Juncea)* suggested that the filament properties ranged widely. The tensile strengths of the filaments of sun hemp varied from about 325 MPa to MPa, while tensile modulus ranged from 27 MPa to 28 MPa [Sanadi *et al.* 1985]. In a natural fiber-thermoplastic composite the lignocellulosic phase is present in a wide range of diameters and lengths, some in the form of short filaments and others in forms that seem closer to the individual fiber. The high shearing energy of blending the filaments and the polymer in a mixer results in fiber attrition but can also axially separate the filaments into discrete individual fibers.

Cellulosic fillers/fibers can be classified under three categories depending on their performance when incorporated in a plastic matrix. Wood flour and other low cost agricultural based flour can be considered as particulate fillers that enhance the tensile and flexural moduli of the composite with little effect on the composite strength. Wood fibers and recycled newspaper fibers have higher aspect ratios and contribute to an increase in the moduli of composite and can also improve the strength of the composite when suitable additives are used to improve stress transfer between the matrix and the fibers. The improvement in modulus is not significantly different than the cellulosic particulate fillers. The most efficient cellulosic additives are some natural fibers such as kenaf, jute, flax, etc. The specific Young's modulus and specific flexural modulus, the ratio of the composite modulus to the composite specific gravity of composites with natural fibers such as kenaf are significantly higher than those possible with wood fibers. Thje sprcific moduli (the ratio of the compostie modulus to the composite specific gravity) of high fiber volume fraciton bast fibers-PP composites are high and in the range as glass fibers-PP composites. The most efficient natural fibers are those that have a high cellulose content coupled with a low micro fibril angle resulting in high filament mechanical properties.

Although several plastics have been used with cellulosic fibers, the major part of the work at the University of Wisconsin-Madison and the Forest Producls Laboratory has been on polypropylene. The work reported here will concentrate on this versatile plastic in combination wilth several tyypes of agro-resources [Jacobson *et al.* 1995].

Experimental

Materials

The base resin was a polypropylene homopolymer, Fortilene 1602 (generously donated by Solvay Polymer, Houston, TX) with a melt flow index of 12 gr/10 min at 230°C (ASTM - D238). A maleic anhydride grafted polypropylene (MAPP) modifier, Epole G3002 (donated by Eastman Chemical, Kingston, TN) was used to enhance the surface adhesion between the agro-wastes and the PP matrix. The agro-resources used in this study were kenaf bast and core form AG-Fibers lnc, corncob, hard from Composition Materials Inc., corn fiber from Cargill Inc., oat hulls from Quaker Oats Co., rice hulls from Busch Agricultural Resources Center, peanut hulls from Seminole Peanut Co. and soybean hull residue from WI Soybean Assoc., oat/wheat straw, wood flour and jojoba seed from other sources.

Methods

Thc agro-resources were run through a Wiley mill a 30 mesh screen. The agro-resources, PP and MAPP, were compounded in a 1 liter-high intensity shear-thermokinetic mixer (Synergistics Industries Ltd., Canada). No external heat sources are required due to the high shearing/smearing of the PP which produces friction and generates heat. The shearing action causes softening and flow of the composite system. A thermally controlled monitor regulated the dump temperature at 168°C to 199°C depending on the fiber type. The composites were compounded at 5000 rpm (tip speed = 32.9 m/s) and 150 gram batches were standard. Directly after reaching the dump temperature the material was pressed flat to enhance cooling and prevent fibers in the core of the composite from burning.

The resultant composite blends were then granulated and dried at 105°C for 4 hours to drive off residual fiber moisture in preparation for injection molding. A 33-ton Cincinnati Milacron injection molder was used to produce standard ASTM tensile, flexural and impact specimens.

Testing

Specimens were placed in a controlled himidity room for three days prior to testing to asssure complete thermal stability of the test samples. Test conditions were performed according to the following ASTM standards; tensile testing (ASTM D638), flexural testing (ASTM D790) impact testing (ASTM D256).

Results and Discussion

Physical Properties

Water absorption and specific gravity of lignocellulosic fiber composties are important characteristics that determine end use applications of these materials. Water absorption could lead to a decrease in some of the properties and needs to be considered when selecting applications. It is difficult to entirely eliminate the absorption of moisture in the composites without using expensive surface barriers on the composite surface. Water absorption in lignocellulosic based composites can lead to a build up of moisture in the fiber cell wall and also in the fiber-matrix interphase region. Moisture build up in the cell wall could result in fiber swelling and concerns on the dimension stability connot be ignored. If necessary, the moisture absorbed in the fiber cell wall can be reduced through the acetylation of some of the hydroxly groups present [Rowell *et al.* 1986] in the fiber, but with some increase in the cost. Good wetting of the fiber by the matrix and adequate fiber-matrix bonding can decrease the rate and amount of water absorbed in the interphasial region of the composite. A typical 50% by weight of kenaf-homopolymer PP blend absorbed about 1.05% by weight of water in a 24 hr water soak test. This is considerably higher than any mineral filled systems. It is therefore very important to select applications where this high water absorption is not a critical factor such as in electrical housing components.

The specific gravity of lignocellulosic based composites is much lower than the mineral filled thermoplastic systems. The apparent density of the lignocellulosic fibers in PP is about 1.4 g/cc as compared to mineral fillers/fibers (about 2.5 g/cc). The specific gravity of a 50% (by weight) kenaf-PP composite is about 1.07, while that of a 40% (by weight) glass-PP composite is 1.23. The specific mechanical properties of kenaf-PP composites compare favorably to other filled commodity plastics. Since materials are bought in terms of weight and pieces or articles are in general sold by the number, more pieces can be made with lignocellulosic fibers as compared to the same weight of mineral fibers. This could result in significant material cost savings in the high volume and low cost commodity plastic market.

Table 5 shows a comparison of properties of 50% kenaf and 40% recycled newspaper fiber with 40% talc, 40% calcium carbonate, 40% glass and 40% mica. Tensile and flexural moduli and moisture sorption

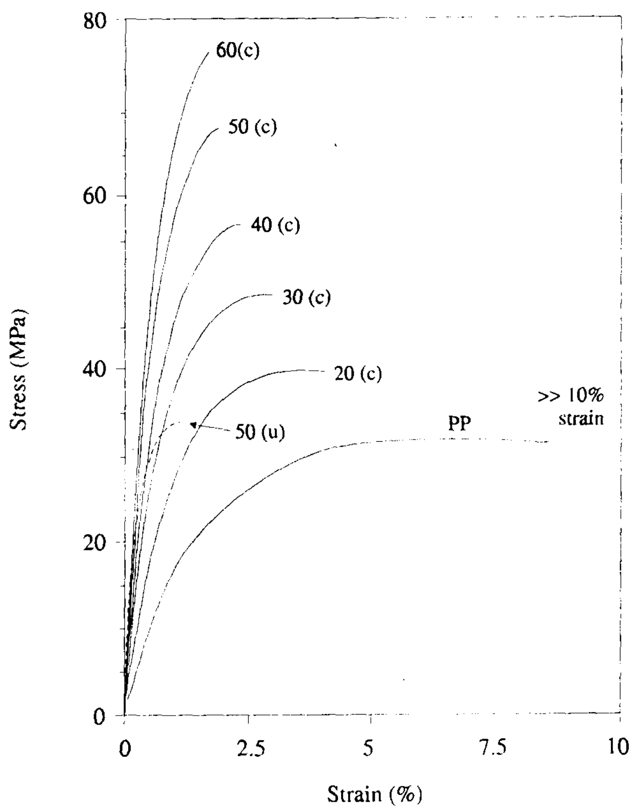

Figure 1. Tensile stress-strain curves of kenaf-PP. The numbers near the end of curves indicate kenaf weight % and (c) indicates couppled and (u) uncoupled composites. All coupled systems containcd 2% by weight of MAPP. PP failure strain was 10%.

Table 5.

Filler/Reinforcement in PP	ASTM standard	none	kenaf	recycled newspa per fiber	talc	$CaCO_3$	glass	mica
% filler by weigth		0	50	40	40	40	40	40
% filler by volume (estimated)		0	39	30	18	18	19	18
Tensile Modulus, Gpa	D638	1,7	8,3	4,4	4	3,5	9	7,6
Specific Tensile		1,9	7,8	4,5	3,1	2,8	7,3	6,0
Modulus, Gpa Tensile Strength, Mpa	D638	33	65	53	35	25	110	39
Specific Tensile		37	61	54	28	20	89	31
Strength, Mpa Elongation at Break %	D638	>>10	2,2	3	—	—	2,5	2,3
Flexural Strength, Mpa	D790	41	98	80	63	48	131	62
Specific Flexural		46	92	82	50	38	107	49
Strength, Mpa Flexural Modulus, Gpa	D790	1,4	7,3	3,9	4,3	3,1	6,2	6,9
Specific Flexural		1,6	6,8	4,0	3,4	2,5	5,0	5,5
Modulus, Gpa Notched Izod Impact J/m	D256A	24	32	21	32	32	107	27
Specific Gravity		0,9	1,07	0,98	1,27	1,25	1,23	1,26
Water Absorption % 24 hrs	D570	0,02	1,05	0,95	0,02	0,02	0,06	0,03
Mold (linear) Shrinkage cm/cm		0,0028	0,003	—	0,01	0,01	0,004	—

are higher for 50% kenaf fiber filled PP as compared to all other filled PP materials.

Figure 1 shows a stress-strain curve for PP alone and in combination with various amounts of kenaf fiber either using a coupling agent (MAPP) or uncoupled. The level of MAPP in these experiments was 2%. A small amount of the MAPP (0.5% by weight) improved the flexural and tensile strength, tensile energy absorption, failure strain and un-notched Izod impact strength. The anhydride groups present in the MAPP can covalently bond to the hydroxyl groups of the surface. Any MA that has been converted to the acid form can interact with the fiber surface through acid-base interactions. The improved interaction and adhesion between the fibers and matrix leads to better matrix to fiber stress transfer. There was little difference in the properties obtained between the 2% and 3% (by weight) MAPP systems. The drop in tensile modulus with the addition of the MAPP is probably due to polymer morphology. Transcrystallization and changes in the apparent modulus of the bulk matrix can result in changes in the contribution of the matrix to the composite modulus and will be discussed later. There is little change in the notched impact strength with the addition of the MAPP, while the improvement in un-notched impact strengh is significant. In the notched test, the predominant mechanism of energy absorption is through crack propagation as the notch is already present in the sample. Addition of the coupling agent has little effect in the amount of energy absorbed during crack propagation. On the order hand, in the un-notched test energy absorption is through a combination of crack initiation and propagation.

Cracks are initiated at places of high stress concentration such as the fiber ends, defects, or at the interface region where the adhesion between the two phases is very poor. The use of the additives increases the energy needed to initiate cracks in the system and thereby results in improved un-notched impact strength values with the addition of the MAPP.

Use of the MAPP increases the failure strain and the tensile energy absorption. Thermodynamic segregation of the MAPP towards the interphase can result in covalent bonding to the -OH groups on the fiber surface. Entanglement between the PP and MAPP molecules results in improved interphase properties and the strain to failure of the composite. There is a plateau after which further addition of coupling agent results in no further increase in ultimate failure strain. Any further increase in the amount of MAPP does not increase the failure strain past the critical amount. However, a minimum amount of entanglements are necessary through the addition of about 1.5% by weight for the critical strain to be reached.

Tensile Strength and Modulus

Figures 2 and 3 show the tensile strength and tensile modulus properties of the agro-resource polypropylene based composites. For purposes of comparison, 100% PP, 40% talc and 50% wood flour composites are included because they are commercially available prod-ucts. The figures show that the tensile strength properties of kenaf core,

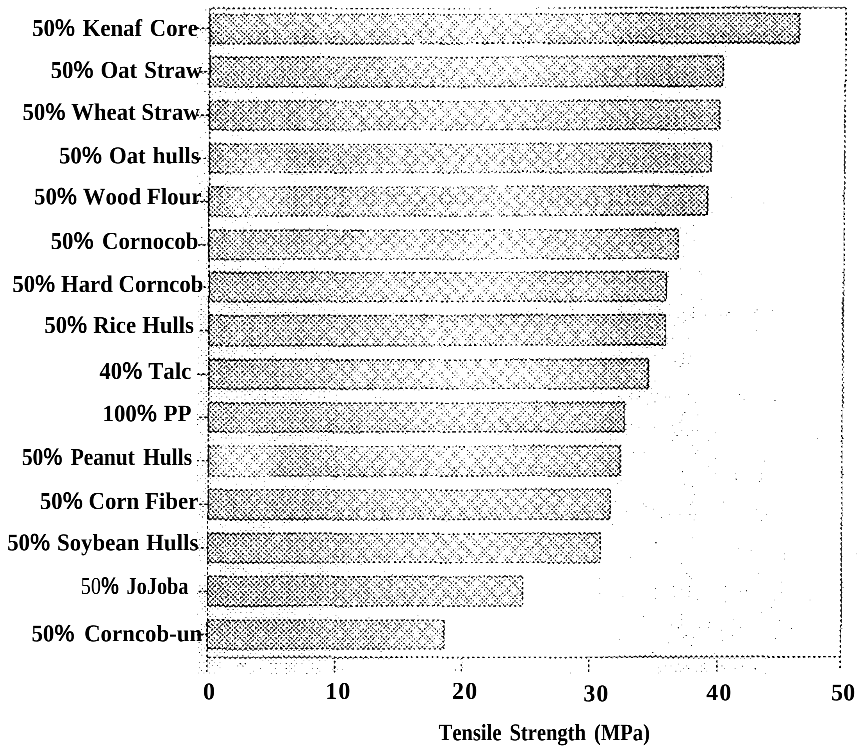

Figure 2. Tensile strength properties of agro-resource-PP composite materials.

oat straw, wheat straw and oat hulls compare favorably to both flour and talc filled PP composites. All other fillers have mechanical properties which are slightly less then the 50% wood flour, but still compare faborably to 40% talc filled PP. the 50% corncob-un is an uncoupled system (*i.e.* no MAPP) and is used for comparison vs. the 50% corncob. The tensile strength of the un-coupled system increased 95% with the addition of 2% MAPP for the coupled corncob composite. All of the filler systems will react in a similar fashion when no MAPP is used during the compounding stage [Jacobson *et al*. 1995].

The tensile modulus of agro-resource composites show dramatic property improvement characteristics vs. 100% PP. Wood fllour shows a 225% increase in modulus, while kenaf core, oat straw and wheat straw show 200% increase in modulus vs. 100% PP. Other systems, such as, oat hulls, corncob, hard corncob and rice hulls show an increase in modulus of 100% vs. virgin PP. For coupled and uncoupled systems, There is little change in the tensile modulus properties between 50% corncob and 50% corncob-un. Previous work at the Forrest Products

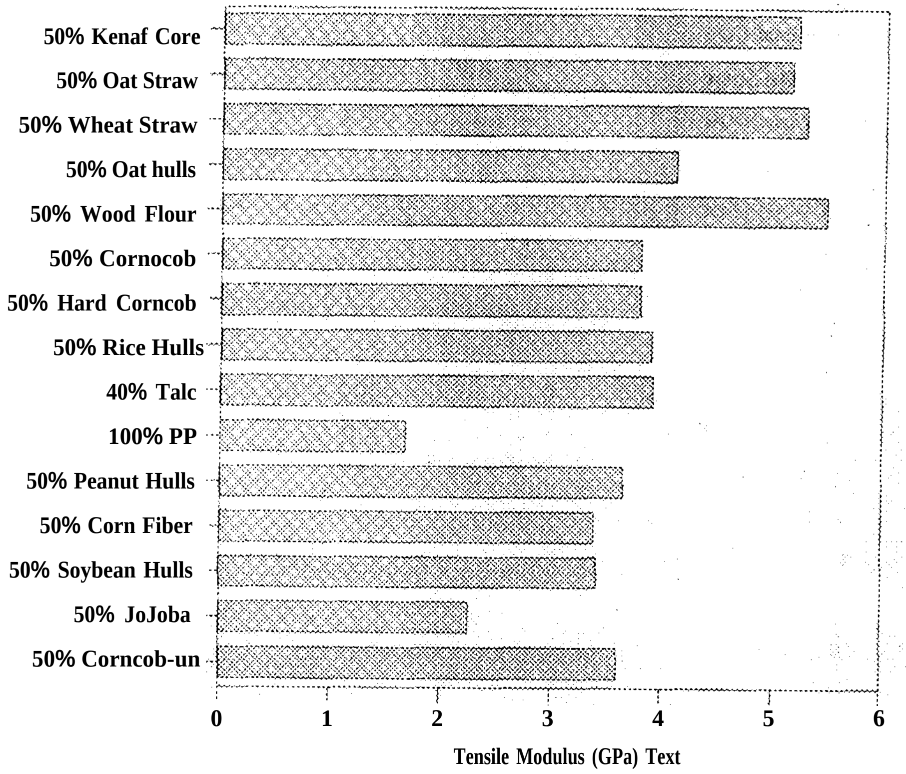

Figure 3. Tensile modulus properties of agro-resource-PP composite materials.

Laboratory [Sanadi *et al.* in press a] indicates that some fiber systems will show a decrease in tensile modulus with a coupled system. Therefore, no discussion of the other fiber systems will be presented until further testing is done.

The addition of MAPP has the most dramatic effect on the tensile strengths of agro-resource composites. The uncoupled fiber systems have strengths approximately half that of coupled systems. MAPP migrates to the interface between the non-polar PP and polar fiber surfaces. In addition, the maleic anhydride present in the MAPP can covalently link to the hyfroxyl groups on the lignocellulosic fibers. Under a tensile load, the improved adhesion at the fiber/matrix interface results in a more efficient stress transfer from the matrix to the reinforced fillers. As a result, strength properties of agro-resource compositres can be improved with a small additions of MAPP.

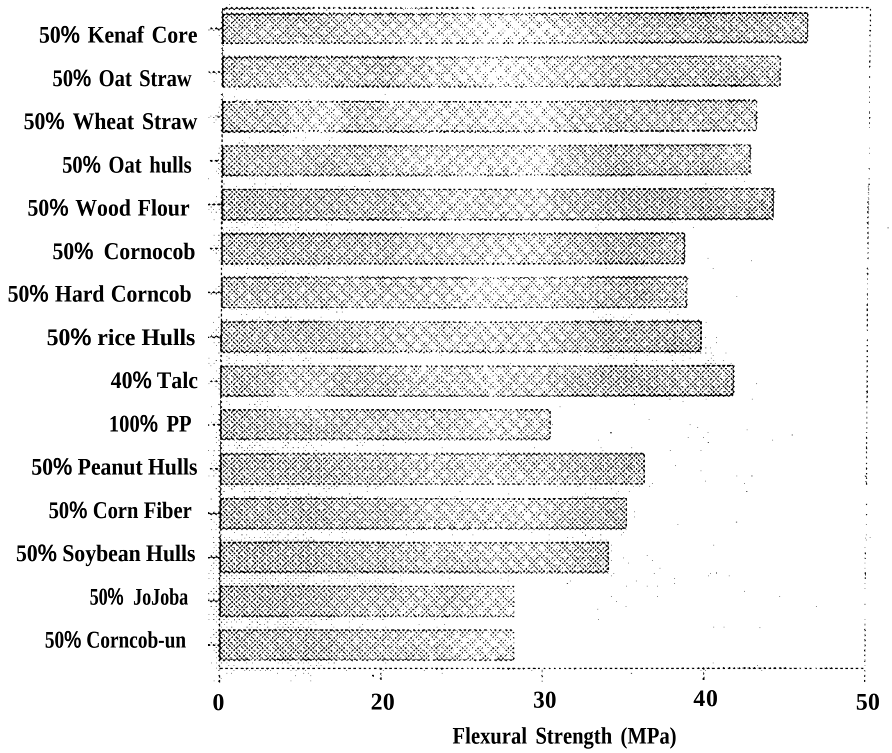

Figure 4. Flexural strength properties of agro-resource-PP composite materials.

Flexural Strength and Modulus

Figures 4 and 5 show the flexural strength and flexural moduli for agro-resource PP based composites. The addition of MAPP increases the flexural strength of these agro-resources composites by approximately 50% of the value of un-coupled systems. Values for the 50% corncob-un and 50% corncob in Fig. 4 shows an increase of 57% for a coupled system and is an indication of how other composite systems flexural strength would be if no MAPP were present in the composites. Increased adhesion between the lignocellulosic fibers and the matrix provides for increased stress transfer from the matrix to the filler. This results in an increased stress at failure and the higher values for flexural strength in the coupled systems verses un-coupled systems the flexural strength of composite systems composed of kenaf core, oat straw, wheat straw and oat hulls are equivalent or superior to both wood flour and talc filled polypropylene composites. The kenaf core com-

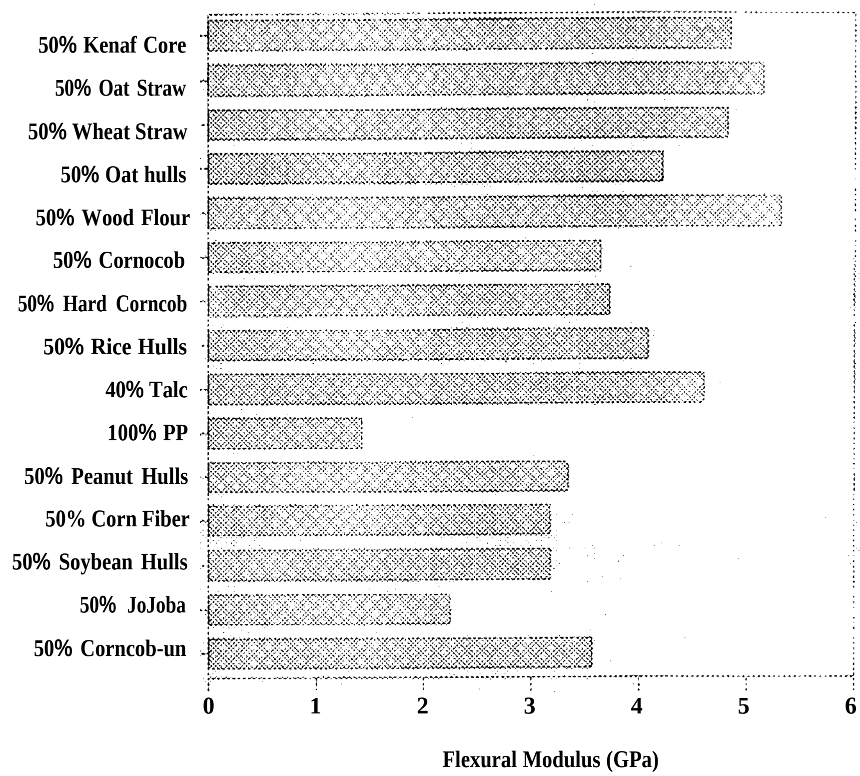

Figure 5. Flexural modulus properties of agro-resource-PP composite materials

posite shows an increased flexural strength of 75% vs. 100% PP. Other systems, such as, corncob, hard corncob and rice hulls have flexural strength slightly less then wood flour and talc filled composites. These systems still show an increased in flexural strength of approximately 50% over virgin PP.

In terms of the flexural modulus, Fig. 5 shows that wood flour has the highest flexural modulus, with an increase of 279% over 100% PP. On average, the top eight agro--waste composites show an increase in flexural modulus of 200% over virgin PP. The kenaf core, oat straw and wheat straw composites have flexural moduli between flour and talc filled composites, while all other agro-resource/PP composites have flexural moduli less then talc and wood flour.

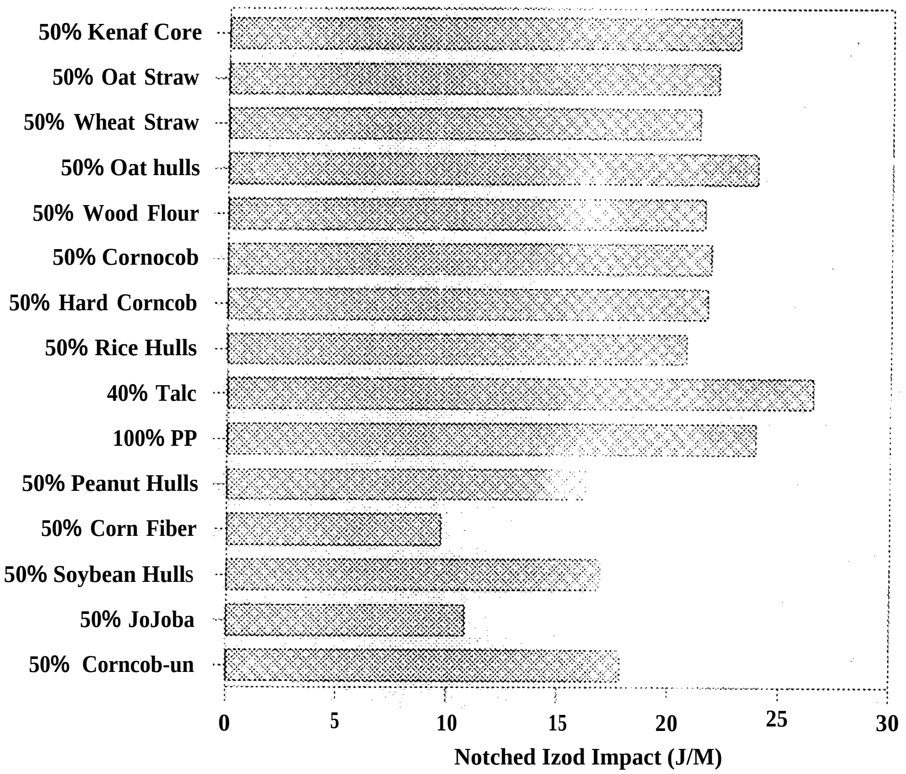

Figure 6. Notched lzod impact properties of agro-resource-PP composite materials.

Notched and Un-notched Izod Impact

Figures 6 and 7 show the notched and un-notched Izod impact properties for the agro-resource composites. The commercially available 40% talc/PP has a notched impact toughness of 26.7 J/M, while 100% PP has a value of 24 J/M. Various types of talc filled PP is available and the notched Izod impact toughness can range up to 75 J/M. The mechanism for the touoghness is due to the plate-like particles of talc which have a higher aspect ratio then the finely ground (i.e. 30 mesh) agro-wastes. Overall, the top eight agro-waste composites notched Izod impact toughness equals 22.1 J/M (i.e. average) and compares favorably to the wood flour. In comparing the agro-resource composites to the talc filled PP and 100% PP, there is an 8-10% decrease in the notched toughness.

Figure 7 shows the un-notched Izod impact toughness of 100% PP equals 640 J/M, while the 40% talc filled PP equals 240 J/M. The average value

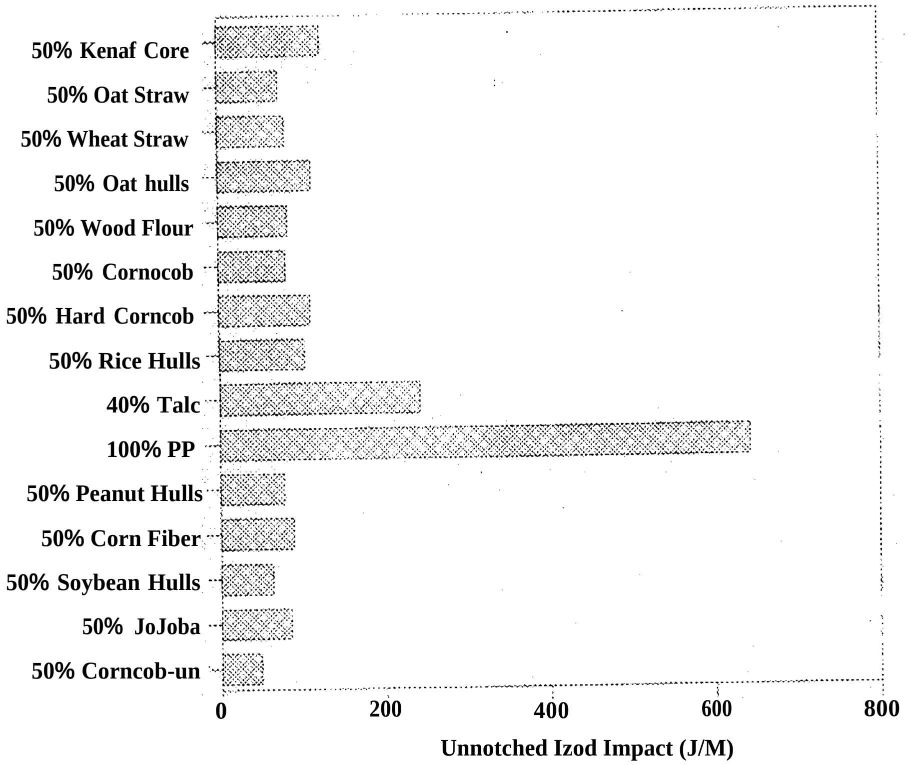

Figure 7. Unnotched Izod impact properties of agro-resource-PP composite materials.

for the agro-resource composites is approximately 95 J/M. The addition of MAPP results in a 100% increase in the un-notched toughness over un-coupled systems. the 50% corncob-un has a value of 40 J/M, while the 50% corncob has a value of 80 J/M. furgher research and development involving impact copolymers will improve the toughness of the composites, but at a loss in strength properties [Jacobson *et al.* 1995].

Impact Properties

The impact strength of the composite depends on the amount of fiber and the type of testing. *i.e.*, whether the samples were notched or un-notched. In case of notched samples, the impact strength increases with the amount of fibers added until a plateau is reached at about 45% fiber weight, irrespective of whether MAPP was used or not. the fibers bridge cracks and increase the resistance of the propagation of the crack. Contribution from fiber pullout is limited since the aspect ratio of the fibers in the system are well below the estimated critical aspect ratio of about 0.4 mm [Sanadi et al. 1993]. In case of the un-notched impact values of the uncoupled composites, the presence of the fibers decrease the energy absorbed by the specimens. Addition of the fibers creates regions of stress concentration that require less energy to initiate a crack. Improving the fiber-matrix adhesion through the use of MAPP increases the resistance to crack initiation at the fiber-matrix interface and the fall in impact strength with the addition of fibers not as dramatic. The impact strength can be increased by providing flexible interphase regions in the composite or by using impact modifiers.

Recycling/Reprocessing

Agro-based fibers are less britte and softer than glass fibers and are likely to result in composites that are easier to recycle than mineral based fibers. Although no post-consumer based recycling studies have been done on agro-based fibers a short study on the effect of reprocessing has

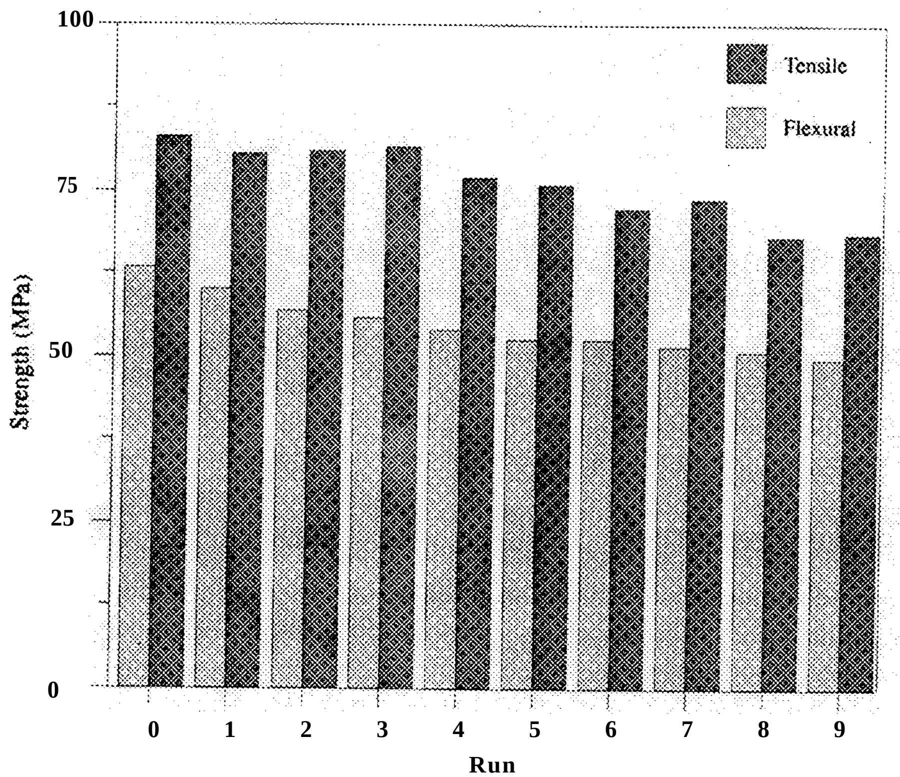

Figure 8. Effect of reprocessing 50% by weight kenaf-MAPP coupled-PP on composite tensile strength. The numbers in the abscissa indicate the number of times the composites were reprocessed.

been conducted at the Forest Products Laboratory and University of Wisconsin-Madison [Sanadi et al. in press b]. Experimental details are as follow:

Short kenaf filaments were compounded with polypropylene (Fortilene-1602, Solvay Polymers) and MAPP using the thermo-kinetic mixer explained earlier in the text. The blend ratio was 50% kenaf to 49% PP to 1% MAPP, based on dry weight of material. the mixer was operated at 5200 rpm. A total of 2.25 Kg (15 batches of 150 g each) of material was blended for the experiment.

All the compounded material was then granulated, dried at 105°C for 4 h and then molded at 190°C using the injection molder. Specimens were randomly selected to evaluate the tensile, flexural and impact properties and five samples were used for each test: this first set of data was the control or virgin data and is denoted by "0" in Figs. 8, 9 and 10.

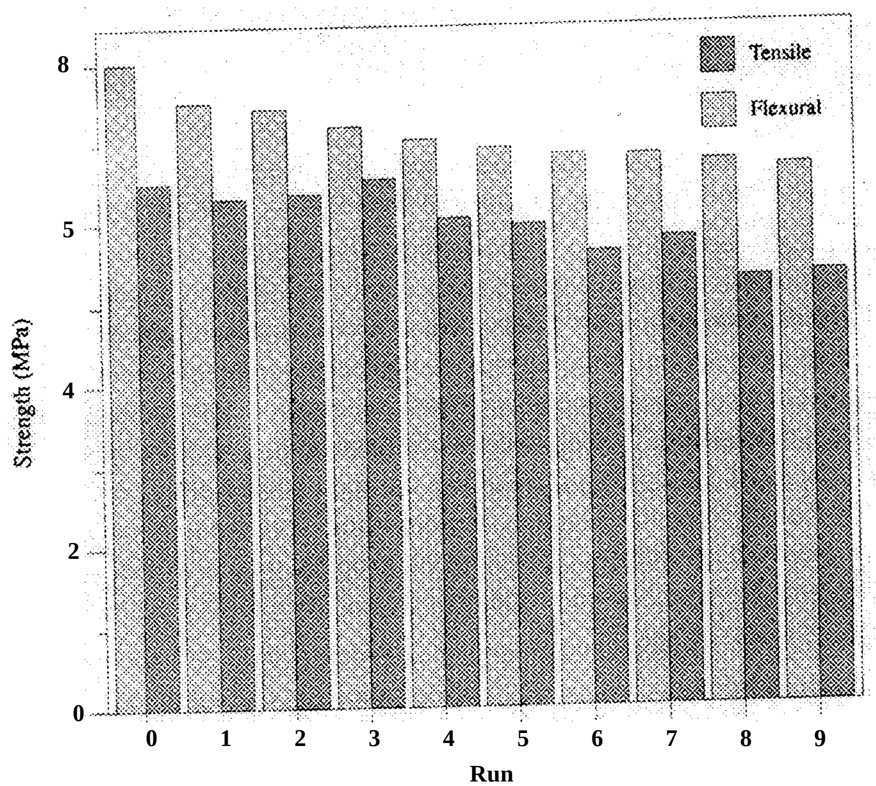

Figure 9. Effect of reprocessing 50% by weight kenaf-MAPP coupled-PP on composite tensilemodulus. The numbers in the abscissa indicate the number of times the composites were reprocessed.

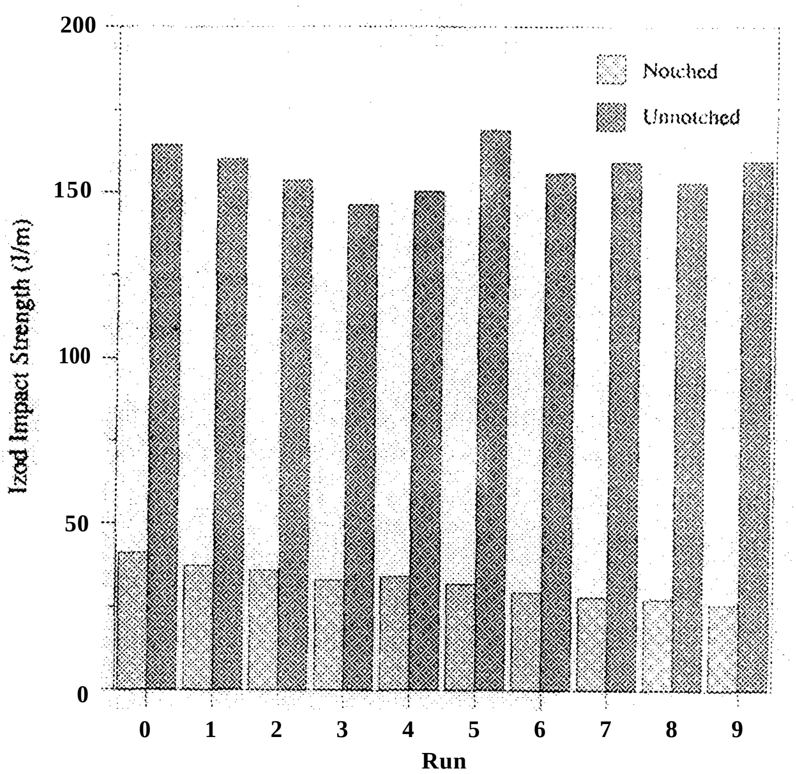

Figure 10. Effect of reprocessing 50% by weight kenaf-MAPP coupled-PP on composite Izod impact strength. The numbers in the abscissa indicate the number of times the composites were reprocessed.

All the remaining non-tested specimens were once again granulated and the injection molded. Once again five specimens were randomly selected for mechanical properties evaluation: this set was labeled as the 1st recycle data point. This procedure of injection molding and granulated was repeated for a total of nine recycle data points. Figs. 8, 9 and 10 show that the repeated grinding and molding does cause a deterioration of composite properties. The loss in properties is a combination of repeated fiber attrition and oxidative degradation of the polypropylene through chain scission.

Conclusion, Economic Aspects and Potential Markets

The cost of natural fibers are in general less than of the plastic and high fiber loading can result in significant material cost savings. The cost of compounding is unlikely to be much more than for conventional mineral/inorganic based presently used by plastics industry. Due to the lower specific gravity's of the cellulosic based additives (approximately 1.4 as compared to about 2.5 for mineral based systems), composite properties considering the weight of the composite is an advantage that may have implications in the automotive and transportation applications. Furthermore using the same weight of plastic/natural fiber, as for example plastic/glass fiber, about 20% more pieces are possible with the cellulosic based system. Cellulosic fibers are soft and non-abrasive and high filling levels are possible. Reduced equipment abrasion and the subsequent reduction of re-tooling costs through the use of agricultural based fibers is a factor that is definitely a factor that will be considered by the plastics industry when evaluating these natural fibers. It is important to point out we do not anticipate nor intend the total replacement of conventional based fillers/fibers with agricultural based fillers/fibers. We do, however, believe that these natural material will develop their own niche in the plastics filler/fiber market in the future.

The volume of thermoplastics used in the housing, automotive, packaging and other low-cost, high volume applications is enormous. Recent interest in reducing the environmental impact of materials is leading to the development of newer materials or composites that can reduce stress to the environment. In light of petroleum shortages and pressures for decreasing the dependence on petroleum products, there is an increasing interest in maximizing the use of renewable materials. The use of agricultural resources as source of raw materials to the industry not only provides a renewable source, but could also generate a non-food source of economic development for farming and rural areas. Appropriate research and development in the area of agricultural based fillers/fibers filled plastics could lead to new value-added, non-food uses of agricultural materials.

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