

COMPOSITES FROM RECYCLED WOOD AND PLASTICS

by

John A. Youngquist
George E. Myers
James H. Muehl
Andrzej M. Krzysik
Craig M. Clemons
USDA Forest Service
Forest Products Laboratory
Madison, WI 53705-2398

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Project Officer

Lisa Brown
Waste Minimization, Destruction, and Disposal Division
Risk Reduction Engineering Laboratory
Cincinnati, OH 45268

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RISK REDUCTION ENGINEERING LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CINCINNATI, OH 45268

NOTICE

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FOREWORD

Today's rapidly developing and changing technologies and industrial products and practices frequently are accompanied by the increased generation of materials that, if improperly dealt with, can threaten both public health and the environment. The U.S. Environmental Protection Agency (EPA) is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. These laws direct the EPA to conduct research to define environmental problems, measure impacts, and search for solutions.

The Risk Reduction Engineering Laboratory is responsible for planning, implementing, and managing research, development, and demonstration programs to provide an authoritative, defensible engineering basis in support of the policies, programs, and regulations of the EPA with respect to drinking water, wastewater, pesticides, toxic substances, solid and hazardous wastes, Superfund-related activities, and pollution prevention. This publication is one product of that research and provides a vital communication link between the researcher and the user community.

Passage of the Pollution Prevention Act of 1990 marked a strong change in U.S. policies concerning the generation of hazardous and nonhazardous wastes. This bill implements the national objective of pollution prevention by establishing a source reduction program at the EPA and by assisting States in providing information and technical assistance regarding source reduction. In support of the emphasis on pollution prevention, the technology evaluation project discussed in this report emphasizes the study and development of methods to reduce waste.

E. Timothy Oppelt, Director
Risk Reduction Engineering Laboratory

ABSTRACT

Wastepaper and wood materials constitute more than 40 percent by weight of municipal solid waste; waste plastics constitute approximately 7 percent. This materials resource offers considerable potential as ingredients in wood fiber-plastic composites. These composite systems, in turn, offer considerable potential for reducing municipal solid waste by the manufacture of an array of value-added products that may themselves be recyclable.

The ultimate goal of this program was to develop technology to convert recycled wood fiber and plastics into durable products that are recyclable and otherwise environmentally friendly, and thereby effectively remove the waste materials from the waste stream. The program employed two processing technologies to prepare the composites: air-laying and melt-blending. Research was conducted in (1) developing laboratory methods for converting wastewood, wastepaper, and waste plastics into forms suitable for processing into composites; (2) optimizing laboratory methods for making composite panels from the waste materials; (3) establishing a database on the effects of formulation and bonding agent variables on composite physical and mechanical properties; (4) establishing the degree to which the composites can be recycled without unacceptable loss in properties; and (5) reaching out to industry to provide education, to develop applications, and to extend the database as appropriate.

Overall, the program demonstrated that both air-laid and melt-blended composites could be made from a variety of wastewood, wastepaper, and waste plastics. These composites exhibit a wide range of properties that should make them useful in a wide variety of commercial applications. Specifically, improvements in laboratory methods to convert waste materials to usable forms and improvements in methods to prepare composites provided the capability to make composites with reproducible properties.

For air-laid composite technology, the waste materials emphasized were demolition wood waste and waste plastics from milk bottles (polyethylene) and beverage bottles (polyethylene terephthalate). Test results showed that air-laid composites made from these waste ingredients possessed properties very similar to those for composites made from the virgin ingredients. In addition, air-laid panels containing 20 percent reground panels as part of their filler possessed some properties that were superior to those of the original panels. This finding provides strong evidence that air-laid panels are recyclable.

For melt-blended composites, the waste materials emphasized were wastepaper, polyethylene from milk bottles, and polypropylene from automobile battery cases or ketchup bottles. Waste magazines were slightly inferior to waste newspapers as a reinforcing filler; the properties of composites made from waste newspaper were better than those of composites made from wood flour, which is currently used in some commercial composites. The various plastics resulted in composite properties that were generally parallel to those of the plastics; thus, different balances in composite properties are possible from using waste plastic. Two melt-blended composites and one air-laid composite were subjected to five extrusion cycles without major changes in properties.

Extensive outreach activities were conducted, including the organization and presentation of two international conferences on wood fiber-plastic composites, presentations at many conferences, publication of several papers, and several spin-off cooperative studies with industry to examine the potential of various waste materials in composites. One major study with industry demonstrated the commercial feasibility of melt-blended composites made from old newspapers and polypropylene.

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LIST OF ABBREVIATIONS

AL	-- air-laid composite
ASTM	-- American Society for Testing and Materials
BPP	-- polypropylene from recycled auto battery cases
BW40	-- Solka-Floc cellulose fiber
DF	-- demolition wood fiber obtained from wastewood
DNR	-- Wisconsin Department of Natural Resources
E10	-- Eastman Chemical Co. Epolene E10, low molecular weight maleated polyethylene
E43	-- Eastman Chemical Co. Epolene E43, low molecular weight maleated polypropylene
E43E	-- emulsified potassium salt of E43
E43S	-- solid powdered form of E43
FPL	-- Forest Products Laboratory
G3002	-- Eastman Chemical Co. Epolene G3002, high molecular weight maleated polypropylene
GPa	-- gigapascal; 10^9 Pa; unit of pressure
HF	-- virgin Western Hemlock fiber
HDPE	-- virgin high density polyethylene
HDPE-MB	-- recycled high density polyethylene from milk bottles
J	-- Joule; unit of energy (work, heat)
KPP	-- recycled polypropylene from ketchup bottles
MB	-- melt-blended composite
MFI	-- melt flow index
MOE	-- modulus of elasticity
MOR	-- modulus of rupture
MPa	-- megapascal; 10^6 Pa; unit of pressure
MSW	-- municipal solid waste
OMG	-- old magazines
ONP	-- old newspapers
PET	-- polyester fiber
PP	-- polypropylene
PR	-- phenolic resin
RH	-- relative humidity
RHDPE	-- recycled high density polyethylene from milk bottles (granulated)
RPET	-- recycled polyester fiber from beverage bottles
SG	-- specific gravity
TAPPI	-- Technical Association of the Pulp and Paper Industry
VHDPE	-- virgin high density polyethylene (granulated)
VPET	-- virgin polyester
VPP	-- virgin polypropylene
WF	-- wood flour

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SECTION 1

INTRODUCTION

Each year the United States has been attempting to dispose of 160 million tons of municipal solid waste (MSW) and is finding it increasingly difficult to find landfill sites that neighbors to landfills will tolerate. In the next 15 years 75 percent of all our landfills will be closed, and by the year 2000 this nation will be short 56 million tons per year of disposal capacity.

In 1986, paper and paperboard, wood, and plastics in the MSW stream accounted for approximately 65, 5.8, and 10.3 million tons, respectively. By the year 2,000, these figures are expected to increase to 86.5, 6.1 and 15.7 million tons annually. In addition to the wood fiber in the MSW stream, there are vast quantities of low-grade wood, wood residues, and industry-generated wood waste in the form of saw dust, planer shavings, chips, and other such items that are now being burned or otherwise disposed of.

Technology is evolving that holds promise for using waste or recycled wood and plastics to make an array of high performance products that are, in themselves, potentially recyclable. However, much research remains to be done. Preliminary research at the USDA Forest Service Forest Products Laboratory (FPL) indicated that recycled plastics such as polyethylene, polypropylene, or polyethylene terephthalate can be combined with wastewood fiber to make useful composites. Advantages associated with these composite products include improved stiffness and strength, acoustic, and heat reformability properties, all at costs lower than comparable products made from plastics alone. In addition, early research has shown that the composite products can possibly be reclaimed and recycled for the production of second-generation composites.

Many of the uses of wood fiber/plastic composites are such as to require them to be opaque, colored, painted, or overlaid. Consequently, recovered fibers or resins used in these composites do not require the extreme cleaning and refinement needed when they are to be used as raw materials for printing paper or pure plastic resins. This fact greatly reduces their cost as raw materials and makes composite panels an unusually favorable option for the recycling of three of our most visible and troublesome classes of MSW—newspapers, wastewood, and plastic bottles.

The research program outlined here focused on melt-blending and nonwoven web technology.

Melt-blending technology—Melt-blending is an inherently low-cost, high-production rate process in which wood and paper are mixed with molten plastic. These blends can then be formed into products using conventional plastics processing techniques such as extrusion or injection molding. The plastic acts as a means to convey the wood/paper during processing and the wood/paper fiber bears the load in the final composite, offering an effective balance between processability and strength of the end-product. With melt-blending techniques, wood flour and wood fibers offer a number of advantages as reinforcements in thermoplastic composites. These include economy on a cost per unit volume basis, desirable aspect ratios, flexibility (hence less fiber breakage), and low abrasiveness to equipment. Composite panels can be produced containing up to 50 weight percent wood fiber and are low cost, thermoformable, and relatively insensitive to moisture.

Air-laid web technology—A wide variety of wood fibers and synthetic plastic fibers can be assembled into a random web or mat using an air-forming web technology. In contrast to melt-blended composites, air-forming technology involves room temperature mixing of lignocellulosic fibers or fiber bundles with other long fibrous materials (usually synthetic

fibers). Nonwoven processes allow and tolerate a wide range of waste paper and wood types depending upon the end product under consideration. With this technology, the amount of paper or wood fiber can be greater than 90 weight percent. After the fibers are mixed they are air-laid into a continuous, loosely consolidated mat. The mat then passes through a secondary operation in which the fibers are mechanically entangled or otherwise bonded together. This low density mat may be a product in itself, or the mat may be shaped and densified by a thermoforming step. Alternatively, a thermosetting resin can be incorporated to provide additional bonding of the fibers. Compared to products made by melt-blending, the air-formed web composites have the potential for better mechanical properties and the capability to make products having more intricate shapes.

Because of the increased processing flexibility inherent in both the melt-blending and air-formed web technologies, a host of new natural fiber/synthetic plastic products can be made. These can be produced in various thicknesses from a thin material of 3 mm to structural panels up to several centimeters thick. A great variety of applications are possible because of the many alternative configurations of the product. Three major use classifications can be defined as packaging products, industrial products, and building materials. Taken together, these two processing techniques provide options for balancing performance properties and costs, depending upon the product application under consideration.

The ultimate goal of this research program was to develop technology to convert recycled wood fiber and plastics into durable, long service life products that are recyclable and otherwise environmentally friendly and will effectively remove the raw materials from the waste stream. In support of this goal, specific research objectives included the following tasks:

Task 1. Raw materials preparation—developing laboratory methods for converting wastewood and waste plastics into forms suitable for subsequent melt-blending and air-laid web processing.

Task 2. Laboratory methods for making composites—optimizing laboratory methods for making composite panels from these waste materials.

Task 3. Development of a performance database—establishing a database on the effects of formulation and bonding agent variables and the resultant physical and mechanical properties of the composite.

Task 4. Recyclability—defining the degree to which the composites can be recycled without unacceptable loss in processability or mechanical and physical properties.

Task 5. Product application and database extension—identifying, in partnership with industry cooperators, product applications and extending the property database to these applications.

Our approach was an exploratory and experimental pursuit of methods that may lead to the fabrication of useful, long-living, recyclable composites from blends of recycled wood and plastic.

SECTION 2

CONCLUSIONS

GENERAL

This program has demonstrated that air-laid and melt-blended composites can be made from a variety of wastewood, wastepaper, and waste plastic materials in the postconsumer waste stream. These composites exhibit a wide range of properties that should make them useful in a wide variety of commercial applications and value-added products.

AIR-LAID COMPOSITES

- (1) The Rando-Webber air-forming equipment can be adapted to handle both long and short synthetic and natural fibers and as well as powders. Nonwoven air-laid webs can be produced that have excellent uniformity in both the machine and cross machine direction.
- (2) Postconsumer high density polyethylene from milk bottles cannot be respun into a fiber using melt processing techniques. Granulating this material for further processing was extremely successful. Recycled and granulated high density polyethylene can be used in the FPL air-forming equipment to produce an air-laid web that can be subsequently made into flat panels or shaped sections.
- (3) Pressure refining techniques can convert postconsumer demolition wood or construction waste into fiber bundles that can be processed very successfully in the FPL air-forming equipment and subsequently into pressed flat panels or shaped sections.
- (4) There were statistical differences in the mechanical and physical properties of test panels made with virgin fiber compared to recycled polyethylene terephthalate and test panels made with hemlock fiber compared to demolition wood fiber. Statistically significant differences were shown in 12 of 40 comparisons. However, the relative magnitude of the difference in these 12 comparisons is small and would most likely have little or no influence on commercial products.
- (5) Demolition wood fiber, which consists of a mixture of up to 15 different hardwood and softwood species, performed as well as panels made from virgin hemlock fiber.
- (6) Panels containing recycled high density polyethylene from milk bottles had mechanical and physical properties similar to those of panels containing virgin high density polyethylene. Sixteen of 40 comparisons exhibited statistically significant differences. However, the character and relative magnitude of the differences in these comparisons would most likely have little or no influence on commercial products.
- (7) Second-generation panels made with a portion of reground first-generation panels performed better than first-generation panels.

MELT-BLENDED COMPOSITES

- (1) Melt-blended composites cannot be prepared with wastepapers as reinforcing filler using a conventional laboratory single-screw extruder. However, they can be prepared with a laboratory high intensity K-mixer. They can also be prepared with an industrial scale K-mixer or with a twin-screw extruder that employs a properly designed feeder for the fiber.

(2) Old newspaper as reinforcing filler provides property advantages over wood flour, which is currently used as filler in commercial composites. With virgin polypropylene as matrix, for example, tensile strength is increased nearly 30 percent and unnotched impact energy is increased over 40 percent. Old magazines can also be used as a filler, but they are less easily dispersed into the matrix plastic and result in somewhat lower properties than those of composites containing old newspaper.

(3) With the same filler, substituting recycled milk bottle polyethylene for virgin polypropylene leads to lower strength (greater than 20 percent reduction), stiffness (greater than 20 percent reduction), and unnotched impact energy (nearly 30 percent reduction), but higher notched impact energy (nearly 30 percent increase).

(4) Use of recycled high density polyethylene from milk bottles and recycled polypropylene from battery cases as a matrix in composites with old newspaper results in improved impact performance when compared with the performance of composites of virgin polypropylene and newspaper.

(5) The addition of Epolene G3002, a coupling agent, at 3 weight percent of filler results in very useful increases in composite properties, for example, a 30 to 50 percent increase in various strengths and a 70 percent increase in unnotched impact energy. These changes are probably brought about by improved dispersion of the fiber.

(6) Select composite systems showed little or no loss in mechanical properties when repeatedly reprocessed (re-extruded and injection molded).

SECTION 3

RECOMMENDATIONS

Recommendations arising from this program, and other closely related efforts, logically fall into two categories—those related to further research needs and those related to further efforts towards commercialization of composites from wastewood fiber and waste plastics. The two categories are not entirely separate, however, because progress in one may well influence the course of the other.

RESEARCH

In several areas, additional research is needed on both air-laid and melt-blended composites made from recycled wood fiber and plastics to improve properties and processing and thereby increase the number of potential applications.

- (1) Evaluate the potential for making valuable composite materials with other major components of the waste stream, including low density polyethylene, polystyrene, and mixed waste plastics. Investigations into some of these materials had to be eliminated from this program because of funding limitations.
- (2) Verify the recyclability of composites made with the above ingredients. In addition, the improved behavior of second-generation air-laid panels, containing portions of reground first-generation panels, may have major ramifications for recycling and should be further investigated.
- (3) Improve melt-blending processes to achieve better fiber dispersion with minimal fiber breakage.
- (4) Improve the bonding between the wood fiber and plastic matrix in order to enhance physical and mechanical properties. The methods and/or materials used to achieve improved bonding will very likely differ with the matrix plastic.
- (5) Improve the impact energy and the creep resistance (decreased deflection under long-term load). These are now the major limiting properties of these composite systems.
- (6) Determine the resistance of these composite systems to relatively extreme environments and develop means to enhance that resistance. Resistance to moisture, to biodegradation, and to fire are important properties for continued research.

COMMERCIALIZATION

- (1) Major, continuing outreach efforts to industry will be needed to acquaint companies with these types of composite systems, to develop applications, and to cooperate with industry in the development work that will be required to bring products to market.
- (2) Commercial acceptance of melt-blended composites containing wastepaper fiber will not occur to a major extent without additional development work in two areas:
 - (a) Improvements are essential in methods to convert wastepaper on a commercial scale to the physical forms and at costs that are acceptable to industrial users. Costs must at least approach the current cost of wood flour at about \$0.22/kg.

(b) Improvements are also critical in methods to melt-blend (compound) the fiber and plastics on a commercial scale at costs acceptable to melt fabricators (extruders, injection molders, etc). Several methods are already available but no systematic small or large scale evaluations have been performed.

SECTION 4

STUDIES ON AIR-LAID COMPOSITES

GENERAL COMMENTS

Many articles and technical papers have been written and several patents have been issued on both the manufacture and use of air-laid mats containing combinations of textile and cellulosic fibers. This technology is particularly well-known in the consumer products industry. For example, Bither (1980) found that polyolefin pulps can serve as effective binders in air-laid products.

Promising technology is evolving for using waste or low-grade wood blended with plastics to make an array of high-performance reinforced composite products. This technology provides a strategy for producing advanced materials that take advantage of the enhanced properties of both wood and plastics. Benefits associated with these composite products include light weight and improved properties (Youngquist and others 1992).

Brooks (1990) reviewed the history of technology development for the production and use of moldable wood products and air-laid nonwoven mat processes and products. Moldable wood product processing technologies developed by Deutsche Fibrit in West Germany (Brooks 1990), Roberts (1955, 1956), Caron and Grove (1966), Caron and Allen (1966a,b, 1968), and Grove and Caron (1966) in the United States involved the use of wood fibers and binder resins.

In the early 1970s, Brooks (1990) developed a process for producing a very flexible mat using a thermoplastic fiber in combination with a thermosetting resin. Opportunities for combining wood with nonwood materials were reviewed by Youngquist and Rowell (1989). Their paper included a discussion of the materials and properties of composites consisting of wood-biomass, wood-metal, wood-plastic, wood-glass, and wood-synthetic fibers.

Air-laid web composites provide options for balancing performance properties and costs, depending upon the application under consideration. However, poor attraction and low interfacial bonding between the hydrophilic wood and hydrophobic polyolefin limit the reinforcement imparted to the plastic matrix by the wood component. In 1991, Krzysik and Youngquist reported on the bonding of air-laid wood polypropylene fiber composites, when maleated polypropylene was used as a coupling agent between wood and plastic materials.

Wood fiber and old newspaper composites provide a favorable option for recycling paper fiber. These fibers can be chemically modified using an acetylation process. When these materials are converted into panel products, the panels possess greatly improved dimensional stability properties and reduced susceptibility to biodegradation by decay fungi (Krzysik and others 1993).

Our research plan called for completing a systematic, statistically valid series of experiments that compared the mechanical and dimensional stability properties of flat panel composites made from the following virgin and postconsumer wastewood and plastics:

AL Series 1. PET systems

- Virgin wood fiber, virgin polyester fiber (VPET), and phenolic resin (PR)
- Virgin wood fiber, recycled polyester (RPET) fiber, and phenolic resin
- Demolition wastewood fiber, virgin PET fiber, and phenolic resin

- Demolition wastewood fiber, recycled PET fiber, and phenolic resin

AL Series 2. HDPE systems

- Virgin wood fiber and granulated virgin high density polyethylene (VHDPE)
- Virgin wood fiber and granulated recycled high density polyethylene (RHDPE)
- Demolition wastewood fiber and granulated VHDPE
- Demolition wastewood fiber and granulated RHDPE

The experiments were statistically designed to provide a valid quantitative comparison of the range of mechanical and dimensional stability properties available for air-laid composites made from virgin and postconsumer demolition wood fibers and two major postconsumer waste plastic materials.

GENERAL PROCEDURES

In laboratory investigations of air-laid composites, experimental operations generally proceeded through the following sequence of steps: (a) modifying our air-forming equipment to insure that uniform machine and cross machine direction webs could be produced on a routine basis; (b) converting raw materials into forms suitable for use in our FPL air-forming equipment; (c) producing air-formed webs; (d) selecting and stacking the webs to produce mats of a given weight; (e) consolidating the mats in a platen press under sufficient heat and pressure to produce a test panel from which test specimens could be produced; (f) cutting specific test specimens; and (g) measuring properties. Efforts in this program related to (a), (b), and (c) are discussed in Task AL-1, and to (d), (e), and (f) in Task AL-2. All property data are discussed in Tasks AL-3 and AL-4.

TASK AL-1. RAW MATERIALS PREPARATION

The raw materials studied in the air-forming portion of this research program fall into three general classes: cellulosic fibers, plastics, and additives (Table 1).

Cellulosic Fibers

Two basic types of wood fiber were used in both the AL Series 1 and AL Series 2 studies. The first was a virgin western hemlock wood fiber (HF) which was produced in a pressurized single disk refiner from 100-percent pulp-grade chips. The chips were steamed for 2 min at 759 KPa to soften them before refining. After refining, the fibers were then dried at 160°C in a flash tube dryer. This processing sequence produced fibrous strands made of individual fibers, pieces of fiber, and fiber bundles. In this report, these fibrous strands or bundles are referred to as fibers.

The second type of wood fiber used for these studies was demolition wood fiber (DF), derived from wastewood from buildings which had been torn down in the Boston, Massachusetts, area. The demolition wood, originally received in chip form, had to be converted into fiber form. This process required pilot-scale research trials because of the volume of material needed for this program. It was necessary to determine if wood that was so old could even be processed into a fiber and, if so, whether a random mix of aged hardwoods and softwoods would be feasible to use

as a postconsumer wastewood fiber source. The wood waste chips were shredded, hammermilled, and cleaned before being pressure refined in a manner similar to the hemlock. Laboratory trials conducted at FPL confirmed that fiber produced from these wood chips was very similar in appearance and processing capability to that of western hemlock.

Both the hemlock fibers and demolition wood fibers were screened through a +0.9-mm screen to remove the fine dust like material. Nineteen percent of the hemlock material was removed through screening, and approximately 17 percent of the demolition wood was removed as fines. The screening process was necessary for the Series 1 experiments because phenolic resin was used as a binder for the experimental panels. In AL Series 2, phenolic resin was not used and, therefore, the wood fiber was not screened.

Old newspapers were used in an ancillary experiment to determine what effect it would have when added to hardboard. To be used in this study, the newspaper had to be reduced back to fiber form. The refiberization involved hammermilling followed by repulping. The fibers were then mechanically dewatered and oven-dried to a moisture content between 30 and 40 percent. They were again hammermilled and then dried to a final moisture content of 4 to 5 percent. Half of the ONP fibers was used at this point. The other half was hammermilled again using a smaller screen to obtain a smaller fiber.

Plastics

The virgin polyester (VPET) was 5.5 denier (6.1×10^{-7} kg/m), 38 mm long, crimped, and had a softening temperature greater than 215°C. The recycled polyester fiber was spun from recycled soft drink containers and was 6.00 denier, 51 mm long, and crimped. For all of the air-laid experiments, this fiber was used as a matrix fiber which served to hold the wood fibers together within the mat.

The virgin high density polyethylene (VHDPE) was a blow-molding polymer normally used as a feed stock for plastic milk bottles. It came in pellet form with a nominal melt flow index of 0.7. Because the HDPE pellets were too large to be used in the air-laid mat forming process, they were cryogenically ground to a nominal (-)35 mesh size. The recycled HDPE was prepared from milk bottles which had been chopped into approximately 6-mm flakes. These flakes were also cryogenically ground to a (-)35 mesh size in a manner similar to the virgin HDPE. The melt flow index of the recycled HDPE was also 0.7.

We made a very intensive effort to obtain recycled HDPE in fiber form, but were unsuccessful. Although we had granulated RHDPE available to blend with the wood fiber, a textile fiber was preferred if it was available because previous research indicated that a granulated thermoplastic had a greater tendency to shake out of the mat during the fabrication process.

Mr. Harry Fitzgerald, a well-known consultant in the textile industry, was assigned the task of trying to locate a commercial source of HDPE fibers. He made hundreds of contacts with various recycling firms and with companies that process recycled plastics into fibrous forms of one type or another. He also approached commercial laboratories to determine if they could produce this type of fiber. All of these contacts were negative. We found there were a number of reasons it was not available. First of all, economic factors make it very difficult to produce a recycled HDPE fiber cheaper than a virgin HDPE fiber. The cost per pound of the virgin resin to produce HDPE fibers is less than the cost of collecting, sorting, cleaning, reprocessing, and spinning recycled HDPE. Secondly, it is very difficult to get a 100 percent pure grade of HDPE. With impure plastic resin, the melted plastic filament usually has a large variability in diameter. Because of this, it often breaks as it is being spun. This results in chunks of plastic being thrown out from the

spinnerette. These problems cause production of a low quality fiber which is not competitive with virgin HDPE processed in a similar fashion.

Because of the lack of commercial availability of this type of material, we felt that it was worthwhile to try to produce a recycled HDPE fiber in the laboratory. We had a hot-melt extruder for which we purchased a spinner head to make the fibers. When the recycled HDPE milk bottle flakes were introduced into the extruder we found that the material was too viscous for our equipment. We then coupled the spinner head to a vertical single-screw extruder in an effort to increase the head pressure. This change resulted in some improvement, but we were still unable to produce an acceptably uniform fiber with this equipment.

Since our work indicated that it was virtually impossible to buy or produce a recycled HDPE fiber, we found a tackifier, that when sprayed on the wood fiber prior to web formation, resulted in the granular HDPE being retained in the web during the web formation process.

Additives

Phenolic Resin--

The binder for the AL Series 1 panels was a liquid phenolic resin (PR) which was sprayed on the wood fiber at 25°C as it rotated in a drum-type blender. The resin was applied with a pneumatic spray gun at a level of 10 percent solids by weight. The resin had a solids content of 51 to 53 percent, a pH of 9.5 to 10.0 at 25°C, and is infinitely dilutable in water. After application of the resin to the wood fiber, the blend was allowed to air dry over night. This drying period was necessary to prevent the wood fibers from clogging the Rando-Webber forming equipment because the resin was tacky immediately after applying it to the wood fiber.

Tackifier--

In the AL Series 2 boards, the wood fiber was blended with granulated HDPE. During the processing, web manufacturing, and panel making, we found that a lot of the HDPE was being lost throughout the process. This was because the HDPE granules were not held in the matrix of the nonwoven mat. We discovered that the addition of a retention aid in the form of a tackifier on the wood fiber solved this problem. We found the addition of this material almost eliminated the loss of granulated HDPE from the fiber mats as they moved through the processing steps necessary to produce test panels. Preliminary testing also indicated that the tackifier, which was a wax emulsion of oxidized low molecular weight polyethylene (E-10), did not have an adverse effect on the properties of the resultant test panels. This tackifier was applied to the wood fibers in a rotating drum blender with an air spray gun in a manner similar to the way the phenolic resin was applied to the wood fibers in the AL Series 1 study.

Preparation of Second-Generation Recycled Panels

The recyclability part of the study (Task 4) required that we determine the feasibility of recycling panels made for AL Series 2, which contained DF and RHDPE. The panels were cut up into approximately 2- to 4-cm. squares for processing. Hammermilling was found to reduce the fiber length too much and this operation created too many fines. We therefore found we had to recycle the boards through the pressurized refiner to be able to produce the desired fiber length and bundles. Before refining, the composite material was softened by presteaming for 20 min at 68.9 kPa steam pressure. Refining was done using course plates set at a gap of 0.127 mm. When finished, the fiber had an approximate moisture content of 60 percent, requiring it to be dried before further processing. This was done by spreading it out in a thin layer to air dry for 2 days after which it was ready for forming into a nonwoven web. This procedure resulted in a high quality fiber with few noticeable fines.

TASK AL-2. LABORATORY METHODS FOR MAKING COMPOSITES

Equipment Additions and Modifications

The air-laid portion of this study used nonwoven mats made with a 305-mm-wide, lab scale, Rando-Webber forming machine. Our initial efforts in this research program focused on attempting to improve the density profile across the web as it was produced. Preliminary experiments indicated that there was generally more fiber at the center of the web as it formed on the condenser screen than there was on the edges of the mat. Thus, a density gradient across the web was created. This, in turn, caused a density gradient in the test panels and erroneous test results. The effect of the density gradient in an individual web was magnified in the finished panel because as many as six mats were stacked together to produce a finished test panel of the desired thickness and specific gravity.

A number of alterations and improvements were made to the machine in an effort to minimize the density gradient across the web. First, the drive for the elevator which is used to feed the fiber to the feed rolls (Figure 1) was separated from the stripper apron. This allowed both the elevator and the stripper to be driven independently, and provided more flexibility in delivering fiber to the feed rolls. Further improvement in the web density gradient was made by devising baffles in the feeder condenser to divert air flow from the center of the feeder condenser. This directed more fiber to the edges of the mat and away from the center as the fiber was fed into the feed rolls.

Several other changes were made in an effort to further improve density profiles. New feed rolls were machined with a variety of surface patterns to handle the various fibers and fiber blends used in this experiment. These new feed rolls have helped to improve continuous feeding of the fiber into the lickerin, thus allowing a more constant thickness formation of mat in the machine direction. A new nose bar was purchased which had a larger radius than the one used previously. The original nose bar was very sharp and caused some fiber breakage and the generation of a relatively large proportion of fine material. The larger radius bar improved both of these problems dramatically. Further improvement was made in reducing fiber damage and fines creation by using a lickerin with many fewer teeth per increment. Finally, we found it is necessary to use different patterns and styles of needles in the web needler, depending upon the material being processed and the desired configuration of the final web product.

Two hydraulically operated, steam heated, platten presses were obtained through government surplus from the Agricultural Research Service at Peoria, Illinois. These two presses dramatically improved our capability to make high quality panels for this project in a timely manner. Although the presses were serviceable, considerable upgrading and retrofitting was necessary to bring them to an acceptable condition for use on this study. Both presses were steam heated and required new steam controls and temperature regulators. The platens were machined to remove surface flaws and bring them back into parallel alignment. Exhaust fans were installed for the presses, and exhaust hoods were placed above them to remove vapors present when panels are pressed. Both presses were also fitted with thickness measuring gauges, thermocouple meters, and cold water plumbing and controls to provide rapid platen cool down capabilities.

Web Selection Criteria

Webs measuring 305 by 305 mm were weighed and sorted into various weight categories. Appropriate webs were then selectively stacked to arrive at the target panel basis weight. All web sections containing any type of defect were discarded at this point in the panel fabrication process.

Panel Fabrication

The 305- by 305-mm mats were stacked so as to construct multi-layer mattresses of the panel target specific gravity. A manually controlled, steam heated press was used to press all panels. Pressing was at 190°C for 4 min at a maximum pressure of 8.47 MPa; 3 min of cooling time were needed while the board was still under compaction to reduce steam vapor pressure in the pressed board and to maintain the target thickness. Stops were used to produce 3.2-mm-thick panels. After pressing, the panels were trimmed to a final size of 279 by 279 mm.

TASK AL-3. DEVELOPMENT OF A PERFORMANCE DATABASE

Under this task, we carried out the performance evaluation of the panels made for AL Series 1 (PET) systems and AL Series 2 (HDPE) systems. Within each of these two series, a comparison of virgin and recycled materials was made. A statistically valid quantitative comparison of the range of mechanical and dimensional stability properties obtainable for air-formed composites made from virgin and wastewood fibers and two major postconsumer waste plastics is presented in the following discussion.

Prior to mechanical and physical property testing at room temperature (about 23°C) for both AL Series 1 and AL Series 2 studies, the specimens were conditioned at 65 percent relative humidity (RH) and 20°C until equilibrium conditions were achieved. Specimens had minimal exposure to ambient humidity during the time required to complete the testing. Three-point static bending modulus of rupture (MOR) and modulus of elasticity (MOE) and tensile strength and MOE tests were performed in conformance with ASTM D1037 (ASTM 1991) using an Instron testing machine. Impact energy was measured in conformance with TAPPI standard T803 om-88 (TAPPI 1989) using a General Electric impact tester. Thickness swell and water absorption measurements were made by immersing specimens in water in a horizontal position for 24 h at ambient temperature. This test was performed in conformance with ASTM D1037. For linear expansion tests, the specimens were first oven-dried prior to taking specimen length measurements. Linear expansion tests were conducted on length measurements made at equilibrium at 30, 65, and 90 percent RH at 27°C. Linear expansion values were calculated over the following ranges: oven-dry to 30 percent RH, oven-dry to 65 percent RH, and oven-dry to 90 percent RH. Linear expansion tests were determined in conformance with ASTM D1037.

The mechanical, water resistance, and dimensional stability properties of the composite panels in AL Series 1 and 2 are presented in Figures 2 through 21. Each of these figures includes four sets of bar graphs. The first two sets of bar graphs compare performances between composites made with virgin or recycled plastic blended with hemlock fiber in one set and demolition fiber in the second set. Statistical results are discussed for these comparisons. The third and fourth sets of bar graphs in each figure are included for the convenience of the reader and present the same data presented in bar graph sets 1 and 2, but rearranged to point out different comparisons between different combinations of materials.

AL Series 1. PET Systems

Background Information--

This experimental series compared the performance of panels made from virgin wood fiber, virgin polyester fiber, recycled demolition wastewood fiber and recycled polyester fiber. The virgin wood fiber was hemlock (HF) and the demolition wastewood fiber (DF) came from fiberized wood waste from demolished buildings. The recycled polyester fiber was respun from recycled soft drink bottles (Table 1).

Experimental Details--

The experiment consisted of four formulations of wood fiber/synthetic fiber/phenolic resin contents, based on the oven-dry fiber weight (Table 2). A total of 160 panels was made for this AL Series 1 experiment. Each formulation was considered a replicated set that consisted of 40 individual panels. A total of 15 panels was then selected from each set of 40 based upon how close they were to the target specific gravity (Table 3). The selected panels were then cut into specimens for mechanical and physical property testing.

Mechanical and physical property tests were conducted on specimens cut from the selected experimental panels. For all of the formulations, each panel was weighed, measured and the specific gravity was calculated. Panels, from which test specimens came, were selected on the basis of which ones were closest to the target specific gravity of 1.0 ± 0.03 (Table 3), and the target thickness of 3.2 mm. This method of panel selection allowed us to narrow the variability in specific gravity between individual experimental panels.

In this test series, no data outliers were found. Each data set was tested for normality at the 95 percent confidence level using the Shapiro-Wilk statistical analysis. Because there was no indication of non-normality, an analysis of variance was performed and the means were compared at the 95 percent confidence level using Tukey's method of multiple comparisons.

Results and Discussion--

Mechanical and physical property data are presented in Table 4 and in Figures 2 to 11 as bar charts. In all of these figures, bars connected by a solid line indicate that the results are not significantly different at the 95 percent confidence level. A gap in the line indicates a statistically significant difference exists. Each value is an average of 20 tests, with the exception of impact energy, which is an average of 5 tests.

Mechanical properties--The panels made with the HF/VPET formulation had the highest bending MOR value at 50.6 MPa, although no statistically significant differences were observed for MOR values for either wood fiber or PET variations (Figure 2).

A pattern different to that found for bending MOR values was noted for MOE values (Figure 3). In both wood fiber groups, the MOE values of boards containing RPET were significantly higher than those of boards containing VPET. The total average increase for this property was 16 percent for both groups.

For the HF/VPET formulation, tensile strength was 33.0 MPa, whereas it was 14 percent less when RPET fibers were used (Figure 4). However, when RPET or VPET fibers were used with DF fibers, no significant differences were noted.

In contrast to tensile strength, tensile modulus (MOE) (Figure 5) for both HF and DF formulations were increased by 6 and 7 percent by incorporating RPET fibers, although these differences were not statistically significant.

Impact energy of specimens from the HF and DF formulations showed a consistent trend (Figure 6). Formulations containing VPET fibers had impact energy values that were higher by 20 and 10 percent, respectively, compared to formulations containing RPET.

Physical and dimensional stability properties--A 24-h water soak test was used to measure thickness swell and water absorption of boards made from the four formulations. In this AL Series 1 study, formulation had a consistent influence on thickness swell values (Figure 7). Specimens of HF/RPET had the smallest thickness swell values at 22.3 percent, whereas specimens of the DF/VPET formulation had the largest values at 29.8 percent. For these two groups, the values obtained were statistically different from each other.

Results similar to those for the thickness swell test were observed for the water absorption tests, except that no statistical differences were noted for any of the formulations (Figure 8).

Linear expansion values at 30 percent RH were statistically equivalent (Figure 9). At the 65- and 90-percent RH conditions (Figures 10 and 11), the HF/RPET and DF/RPET formulations had slightly higher values and were statistically different from the other two formulations of HF/VPET and DF/VPET.

Conclusions--

In general, the mechanical, water resistance and dimensional stability properties of all panels made from recycled materials were all equivalent to similar properties obtained from panels containing all virgin or virgin/recycled materials. Therefore, the recycled ingredients tested in AL Series 1 could replace virgin materials with minor consequences.

AL Series 2. HDPE Systems

Background Information--

This experimental series compared the performance of panels made from virgin wood fiber, virgin granulated high density polyethylene (VHDPE), virgin polyester fiber (VPET), recycled demolition waste fiber (DF), and postconsumer recycled high density polyethylene (RHDPE) particles. The virgin wood fiber was hemlock (HF) and the demolition wastewood fiber (DF) came from fiberized waste from demolished buildings. The recycled high density polyethylene (RHDPE) particles came from chipped and ground postconsumer milk bottles.

Experimental Details--

This experiment was similar to AL Series 1, except that VHDPE or RHDPE were used as the thermoplastic panel bonding resin, in place of the thermosetting phenolic resin. A tackifier (Epolene E-10) was used in this Series as an aid in retaining the granulated HDPE in the web during the formation process. The experiment consisted of four formulations of wood fiber/granulated HDPE/PET fiber/tackifier content based on oven-dry fiber weight (Table 5). A total of 160 panels (40 per formulation) was made for this AL Series 2 experiment.

For the AL Series 1 composite panels, each panel was weighed, measured and the specific gravity (SG) was calculated. The panels, from which test specimens were obtained, were then selected on the basis of which panels were closest to the target SG of 1.0 and target thickness of 3.2 mm. We found that this method of selection led to a large variability in specimen specific gravity due to a considerable variation in specific gravity within the panel. This fact resulted in us having to make many more panels than were needed to do the actual tests. Therefore, a change in the method of specimen selection was made for the AL Series 2 portion of the program. Specimens were cut from test panels and the specimen specific gravities were calculated. Then, only specimens closest to the specific gravity of 1.0 were used for testing. For AL Series 2 the acceptable range for specimen SG was 1.0 ± 0.06 (Table 6).

Data were first checked for unusual values. Where there were outliers, the analysis was done with and without these values. Each data set was tested for normality at the 95 percent confidence level using the Shapiro-Wilk statistical analysis. If the Shapiro-Wilk statistic indicated that the data were normal, an analysis of variance was done and the means were compared at the 95 percent confidence level using Tukey's method of multiple comparisons. In the cases where normality was rejected, a ranked analysis of variance was done and means (of ranks) were again compared using Tukey's method of multiple comparisons.

Results and Discussion--

Mechanical and physical property data are presented in Table 7 and Figures 12 to 21 as bar charts. In all the figures, the bars connected by a solid line indicate that the results are not

significantly different at the 95 percent confidence level. A gap in the line indicates that a statistically significant difference exists. Each value is an average of 20 tests for static bending MOR and MOE, 20 tests for tensile strength and MOE, 10 tests for impact energy, 40 tests for water soak, and 25 tests for linear expansion.

Mechanical properties--The DF/VHDPE panels had the highest bending MOR value at 19.1 MPa, followed by 18.7 MPa for HF/RHDPE formulation (Figure 12). Generally no statistically significant differences were observed for MOR values for either wood fiber or HDPE variations.

In contrast to MOR values, bending MOE values for panels containing HF/RHDPE exhibited the highest value of 2.13 GPa. The panels made from DF/VHDPE formulation had the lowest value of 1.75 GPa. No statistical differences were noted for MOE values for these test panels (Figure 13).

For tensile strength, the highest value of 12.4 MPa was observed for the DF/VHDPE formulation, whereas the DF/RHDPE panels were 7 percent lower (Figure 14). With both wood fiber variations, the use of either virgin or recycled HDPE did not significantly influence tensile strength values.

The tensile modulus of the HF/VHDPE panels had the highest value at 2.81 GPa, whereas incorporation of RHDPE lowered these values by 21 percent, and the noted difference was statistically significant (Figure 15). The tensile modulus of the DF formulations were about equal, averaging 2.11 GPa.

Impact energy values for specimens made from HF and DF fibers and for specimens made from virgin or recycled HDPE were not statistically different from each other (Figure 16).

Physical and dimensional stability properties--A 24-h water soak test was used to measure thickness swell and water absorption properties of panels made from the four formulations.

Thickness swelling increased by an average of 22 percent as the formulation changed from HF to DF, and the highest value of 53 percent was observed for the DF/RHDPE formulation (Figure 17). It was particularly noticeable that the use of RHDPE significantly influenced only the DF formulation and not the HF formulation. The HF/RHDPE panels exhibited the lowest thickness swell values of 43 percent.

In this study, formulation had a consistent influence on water absorption values (Figure 18). Incorporating RHDPE with either type of wood fiber produced a statistically significant increase in this property by 13 percent on the average. Specimens of the HF/VHDPE and DF/VHDPE formulations exhibited the lowest water absorption values of 55 percent and 59 percent, respectively.

Linear expansion values for all formulations at the 30 percent RH condition ranged from 0.15 to 0.17 percent (Figure 19). At 65 and 90 percent RH, the HF/RHDPE and DF/RHDPE formulations had slightly higher values (Figures 20 and 21). Generally, no obvious trends were observed for linear expansion values at the 30, 65, and 90 percent RH levels for any of the formulations.

Conclusions--

Mechanical and physical properties of panels containing virgin and recycled wood fiber and virgin and recycled polyethylene milk bottle stock had equivalent property values. Therefore, as in AL Series 1, the recycled materials used in AL Series 2 could also replace virgin materials with minor consequences.

TASK AL-4. RECYCLABILITY

Background Information

The purpose of research presented here was to study the recyclability of air-laid composites back into an air-laid composite panel. For this series, the first-generation panels consisted of 60 percent DF, 30 percent RHDPE, 5 percent VPET and 5 percent E-10. The second-generation panels contained 40 percent DF, 20 percent refiberized first-generation, 30 percent RHDPE, 5 percent VPET, and 5 percent E-10.

To be able to recycle the panels made from the first-generation panels, they first had to be cut into approximately 2- by 4-cm squares and then processed into fiber form through pressurized refining. It has been suggested that this process of fiberizing the first-generation panel created shorter and narrower fiber bundles and more small particles, or fines, than were present in the materials that originally were used to make the first-generation panels. This may have allowed a better interfelting of the fibers and could have allowed the fines to fill in gaps between the plastic and wood fibers in the second-generation panels.

Experimental Details

The experiment involved two formulations of wood fiber, granulated HDPE, PET fiber, and tackifier based upon the oven-dry fiber weight (Table 8). Each formulation was considered a replicated set that consisted of 40 individual panels. A total of 80 boards was made for this experiment. The panels of each formulation were cut into specimens for testing mechanical and physical properties (Table 9).

The data were checked for unusual values. Where there were outliers, the analysis was done both with and without these values in the data set. Each data set was tested for normality at the 95 percent confidence level using the Shapiro-Wilk statistical analysis technique. If the Shapiro-Wilk statistic indicated that the data were normal, an analysis of variance was done and the means were compared at the 95 percent confidence level using Tukey's method of multiple comparisons. In the cases where normality was rejected, a ranked analysis of variance was done and means (of ranks) were again compared using the method of Tukey.

Mechanical and physical property tests were conducted on specimens cut from the experimental panels which were made according to AL Series 2 procedures, and test procedures for Task 4 were similar to those used for Task 3, AL Series 1 and 2.

Results and Discussion

Mechanical and physical property data are presented in Table 10 and Figures 22 to 31 as bar charts. These charts are described in the Results and Discussion section for AL Series 2. Each bar on a chart is an average of 20 tests for static bending MOR and MOE, 20 tests for tensile strength and MOE, 10 tests for impact energy, 40 tests for water soak, and 25 tests for linear expansion.

Mechanical Properties--

The second-generation panels had higher bending modulus of rupture (MOR) values at 19.6 MPa, compared to 17.4 MPa for the first-generation formulation (Figure 22). On the other hand, the bending MOE values of panels containing the first-generation formulation exhibited higher values than the second-generation panels (i.e., 2.01 to 1.77 GPa, respectively (Figure 23)). No statistical differences were noted for the formulations.

Second-generation panels exhibited tensile strengths which were 19 percent higher than first-generation panels (Figure 24). Similar statistically different results were obtained for tensile modulus (Figure 25).

The higher property values from the second-generation panels indicate that wood/recycled high density polyethylene composites can benefit by the addition of refiberized first-generation panel material.

Impact energy values of specimens made from first-generation panels and second-generation panels were nearly equal and not statistically different (Figure 26).

Physical and Dimensional Stability Properties--

A 24-h water soak test was used to measure the thickness swell and water absorption properties of panels made from the two formulations.

For thickness swell, a value of 53 percent was observed for the first-generation specimens (Figure 27). Particularly noticeable was the fact that incorporation of first-generation panel fibers into second-generation panels improved this property by 21 percent.

Similar trends were also observed for water absorption values (Figure 28). Specimens of second-generation panels had a value of 54 percent, and was lower than the first-generation specimens by 18 percent. These differences were statistically significant. These results suggest that the additional HDPE from refiberized first-generation panels further encapsulated the wood fibers, thus limiting water uptake by the wood fibers.

Linear expansion values for both formulations at 30 percent RH conditions were very similar, although statistically significant (Figure 29). The incorporation of material from the first-generation panel to the second-generation panel substantially decreased linear expansion at both the 65- and 90-percent RH condition, and these improvements were also statistically significant (Figures 30 and 31).

The positive influence created by incorporating 20 percent of the first-generation panel into the second-generation panel may be due to several factors. When the refiberized first-generation panels were added to the formulation of DF and RHDPE, the percentage of DF was reduced from 60 to 40 percent to accommodate the addition of 20 percent fiberized reground panel. By this method of material addition to the composition, the actual amount of total wood fiber was reduced from 60 to 52 percent, and the total amount of RHDPE increased from 30 to 36 percent. Likewise, the percentage of PET and E-10 each increased by 1 percent. The increase of these components, particularly the HDPE, and the decrease of the wood fiber, may be a direct cause for some of the improvements in property values. By increasing the plastic content, more wood fiber has the possibility of becoming encapsulated by plastic, thereby reducing its exposure to moisture.

Conclusions

In general, the mechanical, water resistance, and dimensional stability properties of all second generation panels made from recycled materials were essentially equivalent or better than properties obtained from first-generation panels. Therefore, the second generation composites, or possibly higher generation composites, can be produced using recycled materials without the consequence of reduced property values.

SECTION 5

STUDIES ON MELT-BLENDED COMPOSITES

GENERAL COMMENTS

Systematic comparisons of composites made from four waste or virgin plastics, three waste lignocellulosics, and one virgin cellulose were carried out in this phase of the research program. Wood flour (WF) was used as the filler in composites using recycled milk bottle high density polyethylene (HDPE-MB) because the high melt viscosity of HDPE-MB leads to potential processing problems when combined with paper fibers. However, the lower melt viscosities of the relevant polypropylenes (PPs) allowed us to incorporate two kinds of wastepaper fibers into the PP composites. The following systems were investigated. Studies MB-1 through MB-4 refer to Task 3; study MB-5 refers to Task 4.

Study MB-1

- Old newspaper (ONP) in virgin polypropylene (VPP) (melt flow index (MFI) 3, 12, and 30) at three filler/plastic ratios
- WF in VPP (MFI 3, 12, and 30) at one filler/plastic ratio
- Epolene E43 at one level (coupling agent)

Study MB-2

- ONP or cellulose (BW-40) in VPP (MFI 12) at one filler/plastic ratio
- WF or BW-40 in HDPE-MB or virgin polypropylene at one filler/plastic ratio
- E43 at one level (coupling agent)

Study MB-3

- ONP in VPP (MFI 12) at one filler/plastic ratio
- E43 or Epolene G3002 at zero or 3 percent (coupling agent)

Study MB-4

- ONP or old magazine (OMG) in recycled ketchup bottle polypropylene (KPP) or recycled auto battery polypropylene (BPP) at one filler/plastic ratio
- G3002 at one level (coupling agent)

Study MB-5 (Recyclability)

- System 1
 - ONP in VPP (MFI 12) at one ONP/PP ratio
 - No coupling agent
- System 2

- WF in HDPE from milk bottles (HDPE-MB) at one WF/ONP ratio
- No coupling agent
- System 3
- Air-laid composite as filler in HDPE-MB at one filler/HDPE-MB ratio
- No coupling agent

GENERAL PROCEDURES

In laboratory investigations of melt-blended composites, experimental operations generally proceed through the following sequence of steps: (a) conversion of raw materials into forms suitable for preparing dry mixtures quantitatively and feeding those mixtures into the melt-blending apparatus; (b) quantitative dry mixing; (c) melt-blending; (d) injection molding of test specimens; and (e) measurement of properties. Efforts in this program related to (a) and (b) are discussed in Task 1, and to (c) and (d) in Task 2. All property data are discussed in Tasks 3 and 4.

Most studies were statistically designed and analyzed, and the results of those analyses are presented in the following. However, we note that statistical analysis has two opposing limitations. First, statistical analysis has a tendency to partially mask the magnitude of effects of specific variables because it averages effects over other variables; we attempt to reduce this tendency by presenting data as graphs in which the influences of specific variables are more readily apparent. On the other hand, it is often the case that an observed change may be statistically significant at a chosen confidence level but still too small to be practically useful in a particular application. To provide statistical rigor and to identify real effects of variables, all statistical comparisons are made at a confidence level of 95 percent. Discussion, however, centers on the larger effects of the variables because of their relative importance and greater impact on composite performance.

TASK MB-1. RAW MATERIALS PREPARATION

Three general classes of raw materials were studied: plastics, cellulose, and additives. These materials are described in Table 11.

Plastics

Plastics act as the matrix in the composite. In this program we employed virgin polypropylene (VPP) homopolymer as our baseline matrix plastic and compared that against three waste plastics. VPP can be purchased at approximately \$0.88/kg. Polypropylene from auto battery cases (BPP) appeared to have considerable utility. The battery cases are nearly 100 percent recycled, and the BPP is available in large quantities in a cleaned form and is currently cheaper than virgin polypropylene (BPP sells for \$0.46/kg to \$0.64/kg). Moreover, BPP is actually an ethylene-propylene block copolymer with small ethylene content, which imparts higher toughness but somewhat lower strength and stiffness compared with polypropylene homopolymer. Recycled ketchup bottle polypropylene (KPP) is an ethylene-propylene random copolymer that is not yet widely available, but it is readily separable from MSW. We felt it would be useful to include it as another representative of recycled polypropylenes. Recycled high density polyethylene from blow-molded milk bottles (HDPE-MB) is one of the two most readily available postconsumer plastics, and the program would have been incomplete without it. However, HDPE-MB has one drawback for melt-processed composites--its relatively high melt viscosity (low MFI) can lead to some difficulties in melt processing operations. Despite the ready availability of PET from recycled beverage bottles, we did not include that material in the melt-processing portion of the program.

because PET melts at temperatures well above those at which cellulose begins rapid thermal degradation.

The primary restriction on the physical form of the plastic feed is that it be of a size (perhaps 1 to 3 mm) that it can be easily handled, easily dry-mixed with the reinforcing filler when that step is necessary, and easily fed into the melt-blending apparatus. Virgin plastics are readily available in the appropriate forms. Waste plastics, however, are often received in larger particles and must be ground. For that purpose, a granulator (Ball and Jewel BP-68-SCS, Minigranulator) was obtained and put into operation.

Cellulosics

Celluloses act as the reinforcing filler in the composite. In this program we employed wood flour (WF) as the primary baseline filler because it is currently used commercially in extruded WF/PP sheets for automobile interior panels. Wood flour currently sells for approximately \$0.22/kg. We included relatively expensive (several times the price of WF) pure cellulose fibers (BW40) as another baseline filler for comparing against recycled fibers. Waste newspaper (ONP) was employed as the primary recycled paper because of its ready availability in large quantities (approximately 6 percent of MSW) and in reproducible form at low cost (up to \$0.30/kg for the smallest sizes; considerably less for the larger fibers). More limited effort was devoted to old magazines (OMG) to determine whether that material, with its high content of clays and other materials, possessed advantages or disadvantages over ONP in composites. The OMG material is, of course, readily separable from MSW.

Preparation of Cellulosics for Laboratory Studies--

The cellulosic filler has restrictions similar to those for plastics. In addition, however, it must be in a form that can be completely dispersed into the molten plastic by the shear forces exerted during melt-blending. Wood flour was readily disaggregated and fully dispersed into individual particles in the plastic using a simple laboratory single-screw extruder. The BW40 cellulose fibers were more difficult to disperse with an extruder but a usable blend was obtained in that way. However, wastepaper fibers were much more difficult to work with because of their low bulk density. For melt-blending in a high intensity K-mixer (described in Task 2), the paper was simply milled or ground into flakes approximately 4 to 8 mm in size. The above-mentioned granulator proved to be excellent for this step. For melt-blending in an extruder, however, the paper must first be reduced back into fibers. For this purpose, we modified a small hammermill. Task 3, Study MB-2 presents results from a study that compared the properties of composites prepared by the two combinations: (a) grinding to flakes plus melt-blending in a K-mixer or (b) hammermilling to fibers plus melt-blending in a laboratory extruder. No major differences in properties were seen.

Preparation of Cellulosics for Commercial Use in Composites--

This program concentrated on laboratory investigations of composite preparation and characterization. Early on, however, it became obvious that some of the processes developed for laboratory investigations could not necessarily be translated directly into commercial practice. We deemed it essential, for example, to assess the commercial feasibility of methods for converting wastepaper into flakes or fiber and for melt-blending those forms of paper with plastic. Accordingly, we have carried on discussions and cooperative investigations with a variety of industrial firms--independently of this program for the most part. The major program in this connection was funded by the Wisconsin Department of Natural Resources (DNR) and by in-kind contributions of eight companies; this program will be referred to henceforth as the DNR program and will be summarized in Task 5. It included pilot plant and full plant tests with ONP that had been ground to flakes or milled to fiber. Discussions have also been held on conversion of ONP fiber to pellets in order to simplify handling and feeding to compounding equipment, on optimizing

grinding methods, and on obtaining ONP fiber as the fines from recycling pulp operations. We expect some of these options will be pursued in future cooperative programs.

Additives

Additives aid the dispersion of filler into the matrix plastic and/or enhance the bonding (act as a "coupling agent") between filler and matrix. Such materials can be very helpful to the processability and/or the mechanical properties of these composites by enhancing the compatibility of the highly nonpolar matrix (PP, HDPE) with the highly polar filler (cellulosics). In this program, we restricted ourselves to Eastman Epolenes. These are maleated polypropylene "waxes" in which the small degree of maleation provides polar groups capable of bonding to the cellulose while the polypropylene segments in theory offer compatibility with the polypropylene matrix. Earlier work demonstrated that they improved the mechanical properties of WF/PP composites (Olsen 1991, Myers and others 1991a,b). Although these additives are in the \$3.85/kg to \$4.95/kg range, they are used at low levels (usually around 1 to 2 percent of the total weight), can yield significant improvement in mechanical properties, and are not as expensive as other coupling agents such as silanes, zirconates, and titanates.

Incorporation of Coupling Agents

One method of incorporating dispersing aids or coupling agents is simply to add the dry powder to the mixture before blending; during subsequent melt-blending the additive must melt, become molecularly dispersed, and concentrate at the interface between solid cellulosic and plastic. Alternatively, it seemed probable that the additive would be more effective if it were placed at the cellulosic surface by precoating the solid before melt-blending. Accordingly, equipment was fabricated to permit uniformly coating the cellulosic filler by spray-coating the additives as solutions or emulsions. A third option was also examined in which the filler was coated by forming a slurry of the filler in the additive solution or emulsion and evaporating the solvent while stirring. An aqueous dye was used to optimize the spray-coating parameters for coating uniformity and to compare the uniformity of coatings by spraying or slurring. The latter procedure produced more uniform distribution of the dye. In either case, however, it is likely that low molecular weight additives on the cellulosic surface will be further spread on the surface or even partially removed from the surface during melt processing to prepare composites. An experiment was performed to compare the mechanical properties of ONP/PP composites made by direct addition of E43 powder to the dry mixture before melt-processing or by precoating the ONP using the slurry process. The results are presented in Task 3, Study MB-2.

TASK MB-2. LABORATORY METHODS FOR MAKING COMPOSITES

Compounding of Plastic and Cellulosic Filler in the Laboratory

Previous investigations demonstrated that wood flour can be readily melt-blended into polypropylene (MFI 3) using a small laboratory single-screw extruder (Figure 32) (Myers and others 1991a,b). Attempts to duplicate that melt processing step with ONP fibers and PP under this program were unsuccessful. The ONP fibers were too light and fluffy to feed properly into the extruder barrel, and homogenous blends could not be made. However, experiments reported from the University of Toronto indicated that melt-blending of ONP into PP could be accomplished with a laboratory (1-L) K-mixer, yielding highly promising properties (Woodhams and others 1991). The K-mixer (a high intensity mixer) is a cylindrical chamber containing several blades which rotate at tip speeds up to 40 m/s (Figure 33). The system being treated is heated by kinetic energy transfer from the blades until the plastic melts, at which time the filler material begins to disperse into the fluxing plastic. At a preset temperature, a bottom door opens, and the molten mix is discharged. Total mix times generally run from 30 to 120 s for wood fiber/plastic composites.

We carried out several blending experiments with the University of Toronto's K-mixer and were able to verify their findings. Microscopic examination of molded samples made from the K-mixer blends showed the blends to be more uniform than those from extrusion blending. Moreover, preliminary measurements of mechanical properties showed that the K-mixer blends of ONP and PP possessed better mechanical properties than those of WF in PP—as they should if the ONP fibers are well-dispersed.

As a consequence of these results, a 1-L K-mixer was ordered and placed into operation at the FPL shortly before the end of the first year of this program. During the interim, compounding with wastepaper fibers was carried out at the University of Toronto. Upon receipt of the K-mixer at the FPL, screening experiments were carried out to approximately optimize the operating parameters for blending wastepapers and several plastics, as a prerequisite to conducting the studies described in Task 3. The relevant parameters included chamber starting temperature, blade speed, mixing time, batch size, cooling water flow rate, and discharge temperature. As noted under Task 1, Task 3, Study MB-2 also describes a study that included comparisons of the properties of composites (non-ONP) prepared by compounding in the 1-L K-mixer or in the laboratory single-screw extruder.

Compounding of Composites for Commercial Applications

As in Task 1, the success in preparing ONP/plastic composites with the laboratory-sized K-mixer by no means meant that similar success would be met with that type of apparatus on a commercial scale or that the K-mixer was the best compounder for commercial applications of ONP/plastic composites. Consequently, our DNR program included tests of scale-up to a 40-L pilot plant K-mixer and of the ability of a pilot plant twin-screw extruder to compound ONP and PP (Task 5). In addition, we have held discussions on this aspect with a variety of compounding equipment manufacturers and expect to carry out a cooperative program to compare several types of equipment.

TASK MB-3. DEVELOPMENT OF A PERFORMANCE DATABASE

Under this task, we carried out several studies which were essentially independent of one another. In some cases, the same parameter was examined in the context of different studies; this duplication allowed us to optimize the significance of conclusions within each study. We describe each study separately in the following.

In all studies, test specimens were prepared by injection molding using a Froehring Minijector model SP50. At least five specimens of each blend were tested for each mechanical property. After molding, the specimens were stored over desiccant for at least 3 days before testing. Mechanical properties were measured on the dry specimens at approximately 23°C. Specimens and test methods followed ASTM specifications (ASTM 1984a–c, 1990). Strain rates were 5 mm/min for tensile and 1.5 mm/min for flexure tests. Table 12 summarizes the mean standard deviations observed in the program for various mechanical properties.

Study MB-1. Effects of PP Molecular Weight and ONP/PP Ratio

Background Information--

Study MB-1 was performed under this program, but it eventually also constituted a major portion of Phase I in the DNR program. Results have been published in a somewhat abbreviated form (Myers and others 1992).

A full-factorial experimental design was used to investigate the mechanical and rheological properties of ONP/PP composites over a range of PP molecular weights and ONP/PP ratios that represented the levels we felt to be of primary commercial interest. The molecular weight of plastics strongly influences melt viscosity, hence melt processability, of both the unfilled and filled plastic. Molecular weight also strongly affects mechanical properties of plastics, and presumably composites, at low molecular weights. At a moderate molecular weight, however, the dependence of mechanical properties on molecular weight essentially disappears; that asymptote was not known for our composite systems.

Experimental Details--

We selected the following matrices, in which melt flow index (MFI) is an inverse measure of melt viscosity and hence represents an approximate inverse measure of molecular weight.

Matrix A

PP MFI	3, 12, or 30 g/10 min
ONP/PP ratio	32, 37, 42 by weight

Matrix B

Filler type	WF or ONP (42 percent by weight)
PP MFI	3, 12, or 30 g/10 min

All formulations contained E43 at 5 weight percent of filler. The entire matrix was replicated and all trials performed in random order. All compounding was carried out in a 1-L K-mixer at the University of Toronto. The ONP was added to the K-mixer as flakes and fiberized by mixing with the E43 at 3,300 rpm for approximately 90 s, after which the plastic was added and mixing continued for approximately 60 s until the mix temperature reached 185°C. For each trial, approximately twenty 0.25-kg batches were prepared; the batches were hammermilled, dry-blended, and stored over desiccant until molded.

Melt viscosities were measured on selected blends of the matrix at several shear rates. Apparent viscosities and apparent shear rates were calculated from measured volumetric throughput rates and pressure drops across the die during steady state extrusion in a Brabender Plasticorder single-screw extruder at 190°C. (Absolute values of viscosity and shear rate would have necessitated making several corrections to the data. It is common practice to report only apparent values when the interest is in the relative changes brought on by the variables being examined.)

Results and Discussion--

Table 13 summarizes the mechanical properties, and Table 14 summarizes the results of the statistical analysis. Table 15 summarizes the larger property changes, and Figures 34 and 35 illustrate some effects. The following points are noteworthy:

- No interaction terms were observed in the statistical analysis (Table 14).
- All effects of PP MFI were negligible from a practical viewpoint—they were either not

statistically significant at the 95 percent confidence level or amounted to less than a 10-percent change (Tables 14 and 15).

- The greatest effects of ONP/PP ratio were on modulus, but only amounted to a 12-percent increase at best (Table 15).
- The greatest changes resulted from substituting ONP for WF (Table 15). Flexural strength increased 24 percent (Figure 34), tensile strength 29 percent, and unnotched impact 43 percent (Figure 35).

Apparent melt viscosities are shown in Figure 36. Table 16 summarizes the values obtained by interpolating or extrapolating the linear log-log plots to 100, 500, and 1000 s^{-1} shear rates; the last value approaches the shear rates existing during commercial extrusion. Both filled and unfilled systems were strongly non-Newtonian (shear rate-dependent). At 1000 s^{-1} shear rate, the viscosity of the 42/58 ONP/12 MFI PP was approximately 25 percent above that of the similar 32/68 system and nearly twice that of the 42/58 WF/PP system. Thus, the 42/58 ONP/12 MFI PP system would use more energy during extrusion, and it might necessitate higher pressures or larger diameter gates during injection molding. Clearly, however, the greater viscosity of the ONP systems compared with the WF systems could be obviated by substituting a higher MFI PP without large effects on composite performance.

Conclusions--

- Within the range of variables examined, only the replacement of WF by ONP brought about large changes in mechanical properties. Those improvements were sufficiently large to validate further study of ONP as a reinforcing filler—from both research and commercial points of view.
- With regard to investigations into commercial feasibility of ONP/PP composites, it appeared that a good balance of mechanical properties and processability could be achieved by using a 12-MFI PP at a 42/58 weight ratio of ONP and PP. That is, viscosity could be tailored to a specific manufacturing process without detriment to composite performance.

Study MB-2. Effects of Filler and Plastic Type, Mode of E43 Addition, and Compounder

Background Information--

It was important to clarify the influence of several additional variables on composite mechanical properties, addressing the following questions:

- (a) Do ONP fibers differ from pure cellulose fibers in their reinforcing effects? (WF was included as the customary baseline filler.)
- (b) The PPV currently used as the matrix in commercial WF/PP composites is stronger, stiffer, and less viscous than HDPE-MB but also has lower impact energy. How do those differences translate into composite properties?

(c) Coupling agents may be incorporated into composites in different ways to concentrate the material at the interface where it is active. Does the addition of solid powdered E43 in its anhydride form produce differences in mechanical properties from precoating fiber with the emulsified potassium form of E43?

(d) Fibrous fillers are more effective reinforcing agents if their length is above a so-called critical value and if they are fully disaggregated into individual fibers. The high shear forces existing in the K-mixer should produce good fiber dispersion but probably will simultaneously reduce the fiber length. In contrast, a single-screw extruder will probably be less efficient in dispersing the fibers but less likely to reduce the fiber length. What is the net effect of these influences on mechanical properties?

Portions of this study were published (Gonzales and others 1992).

Experimental Details--

Because ONP could not be dispersed into the plastics using a single-screw extruder, it was necessary to divide this study into two separate experiments. Experiments A and B were both three variable, two-level full-factorial matrices that were replicated and carried out in random order.

Experiment A

Polymer	PPV (MFI 12) or HDPE-MB
Filler	ONP or BW40 cellulose
Form of E43	Powdered anhydride or anionic emulsion

The filler/polymer ratio was kept constant at 40/60 by weight. E43 was added at 5 percent of filler by weight. Powdered anhydride was incorporated during melt-blending. Anionic emulsion was precoated on filler by evaporating a slurry. Compounding was done with the 1-L K-mixer.

Experiment B

Polymer	PPV (MFI 12) or HDPE-MB
Filler	WF or BW40
Compounder	K-mixer or extruder

The filler/polymer ratio was the same as in Experiment A. E43 was added at 5 percent of filler by weight, incorporated as the solid anhydride during melt-blending. Extruder was a 38-mm Modern Plastics Machinery single-screw extruder.

For K-mixing, the fillers were mixed for approximately 1 min, polymer was added, and mixing was continued for approximately another min. A 185°C discharge temperature and a speed of 2800 rpm were used for the HDPE systems; for PPV the conditions were 200°C and 3000 rpm. Extrusion was carried out at 170, 180, and 190°C for the three barrel sections and 190°C for the die and at a screw speed of 15 rpm. Other details were as in Study MB-1.

Results and Discussion--

Table 17 summarizes the mechanical property data. Tables 18 and 19 present the main effects and two-way interactions found at the 95 percent confidence level. Figures 37 to 41 illustrate some of the observed effects by combining the results of Experiments A and B, and Table 20 summarizes the "useful" percentage changes as a result of replacing HDPE-MB with PP, WF with BW40, and BW40 with ONP. We note the following:

- A small preference existed for the solid form of E43, although the effects are relatively small and without practical significance (Table 18). It appears that concentrating the emulsified additive at

the fiber/matrix interface by precoating the fibers was counteracted by the much lower chemical reactivity of the potassium salt with the cellulose compared with the reactivity of the anhydride with cellulose. Perhaps the E43 functions more to enhance dispersion of the cellulosic fillers than to bond the cellulose to the polymer.

- Little preference was shown for either compounding method with WF or BW40 (Table 19). However, those fillers are relatively easily dispersed, and we reiterate that the ONP could not be melt-blended with the extruder employed here. Moreover, the small interaction terms between filler and compounder also indicated some differences in dispersibility between WF and BW40.
- In general, the fibrous fillers provided greater reinforcement (greater strength and modulus) (Figures 37 to 39) to either matrix than did the wood flour. Moreover, the inexpensive ONP was, for the most part, at least as beneficial as the cellulose fiber (BW40). Despite the fiber breakage during melt processing, therefore, the fibrous fillers appear to have retained a greater aspect ratio than did the wood flour.
- The PPV composites consistently had greater strength and modulus than did the HDPE-MB composites (Figures 37 to 39, Table 20). This difference reflects the differences in the matrix polymers themselves and not the fact that the HDPE-MB is recycled (cf. Task 4).
- Impact behavior was less straightforward in its reaction to variable changes. The most meaningful change probably was the loss in notched impact upon substituting PP for HDPE-MB (Figure 40); this too is consistent with the behavior of the matrices themselves.

Table 21 compares the properties of the system using recycled ingredients (HDPE-MB and ONP) with those of the current commercial system (PPV and WF). Notched impact energy for the "recycled" system was greatly improved whereas all other properties were identical within experimental error. Based on these mechanical properties, the recycled HDPE-MB/ONP system could therefore advantageously substitute for the PPV/WF system. However, this mechanical property advantage may be offset in some commercial melt processes by the greater melt viscosity of the HDPE-MB system.

Conclusions--

- Neither E43 form nor compounding method produced meaningful changes on composite properties.
- Fibrous cellulose or ONP caused greater reinforcement of PP and HDPE-MB than does WF. ONP was essentially equivalent to cellulose as a reinforcing filler.
- PP composites were stronger and stiffer than HDPE-MB composites but possessed lower notched impact. Composite properties qualitatively followed those of the matrix polymers.
- A recycled ONP/HDPE-MB system had at least as good mechanical properties as the current commercial WF/PP system.

Study MB-3. Coupling Agent Comparison

Background Information--

Coupling agents are often used in composites to enhance the bonding between matrix and filler, thereby increasing the stress transfer between the two phases and improving strength and stiffness. As noted, previous studies demonstrated that Eastman Chemical's Epolene E43 and G3002 enhanced some properties of WF/PP composites (Olsen 1991; Myers and others 1991a,b), the G3002 being particularly effective. Consequently, it was important to establish the relative effects of these two Epolenes on the mechanical properties of ONP/PP composites.

These materials are relatively low molecular weight polypropylenes onto which small amounts of maleic anhydride have been grafted (Table 22). They differ in molecular weight and amount of grafted maleic anhydride per molecule; however, the frequency of anhydride groups along the Epolene molecular chain (number of polymer repeat units per anhydride) was equal.

Portions of this study are in the process of publication (Sanadi and others, in press).

Experimental Details--

The ONP/PP composites were prepared at a 40/60 weight ratio containing no additive or containing 3 percent by weight of E43 or G3002. Blending was performed with the K-mixer at 4600 rpm for 2 min to complete fiberization and then at 5500 rpm until the discharge temperature of 200°C was reached. Batch sizes were 150 g and the matrix was replicated.

Results and Discussion--

It is important to note that this study was performed as a preliminary examination of the effectiveness of these additives. Therefore, no statistical analysis of the results is presented. The preliminary results are highly encouraging, however, and additional testing is planned.

Table 23 summarizes the mechanical properties. Strength values increased regularly from no additive to E43 to G3002, as illustrated in Figures 42 and 43 where tensile strength increased 50 percent with G3002. Increase in tensile strength was accompanied by greater energy absorption (100 percent with G3002) and by a 50 percent increase in elongation with G3002. Figure 42 shows the tensile stress strain curves for the three systems and graphically illustrates the positive effects of G3002. The greater energy absorption during tensile failure was paralleled by increases in unnotched impact energy (70 percent with G3002) (Figure 44). Surprisingly, the additives, if anything, decreased the moduli.

Conclusions--

The higher molecular weight maleated polypropylene, G3002, was very beneficial to composite strength, energy absorption, and unnotched impact behavior and warranted further investigation. These increases indicate that G3002 is not acting as a strong coupling agent but instead is probably enhancing dispersion of the fibers and reducing aggregates which would act as failure loci. Unnotched impact, for example, is more affected by removal of failure loci than is notched impact because the notch already is an overwhelming failure locus. The apparently greater interaction of G3002 with the PP relative to E43 may be attributable to the higher molecular weight of G3002.

Study MB-4. Effects of Wastepapers and Waste Polypropylenes

Background--

In this study, we extended the database on waste plastic and wastepaper to define the potential of some waste polypropylenes combined with old newspaper or old magazines. The rationale for selecting the particular materials was discussed in Task 1. Most of this study has been published (Clemons and Myers 1993).

Experimental Details--

The following full-factorial matrix was investigated in replicate. Data for the comparable ONP/PPV systems were obtained from other studies. Comparisons were also made with each of the unfilled polymers.

Polymer	KPP or BPP
Filler	ONP or OMG
Filler/polymer ratio	40/60 by weight
Epolene G3002	3 weight percent of filler
Compounding	1-L K-mixer

The ONP and OMG were fed to the K-mixer as approximately 5-mm flakes. Blending was carried out for approximately 1.5 min at 4,500 rpm, with a discharge temperature of 170°C to 190°C. Fiberizing the OMG in the K-mixer was more difficult than fiberizing ONP, resulting in poorer dispersion of the OMG in the matrix plastic.

Results and Discussion--

Table 24 summarizes the mechanical properties, and Table 25 gives the results of the statistical analysis of the two-variable, two-level factorial. No interactions were found at the 95 percent confidence level. Unnotched impact energy showed no statistically significant change, probably because of the relatively large data scatter.

Table 26 shows the property changes brought about when 40 percent filler was added to each polymer. Some property changes are illustrated in Figures 45 to 49. Both tensile and flexure moduli improved more than 100 percent for KPP and more than 150 percent for BPP (Figure 47). Modest improvements in strength properties were observed (~30 to 75 percent), but BPP was again reinforced to a greater extent (Figure 46). Energy to maximum load and elongation at maximum load decreased, and the related impact energy dropped sharply (e.g., ~50 percent reduction in unnotched impact energy for KPP and 80 percent for BPP) (Figures 48 and 49). Thus, incorporating wastepaper fibers led to stiffer and stronger but more brittle materials compared to the unfilled plastics.

All tensile and flexure properties improved when OMG was replaced with ONP (Tables 24 and 25, Figures 46 and 47); the largest increases were 50 percent in tensile energy absorbed and 34 percent in tensile strength. The greater reinforcement produced by ONP is consistent with its better fiberization and dispersion in the the K-mixer. In addition, the large amounts of clay in OMG may have diluted the total reinforcing efficiency of the OMG.

When BPP was used instead of KPP, all properties except notched impact energy were decreased by only small amounts (Table 25). The observed changes qualitatively followed the properties of the matrices. The softer, weaker, and tougher BPP was more sensitive than was KPP to filler addition, but overall BPP did not achieve as good a balance of properties as did KPP (Table 24). In particular, the superior impact energy of the unfilled BPP (an ethylene-propylene copolymer) was poorly transferred to the composite system. Nevertheless, both the KPP and BPP composites had notched impacts significantly greater than that of the VPP composite (Figure 48). Thus, BPP and KPP in certain composite applications could provide attractive cost and performance benefits compared to the virgin PP.

Conclusions--

- Adding 40 percent fiber to recycled plastics leads to stiffer, stronger, but more brittle materials. The composite properties qualitatively followed those of the matrix plastics.

- OMG was more difficult to disperse than ONP and produced a balance of properties that was a little less desirable than the balance produced by ONP.
- Because ONP is more available, is more easily processed and produces better composite properties, it offers more promise for commercial applications than does OMG.
- Because BPP is less expensive, is more available, and produces a good balance of properties, it should be of more interest than KPP and a useful replacement for virgin PP in some applications.

FURTHER CHARACTERIZATION OF MELT-BLENDED COMPOSITES

Background

Work has been started to further characterize melt-blended composites. Soil-block tests are being conducted which will determine the susceptibility of the composites to fungal attack. Tests have also been initiated on composite fire resistance and the effects of water absorption on dimensional stability and mechanical properties. Only the water absorption experiments are sufficiently advanced to report here.

Because of the hydrophilic nature of wood and paper fiber, absorption of water could adversely affect the performance of these composites. The following summarizes a preliminary study investigating these water sorption effects.

Experimental Details

The following three composite systems were investigated at a 42/58 weight ratio of filler to PP. Systems 1 and 2 evaluate the effect of water exposure on ONP/PP composites and the influence of G3002 coupling agent thereon. Systems 2 and 3 compare ONP and WF composites.

System 1. ONP/PP with 3 percent G3002 (by weight of the fiber)

System 2. ONP/PP without coupling agent

System 3. WF/PP without coupling agent

Two different exposure tests were performed:

(1) 24-h submergence in water at ambient temperature. This was performed because it conforms to an ASTM standard for plastics.

(2) 24-h submergence in boiling water. This was performed because early screening tests suggested a more severe exposure than the above was needed to observe large changes in a short time.

Cantilever and impact properties, weight, and thicknesses of composite specimens were measured before (dry) and after (wet) the exposures. Weights and thicknesses were measured on the cantilever specimens. Surface moisture was removed before any measurements were made.

Results and Discussion

Tables 27 to 29 summarize the physical and mechanical properties before and after the water exposures. Table 30 is a summary of the statistics for the study. The following points are noteworthy:

- Little change in physical and mechanical properties was found after the 24-h water soak. Even at this relatively high level of filler (42 percent), the encapsulation of the fibers by the polypropylene matrix inhibits absorption of water.
- Comparing the property values before and after the 24-h water boil (last column of Tables 27 through 29), it is clear that the largest change occurred in cantilever modulus; it is the only property that decreases by more than 10 percent after the exposure to boiling water. This decrease in performance may be attributable to the decrease in modulus of the wood/paper fibers themselves when exposed to moisture (Morton and Hearle 1962). The sharp rise in unnotched impact in System 2 (Table 28) was inconsistent with the other two systems and was probably an anomaly.
- Property decreases were probably independent of filler type and the presence of G3002. The latter observation is consistent with other findings that indicate these maleated polypropylenes are not strongly bonded to the filler.

Conclusions

- Additional testing is desirable to establish whether the moduli in other test modes are also sensitive to moisture and to clarify the conclusion that impact is not altered by the water boil. However, it seems likely that routine testing of sensitivity to moisture will not be necessary in the future.
- If the modulus reduction is shown to be a practical problem, additional investigation of coupling agents that actually bond to the wood fiber may be desirable.

TASK MB-4. RECYCLABILITY

Background

In Study MB-5, we investigated the effects of reprocessing on the mechanical and rheological performance of three different composite blends. Although much work on the use of recycled ingredients in composites has been published, very little work has been done to determine the recyclability of the composites themselves. This is not to say that composites are not routinely recycled as part of industrial processes, but few studies have been published which quantify the effects of continued recycling on the mechanical properties of the composite. One notable exception is work performed at the University of Toronto where composites made from virgin PP or HDPE and telephone directories were put through several cycles of K-mixing and granulation (Schmidt and others 1992). Small changes in some mechanical properties and large changes in melt flow index were observed. However, recycling by repeated K-mixing is not a realistic scenario, and we selected a cycle involving repeated extrusion instead.

Experimental Details

Three composite formulations were investigated:

- System 1 Virgin PP/40 percent ONP (no coupling agent)
- System 2 HDPE-MB/40 percent WF (no coupling agent)
- System 3 HDPE-MB/40 percent air-laid composite (air-laid composite contains 80 percent fiberized demolition wood/10 percent phenolic resin/10 percent PET)

The first two systems are representative of melt-blended systems investigated in other parts of the research program. The filler for System 3 was an air-laid composite panel selected to determine the feasibility of recycling air-laid composites containing both a thermosetting resin and the high melting PET by employing the air-laid composite as a filler in a melt-processed composite. This feasibility was in question because neither the phenolic resin nor the PET would melt at the temperatures suitable for melt processing a wood-containing composite but would retain their particulate form; it was uncertain, therefore, what effect those ingredients would have on either the processing or the mechanical properties of the resultant composite. To use the air-laid panel as a filler, it was ground to -35 mesh and added to HDPE-MB at a level of 40 percent by weight.

Figure 50 is a processing flow chart for the study. Each system was initially melt-blended in a K-mixer and then granulated. The granulated material was used as the initial feedstock for the study. The material was then extruded and granulated five times, each extrusion and granulation constituting a cycle. Initially, and after each cycle, enough material was removed for mechanical/rheological measurement. Additional stabilizers were incorporated after the second and fourth cycles to minimize polymer degradation.

Each system was initially blended in the K-mixer at 5500 rpm for approximately 30 s with a discharge temperature of 194°C. Extrusion was carried out in a Killion 25.4-mm single-screw extruder at 35 rpm with a temperature profile of 192°C and 199°C in the barrel and a die temperature of 204°C.

Mechanical properties of injection-molded specimens were measured initially and after each cycle. The MFI was measured for System 1 only. MFI was not measured on the HDPE systems because the high viscosity of HDPE-MB causes such low MFI values that the variability made the measured values meaningless. Fiber dimensions of ONP and WF (Systems 1 and 2) were measured by image analysis. Fibers from initial blends and after five cycles were measured. Results are summarized as follows:

Fiber	Length (microns)	Width (microns)	Aspect ratio
ONP (initial)	225	19	11.9
ONP (5 cycles)	138	18	7.4
WF (initial)	162	88	2.0
WF (final)	111	57	2.0

Results and Discussion

Tables 31 to 33 summarize the data and statistics for each system, and Table 34 is a summary of the analysis of variance. We note the following.

- For almost all of the mechanical and rheological properties for all three systems, very little change was found as the material was recycled. However, several small, statistically significant, changes in mechanical properties resulted from the first cycle. This was probably the result of further mixing in the extruder and enhanced fiber dispersion as well as some minor fiber attrition. The cantilever strength and modulus for System 1 dropped significantly during the fifth cycle. These changes are not reflected in the tensile properties, however, and are not very large from a practical point of view.
- Mechanical properties for Systems 2 and 3 were roughly equal, showing that the air-laid filler performed as well as wood flour. This fact demonstrates the potential for recycling air-laid composites in this manner if the issues of cost, quality, and quantity could be met.
- Over the course of five cycles, the length and, therefore, the aspect ratio of the ONP fiber decreased. These decreases, however, were apparently not large enough to result in any large reductions in composite performance. WF was reduced both in thickness and length, as smaller bundles of wood fibers were sheared off the larger particles. These changes in dimension resulted in no overall change in aspect ratio and, ultimately, composite performance.

Conclusion

We conclude from these results that there are no serious drawbacks to recycling these representative melt-blended and air-laid composite systems through several melt-processing cycles.

SECTION 6

PRODUCT APPLICATION AND DATABASE EXTENSION (TASK 5)

Efforts on Task 5 were not always easily divided into melt-blended or air-laid technologies. Therefore, we present Task 5 as a separate section.

COMMERCIAL IMPLEMENTATION

The EPA program at the FPL provided opportunities for developing additional experience in the theory and practice of wood fiber-plastic composites and for developing industry and university contacts in this area. As a consequence, the program was instrumental in leading to numerous cooperative studies with industry and academia, all of which had as their ultimate goal the commercialization of composites containing ingredients from the waste stream. In addition, the FPL was able to provide advice and information on these systems to a large number of companies and government agencies. In the following, we summarize some of the cooperative studies that arose out of the EPA program.

Commercial Feasibility of Waste Newspaper-Thermoplastic Composites

Laboratory experiments demonstrated that ONP could be dispersed as fibers into thermoplastics by melt-blending, resulting in substantial improvements in some properties compared with the unfilled plastic or compared with the plastic filled with wood flour (Recommendations, Tasks 2 and 3). The FPL and the University of Wisconsin Industry Research Office held several meetings with a variety of industrial firms to plan a cooperative study that would demonstrate whether the laboratory success could be scaled up to commercial operations to produce commercial products. The program was partially funded by the FPL and the Wisconsin Department of Natural Resources (DNR) and by in-kind contributions from the eight cooperating companies. The program and its accomplishments are described in the accompanying final report from FPL to DNR (Myers and Clemons 1993) and in the accompanying manuscript that has been submitted to *Plastics Engineering* (Clemons and others, submitted). We summarize the major conclusions here:

- (1) ONP/PP composites can be compounded on a commercial scale using either the K-mixer with ONP flakes as feed or using a twin-screw extruder with ONP fibers fed separately from the plastic.
- (2) ONP/PP sheet containing 42 weight percent ONP can be prepared by extrusion on a commercial scale. This sheet meets existing specifications for automobile panels and can be thermoformed into a variety of shapes.
- (3) Given proper design of melt processing equipment, a wide variety of other commercial products could be manufactured from ONP/PP composites with similar ONP content.
- (4) Firm estimates of production costs for ONP/PP composite products must await
 - (a) additional examination of compounding methods to define the optimum balance of dispersion ability, throughput rate, and cost

(b) improvement in methods to deliver wastepaper in a form and at a cost acceptable to a compounder or a manufacturer of plastic products, keeping in mind that wood flour is available in quantity at approximately \$0.22/kg

Waste LDPE Program

A consortium of companies have recently come together to investigate the use of waste LDPE that is "contaminated" by residual fiber from a hydropulping operation that scavenges wood fiber from coated paper stock. The program consists of raw material processors, compounders, plastics processors, and research institutions and is being coordinated by the FPL. A research program has been finalized and work is just beginning. Major hurdles in this program are the residual moisture from the hydropulping process left in the raw material and product applications.

Waste Jute-Polyester Panels As Reinforcing Filler

A U.S. company was interested in the possibility of recycling panels that they produced by impregnating jute fibers with thermosetting polyester. We granulated the panels and investigated the ability to use the resultant mixture as reinforcing filler in melt-blended composites with a polypropylene matrix. Overall, it appeared that this waste material produced composite mechanical properties that were approximately equivalent to those of similar composites containing wood flour as the reinforcement (Schneider and others, submitted).

Waste Kenaf Core As Reinforcing Filler

The Agrecol Corporation of Madison, Wisconsin, had a grant from the Wisconsin Department of Agriculture to investigate the potential of kenaf as a commercial agricultural product in Wisconsin. Kenaf is a very rapidly growing plant whose outer fibers produce high quality paper. However, the kenaf core material is much less fibrous than the outer fibers and of less interest for paper manufacture. Agrecol requested FPL to determine whether the kenaf core material could be useful as a reinforcing filler in plastic composites. We granulated the core material and successfully melt-blended the -40 mesh fraction with polypropylene. The composite properties were also approximately equivalent to those of similar composites containing wood flour. Therefore, where the kenaf core may be readily available at low cost, it very likely could substitute for wood flour as a reinforcing filler (Schneider and others, submitted).

Wastewood Composite As Reinforcing Filler

The University of Tennessee extension requested an evaluation of wastewood composite as a reinforcing filler in thermoplastic composites. Such solid waste is available in large quantity; it contained plywood, particleboard, and fiberboard and several percent of cured thermoset adhesives which might cause problems in melt-processed composites because it would not melt at processing temperatures. We granulated the plywood and successfully melt-blended the -40 mesh fraction with polypropylene. The composite properties were approximately equivalent to those of similar composites containing wood flour. This waste material could therefore substitute for wood flour as a reinforcing filler in melt-processed composites (Schneider and others, submitted).

CONFERENCES, PRESENTATIONS, AND PUBLICATIONS

We have actively pursued efforts to bring wood fiber-plastic composites and their potential for commercial applications to the attention of both the research community and industry. This has been accomplished by sponsoring conferences; by presentations at conferences, workshops, and technical meetings; and by publishing papers in scientific and semitechnical journals.

Wood Fiber-Plastic Composites Conferences—Two international conferences were co-sponsored with the University of Wisconsin and held in Madison, in April 1991 and May 1993. Each was attended by approximately 200 people from the United States, Canada, and several other countries, representing academia, industry, and public agencies. We presented several papers and posters at each conference on results that resulted directly from this EPA program and on other composites studies. Both conferences were highly productive in terms of developing interest in these systems and industrial contacts. The University of Toronto held two similar, but smaller, conferences, in February 1991 and May 1992. We presented two papers at each conference.

American Chemical Society Meeting—A paper entitled "Lignocellulosic/Plastic Composites from Recycled Materials" was presented at the spring 1991 ACS national meeting. It was subsequently published in *Emerging Technologies for Materials and Chemicals from Biomass: Proceedings of Symposium*; 1990 August 26-31; Washington, DC: American Chemical Society; 1992. Chap. 4. ACS symposium series 476.

Focus 95+ Landmark Recycling Symposium—A paper entitled "Alternative Uses of Recovered Fibers" was presented on March 21, 1991 at a symposium sponsored by primary paper manufacturers, technical and management associations, academia, and government.

International Particleboard/Composite Materials Symposium—A paper entitled "Composites From Recycled Raw Materials" was presented at this annual symposium on April 11, 1991.

Annual Technical Meeting of the Society of Plastics Engineers (ANTEC)—Papers were presented at two meetings, in 1992 and 1993. These were published in the meeting proceedings (Myers and others 1992, Clemons and Myers 1993).

Materials Research Society Meeting—A paper was presented at the national meeting in 1992 (Gonzales and others 1992).

Other Papers Submitted to Journals—Several additional papers have been submitted to journals and should be published in 1993 or early in 1994 (Sanadi and others; Clemons and others; Schneider and others). The Task 4 studies on Recyclability will also be submitted to a scientific journal in 1993.

Reports to Cooperators—Several reports have been submitted to industry and public agency cooperators, including the final report for the DNR program on commercial feasibility (Myers and Clemons 1993).

SECTION 7

QUALITY ASSURANCE OBJECTIVES

A Quality Assurance Project Plan for this program was submitted to the EPA in June 1990. The Plan identified the following areas to which quality assurance objectives were primarily applicable:

- (1) Preparation of ingredients for incorporation into composites
- (2) Preparation of composites
- (3) Characterization and testing of composites

With regard to the preparation of ingredients and composites (areas 1 and 2), our work emphasized innovative experimentation and trial and error runs to improve or develop methods to the point where reproducible, but not necessarily optimized, processes could be used. Process parameters were apparatus-specific and were controlled as necessary to provide reproducible material.

The emphasis throughout the program was on product quality, and therefore area 3 was of primary concern. In this case, the situation was much different from that in areas 1 and 2. The criteria, methods, and calculation procedures are mostly well-documented in standards maintained by the American Society for Testing and Materials (ASTM) or by the Technical Association of the Pulp and Paper Industry (TAPPI). Specifications for experimental precision were based on our own laboratory experience, from our in-house statisticians, and as specified by ASTM or TAPPI standards when those were available. Wherever possible, experiments were statistically designed and analyzed, using standard statistical methods.

We summarize below the experimental methods that were actually employed in the program and their precision, following the criteria outlined in the preceding comments.

PREPARATION OF INGREDIENTS FOR INCORPORATION INTO COMPOSITES

Air-Laid Composites

During air-forming, it is essential that all the materials introduced into the air stream remain intimately mixed during the web formation process. This requirement necessitates that materials of similar configuration be used, or that materials, like powders, are introduced into the air stream just prior to web formation to insure that they remain evenly distributed in the formed web. Alternatively, powders can be used if a tackifier is added to either the wood or plastic fibers so that the powder adheres to the fiber during the air-forming operation. The primary criterion for acceptability of the raw materials is that they be capable of being distributed evenly and uniformly during the web formation process.

Hemlock fiber (HF), virgin and recycled polyethylene terephthalate (VPET, RPET), phenolic resin (PR), and tackifier (E-10) were used as received. The following materials required further processing to make them suitable for use in our processing equipment:

Demolition wood fiber (DF)—received in a chipped form and had to be converted into a fiber or fiber bundle format using refining methodology.

Virgin high density polyethylene (VHDPE)—required grinding to a nominal (-)35 mesh size.

Recycled high density polyethylene (RHDPE)—obtained from old milk bottles that were chipped and then ground to a (-)35 mesh size.

Melt-Blended Composites

During melt-blending, the plastic melts and completely loses its original solid geometry. The high shear and extensional forces within the molten plastic cause the reinforcing fiber to be dispersed into the plastic, but they simultaneously break the fiber into shorter lengths. Because of these changes in the starting ingredients, their original form and size are not highly critical. The primary criterion for acceptability of the starting ingredients is that they be relatively easy to handle and to feed into the melt-blending apparatus.

Virgin plastics were obtained in pellet or powder form and were not further treated prior to melt-blending. Waste plastics were obtained as flakes or particles and were granulated to approximately 1 to 3 mm using a Ball and Jewel BP-68-SCS Minigranulator equipped with a 3-mm screen. Granulation times were kept to a minimum to prevent heating and melting of the plastics.

Wood flour was obtained as -40+80 mesh material. Wastepapers were treated in two ways depending on the method used to perform subsequent melt-blending. For blending in the K-mixer, the wastepapers were passed rapidly through the Ball and Jewel granulator equipped with a 6-mm screen to produce flakes approximately 4 to 8 mm in size. For melt-blending with an extruder, the wastepaper was hammermilled in a small hammermill with a 3-mm screen.

PREPARATION OF COMPOSITES

Air-Laid Composites

A uniform air-formed web profile across and along the air-forming machine must be obtained to insure that the pressed composite panel has uniform properties throughout. Extensive efforts were made to optimize our air-forming process to insure that web uniformity was obtained to the maximum extent possible. Uniformity can also be achieved from panel to panel or from test specimen to test specimen by close control over the criteria for acceptability of the composites to be tested. Accordingly, panels for AL Series 1 were selected to ensure that they were within the specific gravity range of 1.0 ± 0.03 . Target thicknesses were maintained at 3.2 mm.

In a further effort to maintain close specific gravity control over the actual test specimens, test specimens for AL Series 2 and AL Task 4 were cut from test panels and the specimen specific gravities were determined. In this series, only test specimens with an acceptable range of 1.0 ± 0.06 were used for testing purposes. Again, target thicknesses were maintained at 3.2 mm.

All of the panels made in this program were fabricated using a pressing temperature of 190°C for 4 min at a maximum pressure of 8.47 MPa. Three minutes of cooling time was necessary during compaction to reduce steam vapor pressure in the pressed board and to maintain the target thickness of 3.2 mm.

Melt-Blended Composites

An ideal melt-blending process would produce a composite in which the reinforcing filler is completely dispersed into individual particles without any damage to either the filler or the plastic arising from the temperature or the stresses within the melt. Unfortunately, complete dispersion and lack of damage are likely to be contradictory achievements. Therefore, in practice one must usually compromise and carry out melt-blending at the lowest temperature, the shortest times, and the lowest shear rates that will produce a degree of dispersion that yields composites with reproducible properties. These conditions differ for different plastics and fillers, for different filler concentrations in the plastics, and for different equipment. In every case, approximately optimized conditions were determined by screening tests. The ranges in blending parameters used in this program were as follows:

Melt-blending by extrusion

Modern Plastic Machinery extruder
Barrel temperature, 170°C to 190°C
Screw speed, 15 rpm
Residence time, 1 to 2 min

Killion extruder
Barrel temperature, 192°C to 204°C
Screw speed, 34 rpm
Residence time, 1 to 2 min

Melt-blending by K-mixing

University of Toronto K-mixer
Speed, 2800 to 3300 rpm
Discharge temperature, 185°C
Residence time, 30 to 300 s

FPL K-mixer
Speed, 4500 to 5500 rpm
Discharge temperature, 171°C to 200°C
Residence time, 30 to 150 s

CHARACTERIZATION AND TESTING OF COMPOSITES

Air-Laid Composites

All tests were done on the INSTRON test machine USPN 8176. Calibration was done at the beginning of each new test and checked throughout the test. The INSTRON has an electric load cell to measure forces of 444.8 N or less, and is calibrated by National Bureau of Standards (NBS) dead weights. It is calibrated by Morehouse rings with NBS-traceable calibration data when more than 444.8 N. Displacement is measured by linear variable displacement transducers (LVDTs) calibrated with gauge blocks. Total system calibration is within 1 percent of full scale.

All testing of the air-laid composite panels in this program were conducted using the test procedures specified in detail in the referenced Quality Assurance Project Plan. The observed values obtained on all of the test specimens for each of the AL Series were consistent with data that we have obtained in other test programs using virgin raw material resources. Our results

demonstrated that both our laboratory techniques and our testing procedures were uniform and consistent.

Melt-Blended Composites

To determine mechanical and physical properties of melt-blended composites, specimens were prepared by injection molding using a Frohring Minijector model SP50. Control parameters for this machine were barrel temperature, residence time, and injection pressure. Here also, compromises must be adopted. High temperature causes polymer degradation, whereas low temperature, particularly when combined with low pressure, makes it difficult to fill the mold cavity and produce a complete specimen. Again, conditions may differ with different systems and formulations. However, exact control of parameters is not necessary. Typical conditions employed in the program were 205°C barrel temperature, approximate residence time of 1 min, and 3.4 to 8.9 MPa injection pressure.

A minimum of five specimens was tested for each type of measurement at each variable being investigated. Except for the moisture sensitivity tests, the dry specimens were stored under desiccant at 25°C ± 2°C for a minimum of 3 days after injection molding and were tested at that temperature immediately after removal from the desiccant.

Table 2.3A of the Quality Assurance Plan lists the various properties measured, the methods used, the ASTM standard, and the anticipated precision. The precisions obtained in actual practice during the program are given in Table 12 of this report. The observed values were consistent with those reported in the literature for these types of materials, thus demonstrating that both our production processes and our testing procedures were satisfactory.

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TABLES AND FIGURES

Tables 1–10 and Figures 1–31 pertain to air-laid (AL) studies. The remaining tables and figures (Tables 11–34 and Figures 32–50) pertain to melt-blending (MB) studies.

TABLE 1. MATERIALS USED IN AIR-LAID STUDIES

Material	Abbreviation	Description	Source
Hemlock fiber	HF	Virgin western hemlock wood fiber	Canfor Ltd., Vancouver, BC
Demolition wood fiber	DF	Waste wood pressurized; refined into wood fiber	Wood Recycling, Peabody, MA
Virgin polyester fiber	PET	Polyester 280, 5.5 denier 38 mm, white, staple, crimped fiber	E.I. DuPont deNemours, Inc., Wilmington, DE
Recycled polyester	RPET	Spun from 2-L soft drink containers, 6.0 denier, 51 mm, bright, crimped fiber	Wellman Co., Inc., Johnsonville, SC
Virgin high density polyethylene	VHDPE	Fortiflex A 60-70-119 blow-molding polymer-milk bottle feedstock	Solvay Polymers, Houston, TX
Recycled high density polyethylene	RHDPE	Chopped, recycled milk bottles; ground to 35 mesh	Plastic World, Madison, WI
Phenolic resin	PR	Liquid phenol-formaldehyde GP 2341, 51 to 53 percent solids	Georgia-Pacific Corp.
Epolene-maleated polyethylene	E10	Wax emulsion of low molecular weight polyethylene	Eastman Chemical Co., Kingsport, TN
Recycled composite panel	Second generation	DF and RHDPE composite panels pressurized, refined into fiber form	Refined at FPL

TABLE 2. AL SERIES 1: PANEL COMPOSITION

Wood fiber content (percent)	PET fiber (percent)	Phenolic resin (percent)
HF 80	VPET 10	10
HF 80	RPET 10	10
DF 80	VPET 10	10
DF 80	RPET 10	10

TABLE 3. AL SERIES 1: PANEL SELECTION

Formulation	Panel size (mm)	Panels per composite ^a	Number of selected panels ^b
HF/VPET	279 by 279	40	15
HF/RPET	279 by 279	40	15
DF/VPET	279 by 279	40	15
DF/RPET	279 by 279	40	15

^aTotal number of panels per composite formulation.

^bFinal number of selected panels at specific gravity range 1.0 ± 0.03 .

TABLE 4. AL SERIES 1: MECHANICAL AND PHYSICAL PROPERTIES OF COMPOSITES

Property	HF-80% VPET-10% PR-10%	HF-80% RPET-10% PR-10%	DF-80% VPET-10% PR-10%	DF-80% RPET-10% PR-10%
Static bending MOR (MPa)	50.6 (16)	47.1 (22)	43.2 (25)	47.8 (20)
Static bending MOE (GPa)	3.66 (19)	4.26 (20)	3.23 (27)	3.74 (19)
Tensile strength (MPa)	33.0 (9)	28.4 (11)	28.3 (10)	30.0 (10)
Tensile MOE (GPa)	4.84 (14)	5.12 (13)	4.26 (18)	4.56 (13)
Impact energy (J)	36.1 (5)	28.7 (8)	34.2 (6)	30.7 (4)
Water-soak 24-h thickness swell (%)	25.2 (9)	22.3 (9)	29.8 (14)	26.9 (8)
Water soak 24-h water absorption (%)	43.4 (20)	41.3 (25)	48.2 (25)	44.1 (16)
Linear expansion at 30% RH (%)	0.19 (10)	0.21 (13)	0.20 (11)	0.20 (12)
at 65% RH (%)	0.42 (4)	0.44 (8)	0.43 (6)	0.45 (6)
at 90% RH (%)	0.61 (6)	0.70 (7)	0.64 (7)	0.71 (6)

^aValues in parentheses are coefficients of variation (%).

TABLE 5. AL SERIES 2: PANEL COMPOSITION

Wood fiber content (percent)	HDPE (percent)	VPET fiber (percent)	E10 wax tackifier (percent)
HF 60	VHDPE 30	5	5
HF 60	RHDPE 30	5	5
DF 60	VHDPE 30	5	5
DF 60	RHDPE 30	5	5

TABLE 6. AL SERIES 2: SPECIMEN SELECTION

Test ^a	Specimen size (mm)	Specimens per composite ^b	Number of selected specimens ^c
Static bending MOR and MOE	51 by 127	60	20
Tensile strength and MOE	51 by 254	50	20
Impact energy	254 by 254	20	10
24-h water soak	51 by 51	150	40
Linear expansion	13 by 152	100	25

^aForty panels were produced in every composite formulation

^bTotal number of specimens per composite formulation.

^cFinal number of selected specimens at specific gravity range
1.0 ± 0.06.

TABLE 7. AL SERIES 2: MECHANICAL AND PHYSICAL PROPERTIES OF COMPOSITES^a

Property	HF 60% VHDPE 30% VPET 5% E10 wax 5%	HF 60% RHDPE 30% VPET 5% E10 wax 5%	DF 60% VHDPE 30% VPET 5% E10 wax 5%	DF 60% RHDPE 30% VPET 5% E10 wax 5%
Static bending MOR (MPa)	16.8 (19)	18.7 (15)	19.1 (14)	17.4 (20)
Static bending MOE (GPa)	2.01 (19)	2.13 (19)	1.75 (16)	2.01 (24)
Tensile strength (MPa)	10.8 (15)	9.5 (19)	12.4 (14)	11.5 (12)
Tensile MOE (GPa)	2.81 (15)	2.23 (22)	2.12 (15)	2.09 (13)
Impact energy (J)	27.6 (9)	27.6 (7)	30.9 (8)	31.1 (7)
Water soak 24-h thickness swell (%)	43.8 (14)	42.7 (21)	45.2 (13)	52.8 (15)
Water soak 24-h water absorption (%)	54.9 (17)	61.8 (19)	58.7 (13)	65.8 (16)
Linear expansion at 30% RH (%)	0.15 (9)	0.17 (12)	0.17 (5)	0.16 (7)
at 65% RH (%)	0.39 (5)	0.42 (12)	0.40 (7)	0.44 (6)
at 90% RH (%)	0.68 (7)	0.69 (12)	0.64 (9)	0.74 (6)

^aValues in parentheses are coefficients of variation (%).

TABLE 8. RECYCLABILITY: PANEL COMPOSITION

Wood fiber content (percent)	Recycled panel content (percent)	HDPE (percent)	VPET fiber (percent)	E10 wax tackifier (percent)
DF 60	0	RHDPE 30	5	5
DF 40	20	RHDPE 30	5	5

TABLE 9. RECYCLABILITY: SPECIMEN SELECTION

Test ^a	Specimen size (mm)	Specimens per composite ^b	Number of selected specimens ^c
Static bending MOR and MOE	51 by 127	60	20
Tensile strength and MOE	51 by 254	50	20
Impact energy	254 by 254	20	10
24-h water soak	51 by 51	150	40
Linear expansion	13 by 152	100	25

^aForty panels were produced in every composite formulation.

^bTotal number of specimens per composite formulation.

^cFinal number of selected specimens at specific gravity range
1.0 ± 0.06.

TABLE 10. RECYCLABILITY: MECHANICAL AND PHYSICAL
PROPERTIES OF COMPOSITES^a

Property	DF 60% RHDPE 30% VPET 5% E10 wax 5%	DF 40% Recycled panel 20% RHDPE 30% VPET 5% E-10 wax 5%
Static bending MOR (MPa)	17.4 (20)	19.6 (13)
Static bending MOE (GPa)	2.01 (24)	1.77 (19)
Tensile strength (MPa)	11.5 (12)	13.7 (12)
Tensile MOE (GPa)	2.09 (13)	2.70 (25)
Impact energy (J)	31.1 (7)	33.2 (7)
Water soak 24-h thickness swell (%)	52.8 (15)	42.0 (12)
Water soak 24-h water absorption(%)	65.8 (16)	54.3 (12)
Linear expansion at 30% (RH) (%)	0.16 (7)	0.15 (11)
at 65% (RH) (%)	0.44 (6)	0.37 (9)
at 90% (RH) (%)	0.74 (6)	0.52 (11)

^a Values in parentheses are coefficients of variation (%).

TABLE 11. MATERIALS USED IN MELT-BLENDING STUDIES

Material	Abbreviation	Description ^a	Source
Plastic			
Virgin polypropylene	VPP	Fortilene 9101, 1602, 1902. Nominal MFI 3, 12, 30	Solvay Polymers, Inc., Deer Park, TX
Recycled polypropylene from auto battery cases	BPP	Cleaned chips. Nominal MFI 10	Gopher Smelting and Refining Co., Eagon, MN
Recycled polypropylene from ketchup bottles	KPP	Cleaned chips. Nominal MFI 3	Wheaton Plastic Recycling Co., Milleville, NJ
Recycled polyethylene from milk bottles	HDPE-MB	Cleaned chips. Nominal MFI 0.7	Recycle Worlds, Madison, WI
Cellulosic filler			
Wood flour	WF	Western pine. -40+80 mesh	American Woodfiber Co., Schofield, WI
Cellulose	BW40	Pure cellulose fiber. Mean length 60 um	James River Corp., Hackensack, NJ
Waste newspaper	ONP	Over production issue	Milwaukee Journal/Sentinel Inc., Milwaukee, WI
Waste magazine	OMG	Representative sample of Madison waste stream	Madison Recycling Center, Madison, WI
Coupling agent			
Epolene E43 powder	E43S	Powdered maleated poly- propylene. M = 4,200	Eastman Chemical Co., Kingsport, TN
Epolene E43 emulsion	E43E	Emulsified potassium salt	Eastman Chemical Co.
Epolene G3002 powder	G3002	Powdered maleated poly- propylene. M = 11,000	Eastman Chemical Co.

^a Meltflow index (MFI) is an inverse measure of melt viscosity in units of g/10 min at 230°C for polypropylene and g/10 min at 190°C for polyethylene.

TABLE 12. COEFFICIENTS OF VARIATION FOR
MEASURED MECHANICAL PROPERTIES^a

Property	COV (percent)
Tensile	
Strength	4
Modulus	5
Energy to maximum load	8
Elongation at maximum load	6
Flexural	
Strength	2
Modulus	5
Cantilever	
Strength	3
Modulus	6
Izod impact	
Notched	11
Unnotched	14

^a Averaged over Studies MB-1 to MB-4.

TABLE 13. STUDY MB-1: MECHANICAL PROPERTIES

MFI of PP (g/10 min)	Filler type	Filler content ^a	Tensile ^b		Flexure ^b		Cantilever ^c		Impact	
			Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
3	ONP	32	50.5	4.17	76.4	3.14	88.6	2.15	22.3	160
		37	50.4	4.33	78.9	3.33	84.5	2.35	22.5	149
		42	48.9	4.44	78.2	3.52	87.9	2.40	27.2	141
12	ONP	32	47.9	4.31	72.9	3.13	82.2	2.31	18.0	134
		37	48.5	4.32	75.2	3.35	83.0	2.33	22.7	158
		42	49.8	4.65	76.1	3.47	81.9	2.49	21.1	106
30	ONP	32	48.7	4.26	71.0	3.09	81.1	2.30	17.7	159
		37	47.7	4.46	73.5	3.36	78.3	2.48	23.6	117
		42	49.9	4.79	74.2	3.51	83.7	2.59	21.1	124
3	WF	42	39.3	4.05	62.7	3.22	77.3	2.26	23.2	105
12			37.6	4.53	61.6	3.53	71.3	2.38	20.5	83
30			38.1	4.50	59.8	3.42	67.8	2.37	24.6	73

^aFiller/polymer weight ratio. Composites contain 5 percent E43 by weight of filler.^bMaximum strength and initial modulus.^cMaximum strength and 9° secant modulus.

TABLE 14. STUDY MB-1: ANALYSIS OF VARIANCE SUMMARY^a

Effect	Variable	Critical ^b F ratio	Cantilever		Flexure		Tensile		Impact	
			Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
Matrix A										
Main effect	MFI of PP (3, 12, or 30 g/10 min)	4.3	22.4	32.9	32.8	0.0	3.9	3.2	2.7	1.0
	ONP content (32, 37, or 42 percent)	4.3	0.3	76.6	11.9	14.8	1.5 *	13.2	3.4	1.9
Interactions	Filler content x MFI	3.6	1.7	4.5	0.4	0.1	2.6	0.9	1.0	1.2
Matrix B										
Main effect	Filler type (WF or ONP)	6.0	253.6	10.8	17.6	4.8	8.9	12.8	0.4	27.1
	MFI of PP (3, 12, or 30 g/10 min)	5.1	99.5	3.4	2.1	2.2	0.8	11.4	1.7	6.2
Interactions	Filler content x MFI	5.1	27.0	0.5	1.0	4.2	1.0	1.1	0.8	1.3

^aValues are the F ratios, which are the variances of response attributable to the particular variable divided by the variances resulting from experimental error.

^bExperimental F ratios must be greater than the critical values for the effect to be significant at the 95 percent confidence level.

TABLE 15. STUDY MB-1: PROPERTY CHANGES^a

Property	Variables ^b	
	ONP/PP ratio (32 to 42)	Filler type (WF to ONP)
Cantilever		
Strength	--	17
Modulus	11	--
Flexural		
Strength	--	24
Modulus	12	--
Tensile		
Strength	--	--
	--	29
Impact		
Unnotched	--	43

^a Values are given only when statistically significant at 95 percent confidence level and change is above 10 percent.

^b Averaged over MFI from 3 to 30 g/10 min.

TABLE 16. COMPOSITE MELT VISCOSITIES^a

PP MFI (g/12 min)	Filler	Filler/PP (weight)	Viscosity at shear rate(s ⁻¹)		
			100	500	1000 ^b
3	--	0	1,230 (3.4)	--	--
12	--	0	359 (1.0)	149 (1.0)	102 (1.0)
	WF	42/58	452 (1.3)	195 (1.3)	136 (1.3)
	ONP	32/68	618 (1.7)	227 (1.5)	148 (1.5)
	ONP	37/63	728 (2.0)	269 (1.8)	175 (1.7)
	ONP	42/58	881 (2.5)	301 (2.0)	190 (1.9)
30	ONP	42/58	572 (1.6)	208 (1.4)	135 (1.3)

^aValues in Pa-s. Parenthetical values are ratios of filled system to unfilled PP at 10 MFI and particular shear rate. E-43 present at 5 weight percent of filler.

^bExtrapolated from log-log plot of data measured between 100 and 500 s⁻¹.

TABLE 17. STUDY MB-2: MECHANICAL PROPERTIES^a

Experiment	Polymer	Filler	Epolene ^b	Compounder	Cantilever ^c		Flexure ^c		Tensile ^d		Impact	
					Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
A	HDPE-MB	BW40	Solid	K-mixer	64.0	2.13	53.3	2.95	36.3	3.79	30.8	68
			Emulsion	K-mixer	61.7	2.01	48.2	2.55	34.3	2.86	34.0	105
		ONP	Solid	K-mixer	65.9	2.32	55.6	3.28	37.6	3.80	28.6	73
			Emulsion	K-mixer	63.0	2.33	53.4	3.24	35.8	4.00	30.6	67
	PPV	BW40	Solid	K-mixer	85.9	2.71	78.5	3.85	48.2	4.80	24.7	114
			Emulsion	K-mixer	81.1	2.48	71.7	3.25	43.7	3.62	19.8	119
		ONP	Solid	K-mixer	83.5	2.58	76.7	4.11	47.1	4.89	20.8	109
			Emulsion	K-mixer	72.0	2.59	66.9	3.92	38.0	4.60	20.7	81
B	HDPE-MB	BW40	Solid	Extruder	65.0	2.04	49.5	2.75	35.8	3.90	28.6	84
				K-mixer	64.0	2.13	53.3	2.95	36.3	3.79	30.8	68
		WF		Extruder	57.0	1.96	40.8	2.63	30.9	3.81	33.8	66
				K-mixer	53.9	1.85	40.3	2.26	27.8	2.61	36.4	81
	PPV	BW40		Extruder	73.4	2.46	62.5	4.03	35.2	4.03	24.9	103
				K-mixer	85.9	2.71	78.5	3.85	48.2	4.80	24.7	114
		WF		Extruder	70.7	2.48	62.1	3.98	37.4	4.62	25.7	88
				K-mixer	68.6	2.46	58.1	3.21	34.1	3.72	18.7	72

^aValues are the means from 10 specimens (five each from the two replicate matrices). All systems at 40/60 weight ratio of filler to polymer.^bEpolene E43 at 5 weight percent of filler.^cMaximum strength and secant modulus at 9°.^dMaximum strength and initial modulus.

TABLE 18. STUDY MB-2-A: MAIN EFFECTS AND INTERACTIONS

Effect	Variable	Cantilever		Flexure		Tensile		Impact	
		Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
Overall mean		72.1	2.39	63.0	3.39	40.1	4.04	26.2	92.0
Main effect ^a	Polymer (HDPE to PP)	+17.0	+0.39	+20.7	+0.78	+8.3	+0.87	-9.5	+27.7
	Filler (BW40 to ONP)		+0.13		+0.49		+0.55	-2.2	-19.1
Interactions ^b	E43 (emulsion to solid)	+5.4		+6.1		+4.4	+0.55		
	Polymer x filler	-3.6	-0.13	-3.6		-2.4			
	Polymer x E43			+2.4		+2.5		+2.5	+12.9
	Filler x E43					+1.1	-0.50		+18.8

^a Change in property resulting from the particular variable, averaged over all other variables. Values are given only when effects are significant at the 95 percent confidence level.

^b X x Y interaction = 1/2 (average effect of X at first level of Y minus average effect of X at second value of Y). Values are given only when effects are significant at the 95 percent confidence level.

TABLE 19. STUDY MB-2-B: MAIN EFFECTS AND INTERACTIONS

Effect	Variable	Cantilever		Flexure		Tensile		Impact	
		Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
Overall mean		65.1	2.25	55.6	3.20	35.7	3.91	28.0	94.7
Main effect ^a									
	Polymer (HDPE to PP)	+10.4	+0.53	+19.3	+1.12	+6.1	+0.77	-8.9	+19.4
	Filler (WF to BW40)	+5.2	+0.15	+10.6		+6.3	+0.44		-15.4
	Compounder (extruder to K-mixer)						-0.36		
Interactions ^b									
	Polymer x filler							+4.1	
	Polymer x compounder							-3.0	
	Filler x compounder			+6.1		+5.0	-0.69		

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^a Change in property resulting from the particular variable, averaged over all other variables. Values are given only when effects are significant at the 95 percent confidence level.

^b X x Y interaction = 1/2(average effect of X at first level of Y minus average effect of X at second value of Y). Values are given only when effects are significant at the 95 percent confidence level.

TABLE 20. STUDY MB-2: USEFUL MECHANICAL PROPERTY CHANGES^a

Property	Variables		
	HDPE-MB to PP ^b	WF to BW40 ^c	BW40 to ONP ^d
Cantilever			
Strength	23	(e)	(e)
Modulus	23	(e)	(e)
Flexural			
Strength	41	21	(e)
Modulus	34	(e)	(e)
Tensile			
Strength	21	19	(e)
Modulus	23	(e)	(e)
Impact			
Notched	-29	(e)	(e)
Unnotched	29	(e)	-19

^aPercent relative to mean values.

^bAveraged over other variables in Study MB2-A and Study MB2-B.

^cAveraged over other variables in Study MB2-A.

^dAveraged over other variables in Study MB2-B.

^eChanges less than 20 percent.

TABLE 21. COMPARISON OF MECHANICAL PROPERTIES OF HDPE-MB/ONP AND PPV/WF COMPOSITES

Property	Ratio ^a
Tensile strength	1.07
Tensile modulus	0.98
Flexural strength	0.95
Flexural modulus	0.97
Notched impact energy	1.55
Unnotched impact energy	0.95

^a Average value for HDPE-MB/ONP systems to value for PPV/WF systems.

TABLE 22. CHARACTERISTICS OF EPOLENES

Epolene	Acid number (mg KOH per g Epolene)	M _n	Anhydrides per Epolene molecule (no.)	Polymer repeat units per anhydride (no.)
E43	47	4,200	1.76	36
G3002	63	11,000	6.18	37

TABLE 23. STUDY MB-3: MECHANICAL PROPERTIES^a

Coupling agent	Tensile ^b			Flexure		Cantilever		Impact	
	Strength (MPa)	Modulus (GPa)	Elongation (percent)	Energy (J)	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Unnotched (J/m)
None	34.0	4.97	2.0	0.29	61.3	4.40	66.4	2.53	113
E43	47.4 (1.4)	4.90 (1.0)	2.0 (1.0)	0.34 (1.2)	81.9 (1.3)	4.21 (1.0)	86.1 (1.3)	2.61 (1.0)	144 (1.3)
G3002	52.3 (1.5)	4.56 (0.9)	3.0 (1.5)	0.59 (2.0)	82.0 (1.3)	4.20 (1.0)	91.8 (1.4)	2.54 (1.0)	190 (1.7)

^aONP/polymer weight ratio = 40/60. Composites contain 3 percent coupling agent by weight of filler. Numbers in parentheses are the property values divided by the corresponding property value without additive.

^bElongation and tensile energy to maximum load.

TABLE 24. STUDY MB-4: MECHANICAL PROPERTIES^a

Polymer	Filler	Tensile			Flexure		Impact		
		Strength (MPa)	Modulus (GPa)	Elongation (percent)	Energy (J)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
KPP	None	36.5	1.62	7.8	1.14	48.4	1.49	62.0	>800
	ONP	52.3	4.03	3.7	0.74	73.8	3.30	30.6	167
	OMG	38.9	3.55	3.1	0.48	61.8	3.01	34.2	138
BPP	None	24.5	1.32	7.4	0.80	31.4	1.05	165.0	>800
	ONP	42.5	3.98	3.4	0.58	59.9	3.16	34.3	150
	OMG	31.8	3.44	3.0	0.40	49.2	2.69	41.8	125
VPP	None	31.5	--	--	--	41.1	1.43	23.8	650
	ONP	52.3	4.12	--	--	82.0	4.20	20.4	190

^a Strength is maximum value; elongation is at maximum strength; energy is area under stress/strain curve to maximum strength. Dashes indicate data not available. Filler/polymer weight ratio = 40/60. Composites contain 3 percent G3002 by weight of filler.

TABLE 25. STUDY MB-4: MAIN EFFECTS OF VARIABLES

Property	Overall mean ^a	Main effect ^b		
		KPP to BPP	OMG to ONP	
Tensile				
Modulus (GPa)	3.8	**	+0.5 (+15)	
Strength (MPa)	61.4	-8.4 (-18)	+12.1 (+34)	
Energy (J)	0.55	-0.13 (-20)	+0.22 (+50)	
Elongation (%)	3.3	-0.2 (-6)	+0.4 (+17)	
Flexural				
Modulus (GPa)	3.0	-0.2 (-7)	+0.4 (+12)	
Strength (MPa)	61.2	-13.3 (-19)	+11.3 (+17)	
Impact				
Notched (J/m)	35.3	+5.6 (+18)	-5.6 (-14)	
Unnotched (J/m)	145	**	**	

^a Average over all variables and trials.

^b Main effect is change in property resulting from the particular variable, averaged over the other variable. Numbers in parentheses are percent change. Interactions were not significant at the 95 percent confidence level.

** Not significant at 95 percent confidence level.

TABLE 26. Study MB-4: CHANGE IN PROPERTY
AFTER ADDITION OF 40 PERCENT FILLER TO PLASTIC^a

Property	Filled/unfilled		
	KPP	BPP	VPP
Tensile			
Modulus	2.49	3.00	--
Strength	1.49	1.66	1.66
Energy	0.65	0.73	--
Elongation	0.47	0.46	--
Flexural			
Modulus	2.29	3.01	2.94
Strength	1.52	1.91	2.00
Impact			
Notched	0.49	0.21	0.86
Unnotched	<0.21	<0.19	0.29

^aBased on values for ONP systems. Dashes indicate data not available.

TABLE 27. WATER SORPTION OF VPP WITH 42 percent ONP AND
3 PERCENT G3002 (BY WEIGHT OF FILLER)^a

	Initial	24-h ambient soak	24-h boil	Boil/initial
Cantilever				
Secant modulus (GPa)	2.69	2.58	1.73	0.6
Max strength (MPa)	83.3	84.9	71.6	0.9
Izod impact				
Notched (J/m)	24.5	25.6	26.2	
Unnotched (J/m)	131	150	145	
Thickness (mm)	3.15	3.15	3.28	1.0
Weight (g)	6.02	6.06	6.32	1.0

^aContinuous lines over numbers indicate that they are not statistically significant at the 95 percent confidence level.

TABLE 28. WATER SORPTION OF VPP WITH 42 PERCENT ONP (NO COUPLING AGENT)^a

	Initial	24-h ambient soak	24-h boil	Boil/initial
Cantilever				
Secant modulus (GPa)	<u>2.63</u>	<u>2.51</u>	<u>1.68</u>	0.6
Max strength (MPa)	<u>68.5</u>	<u>68.3</u>	<u>62.0</u>	0.9
Izod impact				
Notched (J/m)	<u>24.5</u>	<u>24.8</u>	<u>27.0</u>	
Unnotched (J/m)	<u>102</u>	<u>105</u>	<u>145</u>	1.4
Thickness (mm)	<u>3.14</u>	<u>3.15</u>	<u>3.24</u>	1.0
Weight (g)	<u>6.03</u>	<u>6.08</u>	<u>6.43</u>	1.1

^aContinuous lines over numbers indicate that they are not statistically significant at the 95 percent confidence level.

TABLE 29. WATER SORPTION OF VPP WITH 42 PERCENT WF (NO COUPLING AGENT)^a

	Initial	24-h ambient soak	24-h boil	Boil/initial
Cantilever				
Secant modulus (GPa)	2.22	2.24	1.59	0.7
Max strength (MPa)	65.6	64.7	59.4	0.9
Izod impact				
Notched (J/m)	23.9	21.8	24.4	
Unnotched (J/m)	102	99	101	
Thickness (mm)	3.17	3.20	3.26	1.0
Weight (g)	5.97	6.01	6.25	1.0

^aContinuous lines over numbers indicate that they are not statistically significant at the 95 percent confidence level.

TABLE 30. ANALYSIS OF VARIANCE SUMMARY

System	Cantilever			Impact		
	Critical F ratio ^a	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)	Thickness (mm)
PP with 40 percent ONP ^b and 3.0 percent G3002 ^b	3.9	89.2	96.8	1.1	1.2	21.7
PP with 40 percent ONP (no coupling)	3.9	22.6	241.0	2.2	13.3	14.1
PP with 40 percent WF (no coupling)	3.9	17.6	66.1	3.1	0.2	54.0
						14.1

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^a F ratio is the variance of response attributable to the particular variable divided by the variance resulting from experimental error. Experimental F ratio must be greater than the critical F ratio to be significant at the 95 percent confidence level.

^b G3002 was added at a level of 3 percent by weight of filler.

TABLE 31. STUDY MB-5: SYSTEM 1 MECHANICAL PROPERTIES AFTER RECYCLING^a

	Initial	Cycle 1	Cycle 2	Cycle 4	Cycle 5	Cycle 5/initial
Cantilever						
Secant modulus (GPa)	2.46	2.47	2.63	2.63	2.25	0.9
Max strength (MPa)	60.0	62.7	63.9	65.8	52.0	0.9
Izod impact						
Notched (J/m)	21.1	18.1	17.5	17.4	19.2	16.4
Unnotched (J/m)	86	100	91	93	93	97
Tensile						
Modulus (GPa)	4.22	4.85	4.84	4.96	4.57	4.51
Max strength (MPa)	26.9	32.3	33.8	32.6	31.7	29.5
Energy (J)	0.22	0.27	0.30	0.25	0.24	0.23
Elongation (percent)	1.8	1.9	2.2	1.9	1.9	1.9
Rheological properties						
Melt flow index (g/10 min)	0.07	0.12	0.14	0.07	0.11	0.13

^a System 1 is VPP with 42 percent ONP. Each cycle is one pass through the extruder and granulation. Continuous lines over numbers indicate that they are not statistically different at the 95 percent confidence level.

TABLE 32.. STUDY MB-5: SYSTEM 2 MECHANICAL PROPERTIES AFTER RECYCLING^a

	Initial	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 5/initial
Cantilever							
Secant modulus (GPa)	1.88	1.95	1.91	1.93	1.90	1.84	
Max strength (MPa)	52.2	54.1	54.5	52.2	51.0	52.4	
Izod impact							
Notched (J/m)	50.5	42.8	38.1	39.0	37.9	40.6	0.8
Unnotched (J/m)	134	97	94	112	109	111	0.8
Tensile							
Modulus (GPa)	2.65	3.10	3.12	3.23	3.21	3.31	1.3
Max strength (MPa)	26.9	32.3	33.8	32.6	31.7	29.5	1.1
Energy (J)	0.49	0.47	0.46	0.46	0.51	0.49	
Elongation (percent)	4.4	4.1	3.9	4.1	4.5	4.4	

^a System 2 is HDPE-MB with 42 percent WF. Each cycle is one pass through the extruder and granulation. Continuous lines over numbers indicate that they are not statistically different at the 95 percent confidence level.

TABLE 33. STUDY MB-5: SYSTEM 3 MECHANICAL PROPERTIES AFTER RECYCLING^a

	Initial	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 5/initial
Cantilever							
Secant modulus (GPa)	1.62	1.85	1.82	1.72	1.79	1.79	1.1
Max strength (MPa)	50.7	51.4	51.6	48.3	49.2	48.7	1.0
Izod impact							
Notched (J/m)	51.8	45.3	39.1	39.8	38.5	40.3	0.8
Unnotched (J/m)	121	98	83	104	95	97	0.8
Tensile							
Modulus (GPa)	2.72	2.79	2.85	2.90	2.76	2.75	1.3
Max strength (MPa)	26.1	25.3	24.8	26.0	25.2	24.0	0.9
Energy (J)	0.51	0.52	0.48	0.50	0.48	0.52	
Elongation (percent)	4.5	4.6	4.5	4.5	4.9	4.9	

^aSystem 3 is HDPE-MB/40 percent air-laid composite. Air-laid composite = 80 percent DF/10 percent PR/10 percent PET. Each cycle is one pass through the extruder and granulation. Continuous lines over numbers indicate that they are not statistically different at the 95 percent confidence level.

TABLE 34. STUDY MB-5: ANALYSIS OF VARIANCE SUMMARY^a

System	Cantilever		Tensile			Impact		Melt flow index (g/10 min)
	Strength (MPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)	Elongation (percent)	Energy (J)	Notched (J/m)	Unnotched (J/m)
System 1	6.71	4.76	7.00	4.16	0.96	1.54	5.66	1.28
System 2	2.54	0.45	7.00	5.44	1.89	0.60	3.58	1.94
System 3	4.10	4.32	4.52	0.97	0.76	0.31	19.7	3.89

^aValues are the F ratios, which are the variances of response attributable to the particular variable divided by the variances resulting the experimental error. Experimental F ratios must be greater than a critical value of 2.62 for the effect to be significant at the 95 percent confidence level.

RANDO-WEB® PROCESS

MODEL B RANDO-WEB® PROCESS

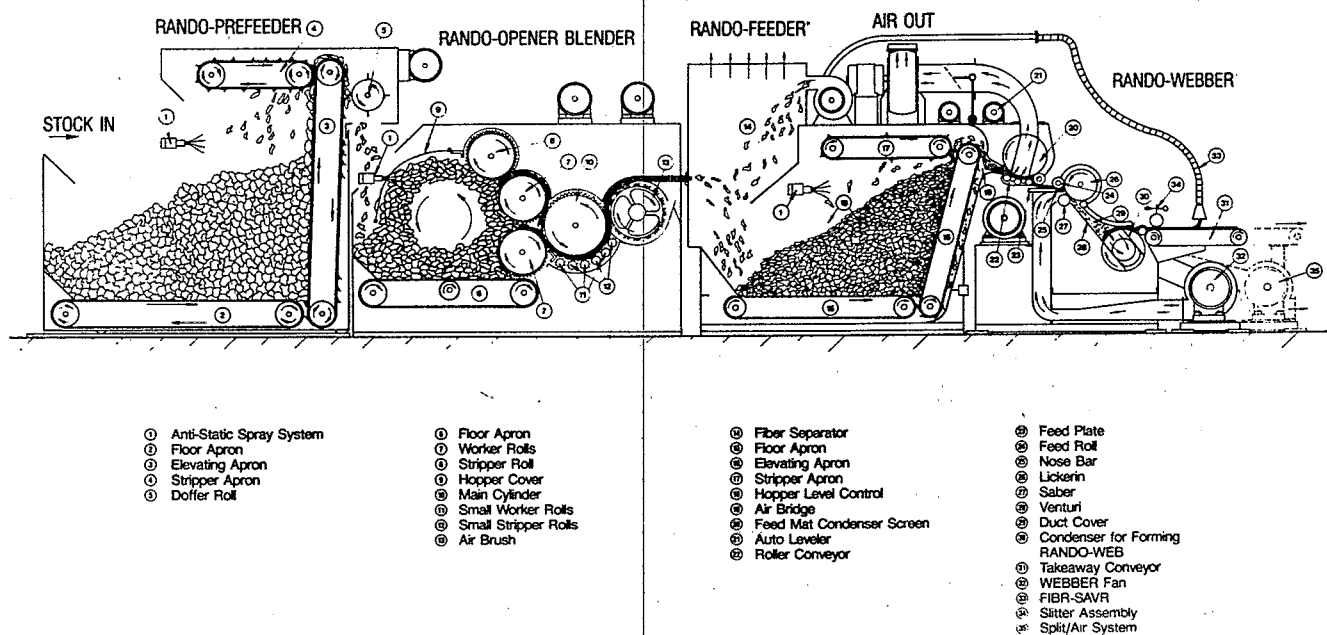


Figure 1. Schematic of air-laid web-forming process.

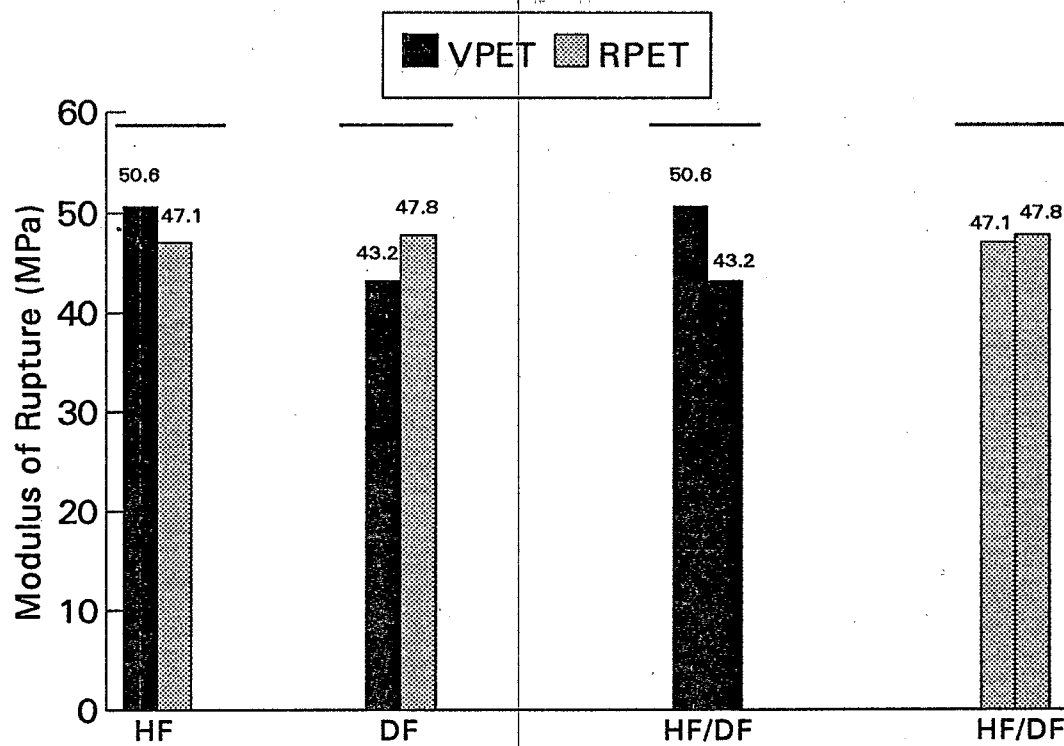


Figure 2. Task 3, AL Series 1. Bending strength (MOR) as a function of virgin hemlock fiber (HF), recycled demolition fiber (DF) and polyester fiber (PET). VPET and RPET designate virgin and recycled polyester fiber, respectively.

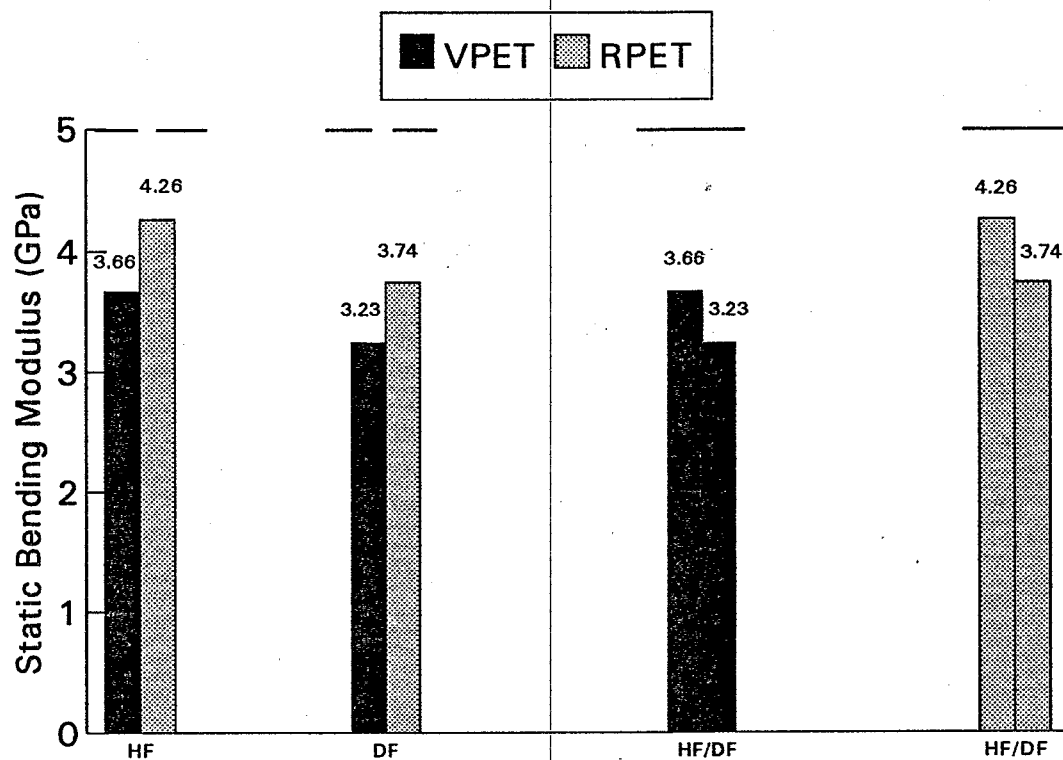


Figure 3. Task 3, AL Series 1. Bending stiffness (MOE) as a function of virgin HF and recycled DF and PET.

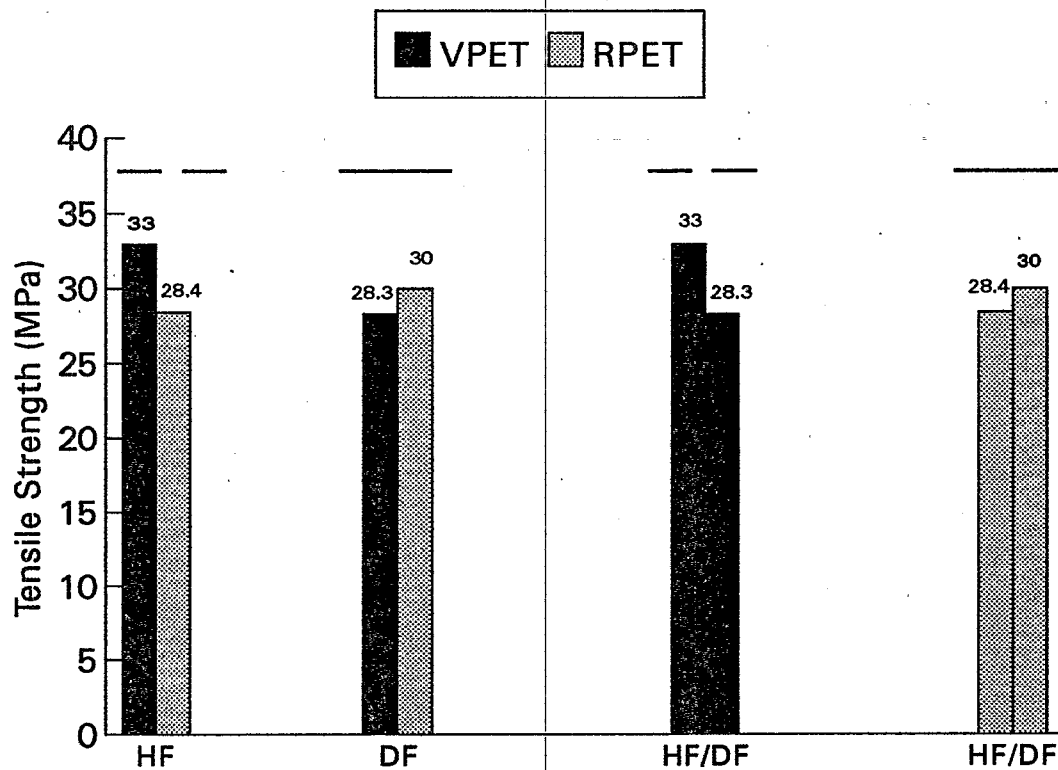


Figure 4. Task 3, AL Series 1. Tensile strength as a function of virgin HF and recycled DF and PET.

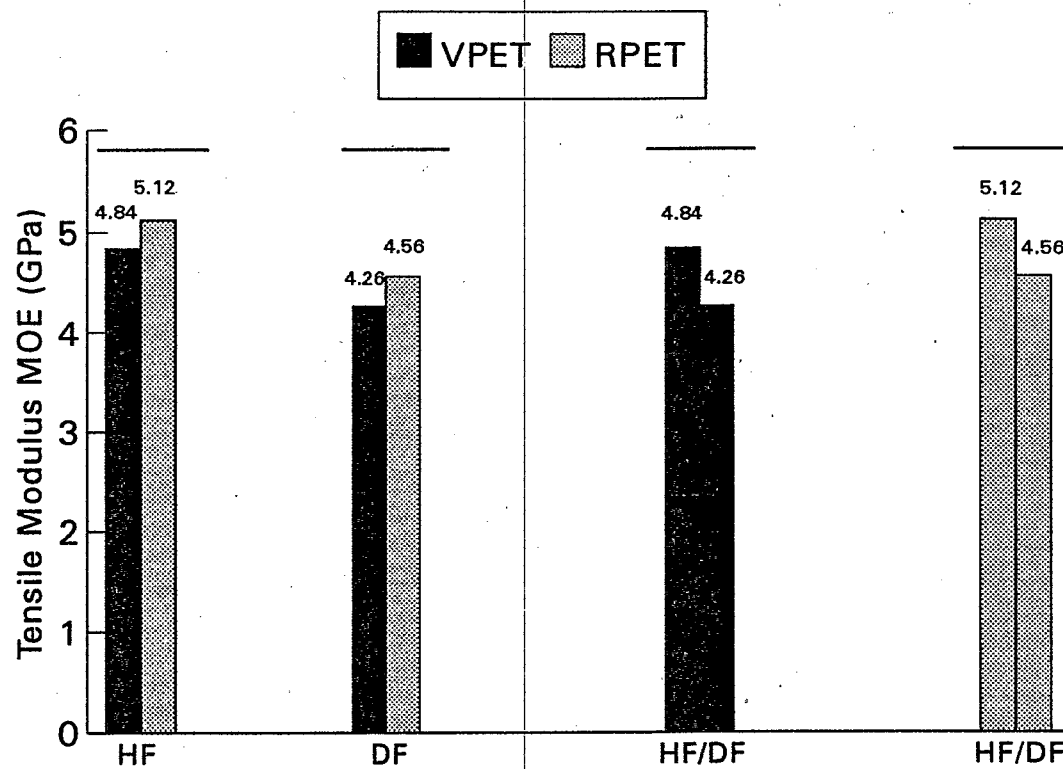


Figure 5. Task 3, AL Series 1. Tensile modulus (MOE) as a function of virgin HF and recycled DF and PET.

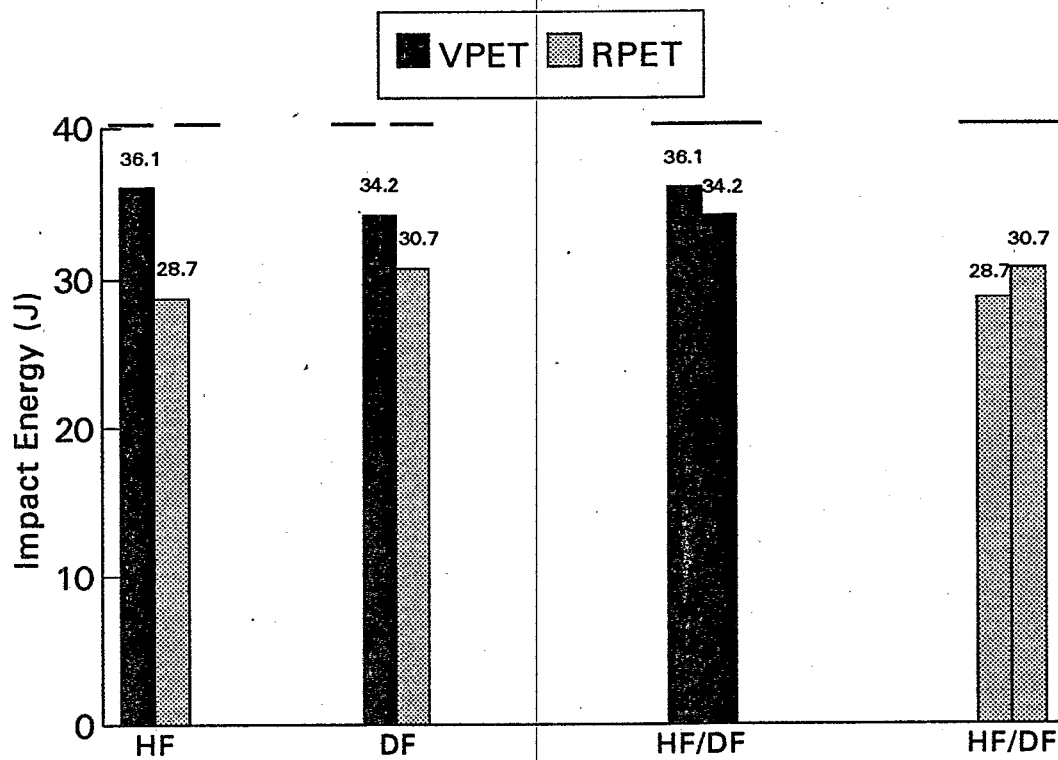


Figure 6. Task 3, AL Series 1. Impact energy as a function of virgin HF and recycled DF and PET.

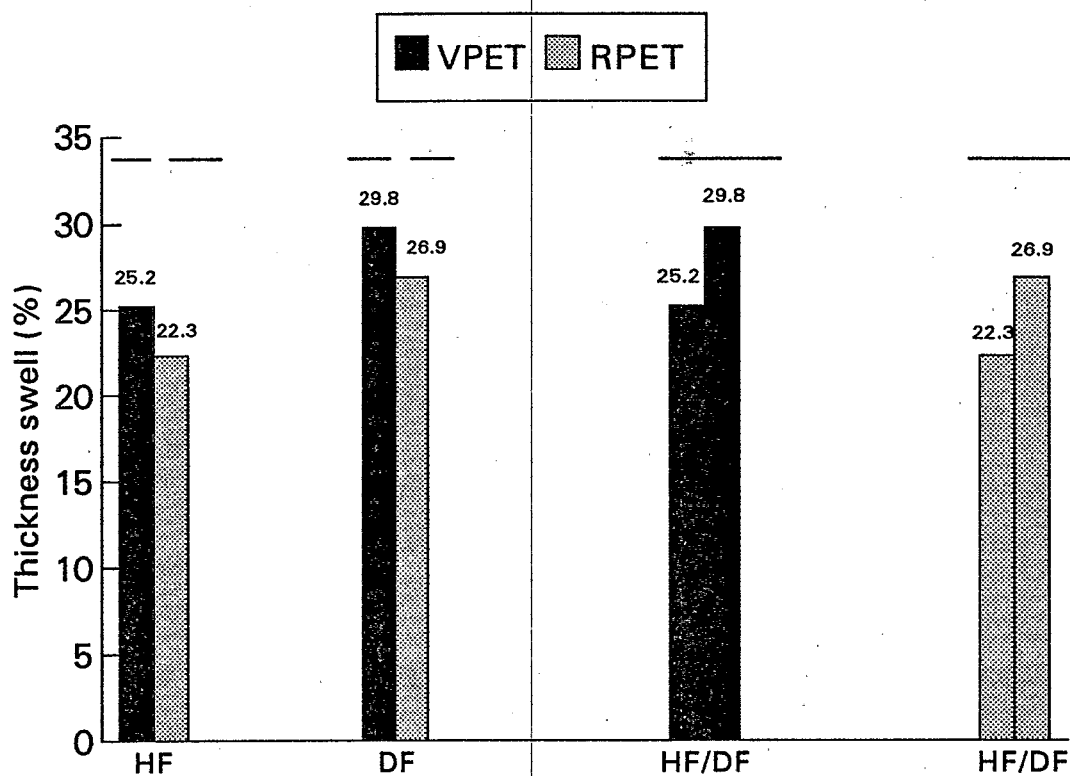


Figure 7. Task 3, AL Series 1. Thickness swell as a function of virgin HF and recycled DF and PET.

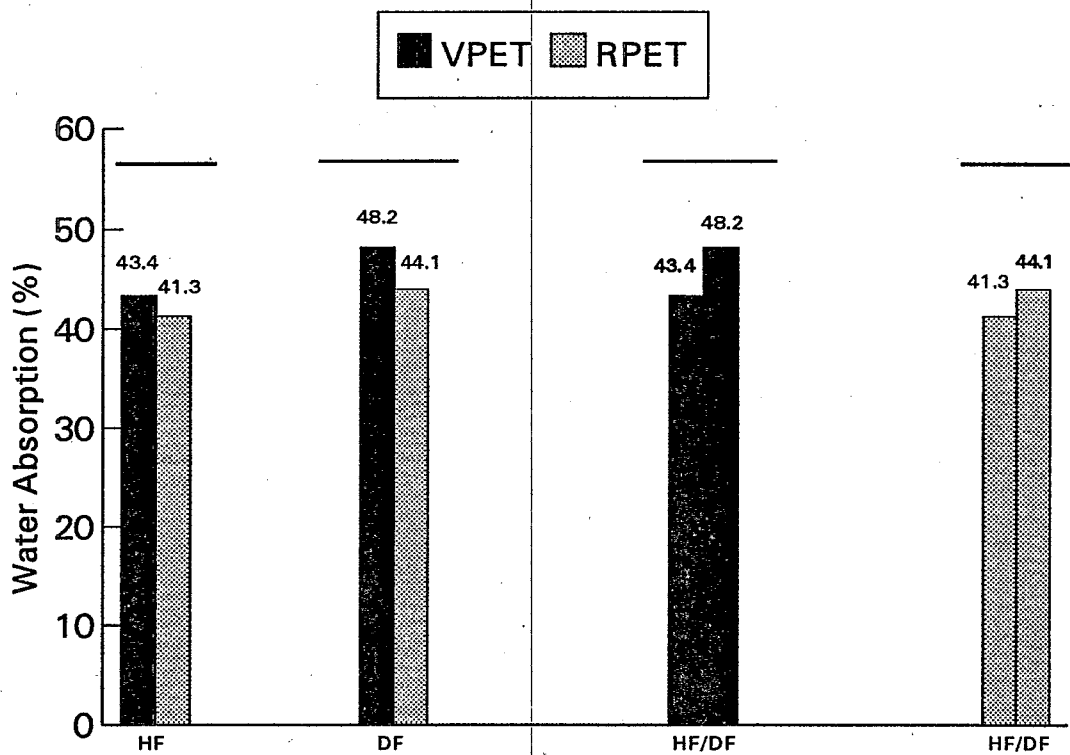


Figure 8. Task 3, AL Series 1. Water absorption as a function of virgin HF and recycled DF and PET.

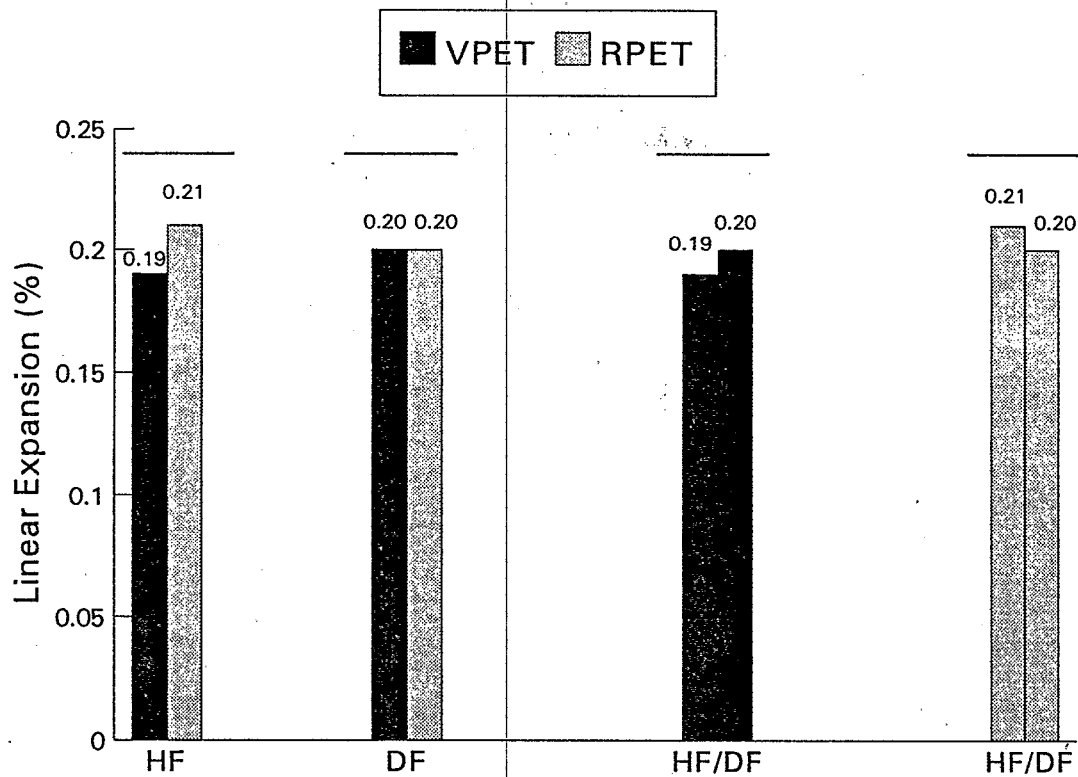


Figure 9. Task 3, AL Series 1. Linear expansion at 30 percent relative humidity (RH) as a function of virgin HF and recycled DF and PET.

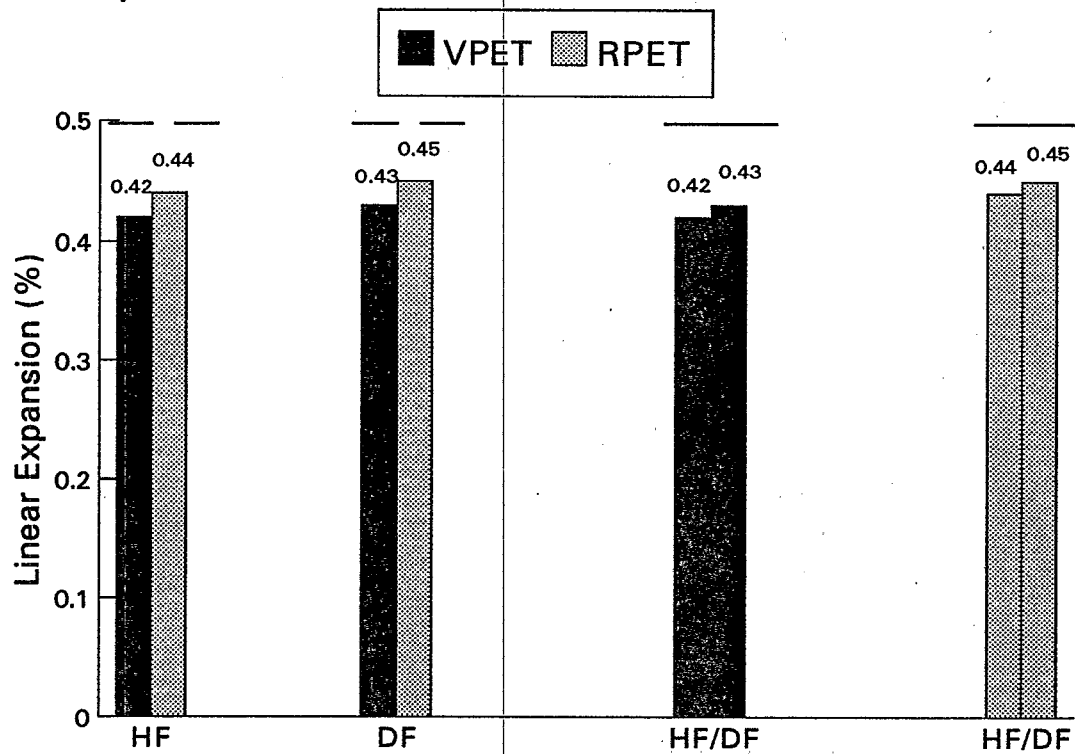


Figure 10. Task 3, AL Series 1. Linear expansion at 65 percent RH as a function of virgin HF and recycled DF and PET.

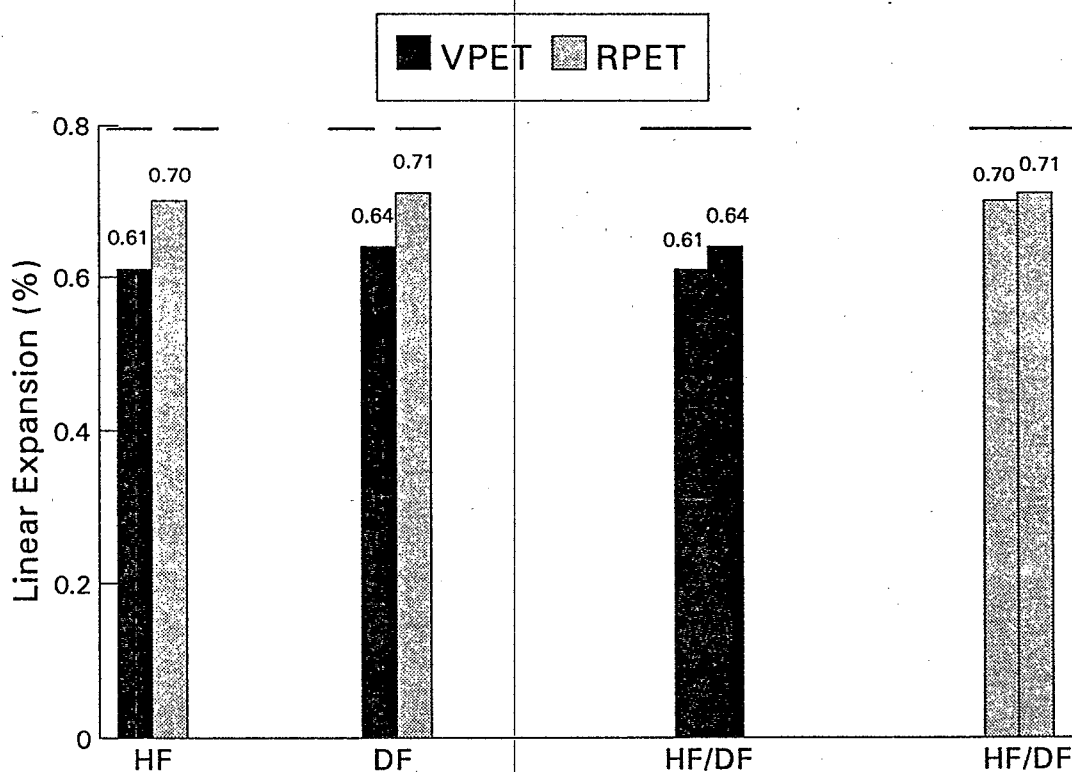


Figure 11. Task 3, AL Series 1. Linear expansion at 90 percent RH as a function of virgin HF and recycled DF and PET.

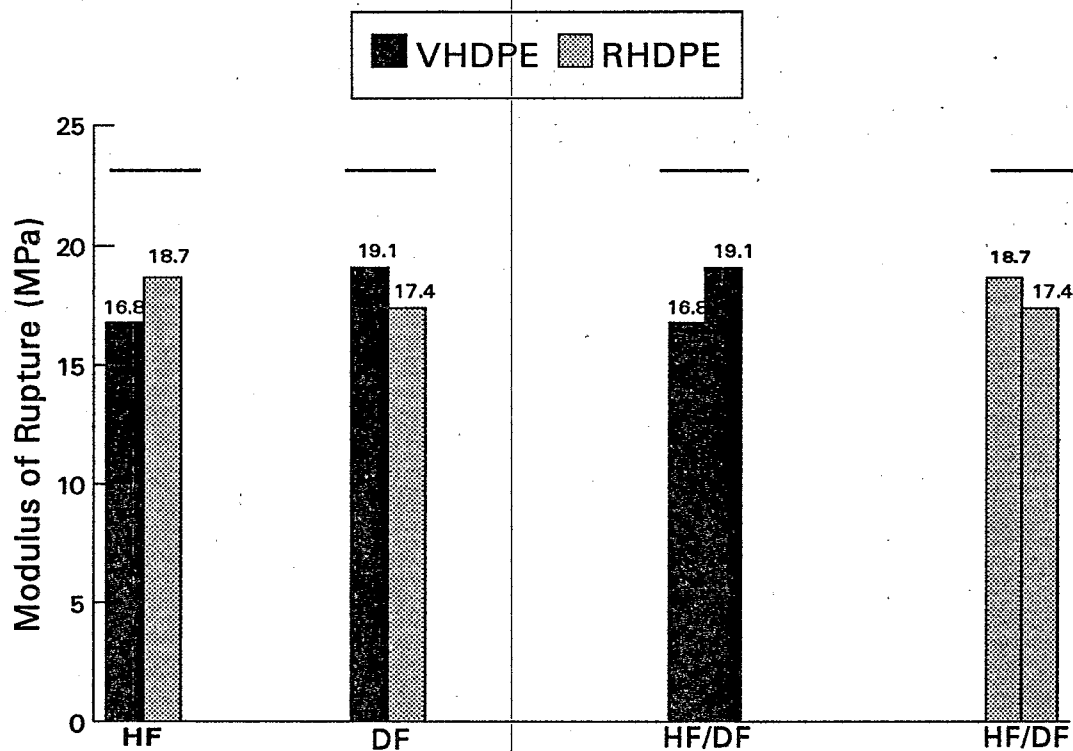


Figure 12. Task 3, AL Series 2. Bending strength (MOR) as a function of virgin HF and recycled DF and high density polyethylene (HDPE). VHDPE and RHDPE designate virgin and recycled HDPE, respectively.

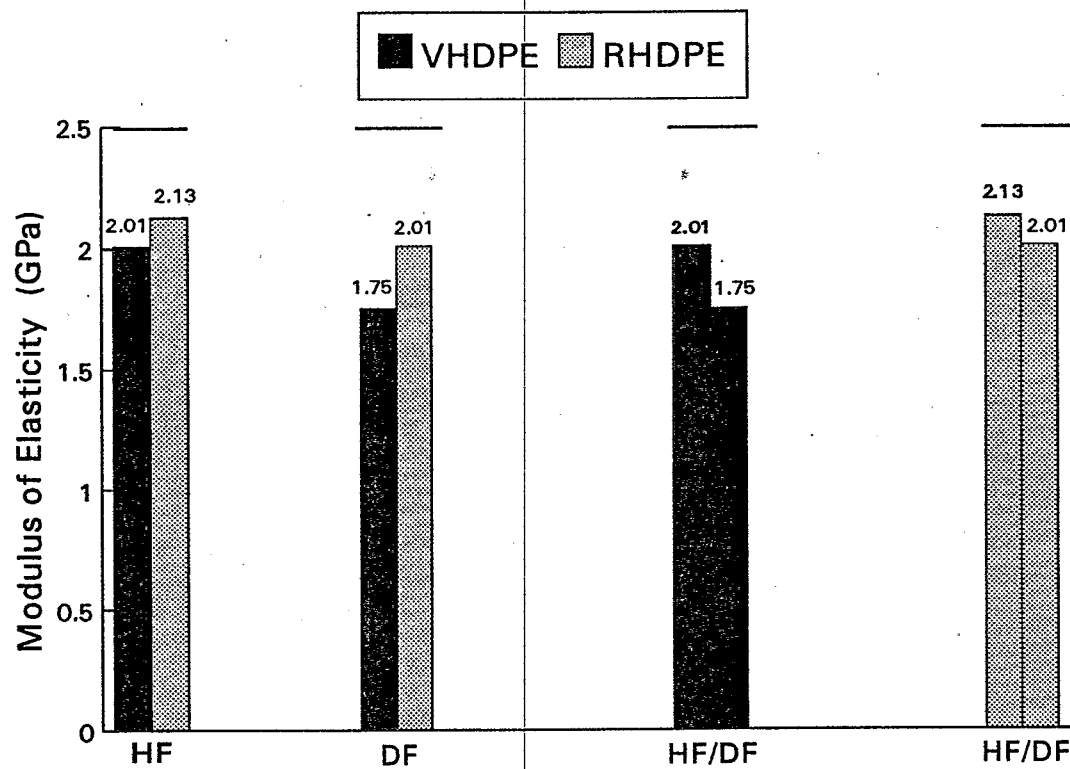


Figure 13. Task 3, AL Series 2. Bending stiffness (MOE) as a function of virgin HF and recycled DF and HDPE.

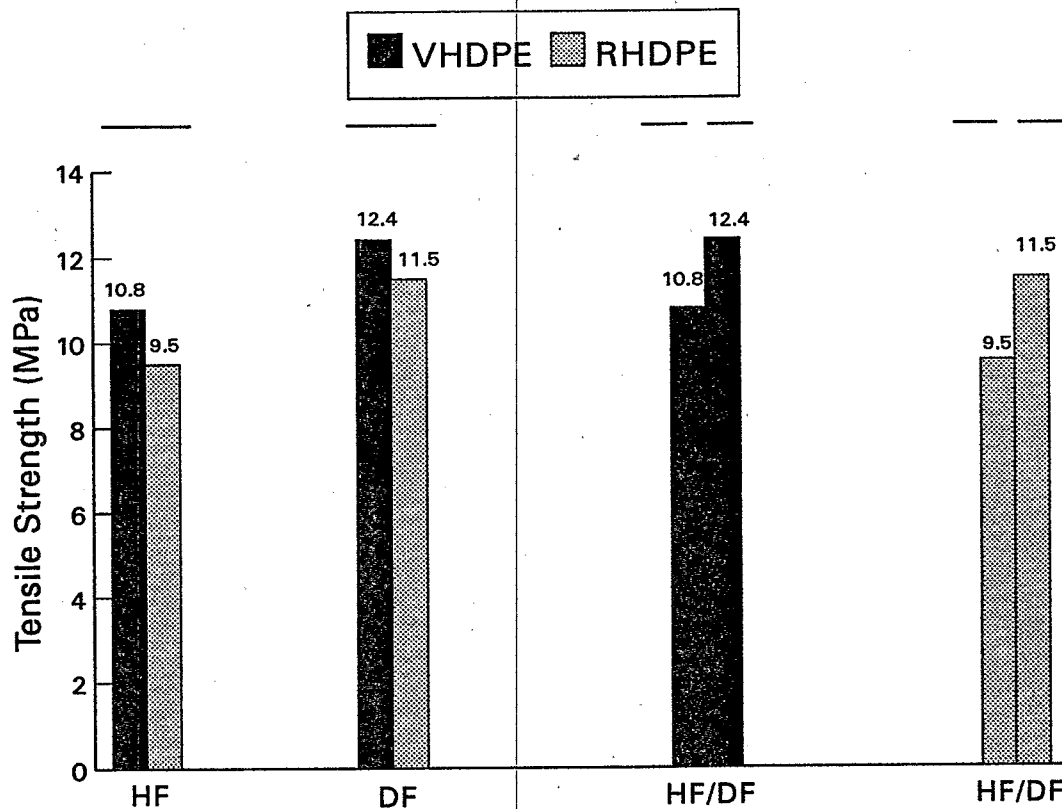


Figure 14. Task 3, AL Series 2. Tensile strength as a function of virgin HF and recycled DF and HDPE.

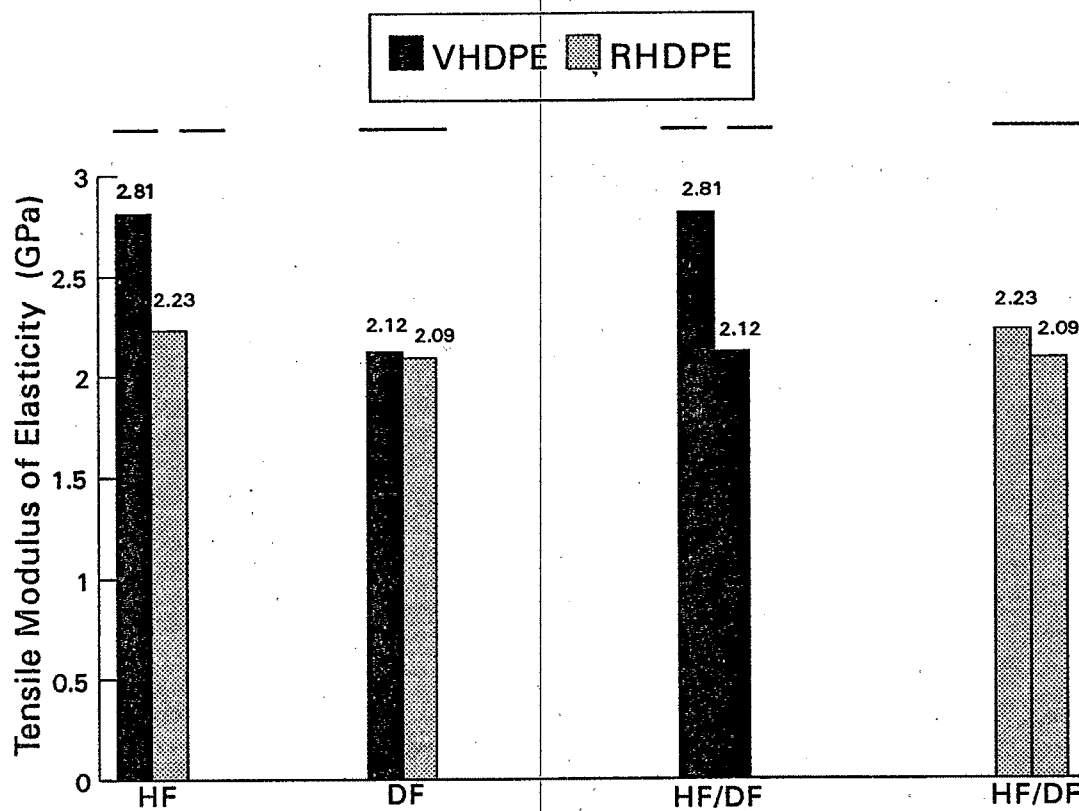


Figure 15. Task 3, AL Series 2. Tensile modulus (MOE) as a function of virgin HF and recycled DF and HDPE.

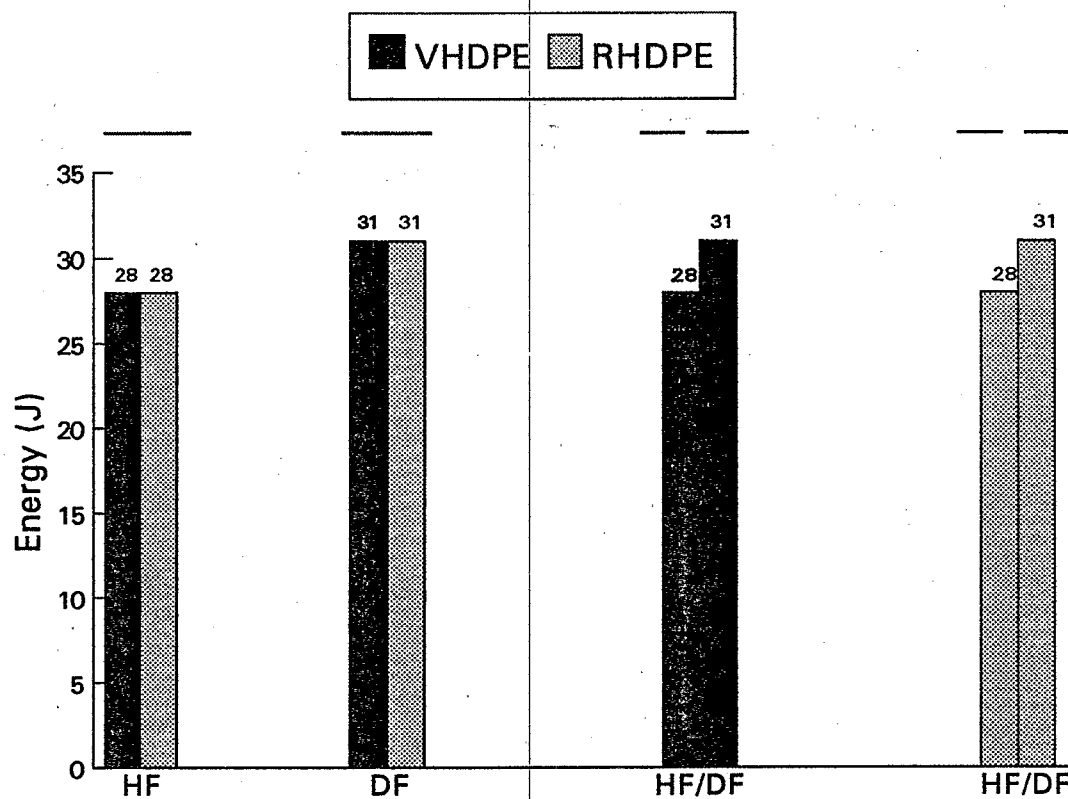


Figure 16. Task 3, AL Series 2. Impact energy as a function of virgin HF and recycled DF and HDPE.

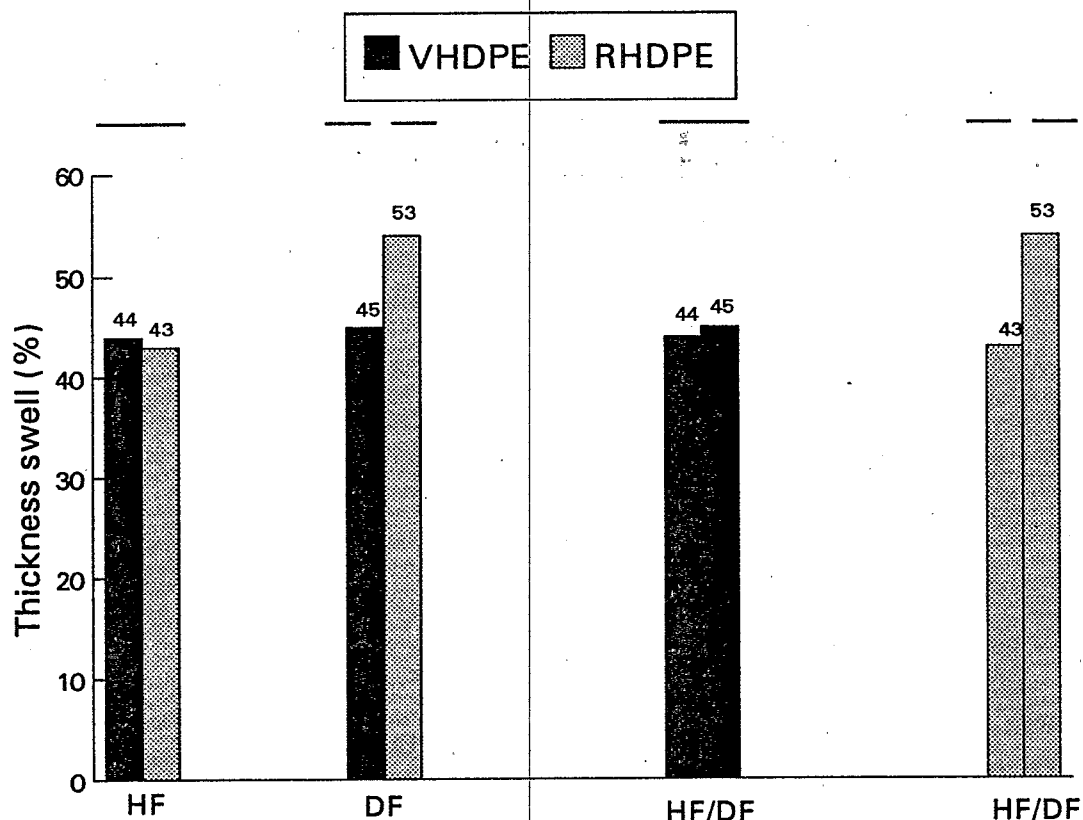


Figure 17. Task 3, AL Series 2. Thickness swell as a function of virgin HF and recycled DF and HDPE.

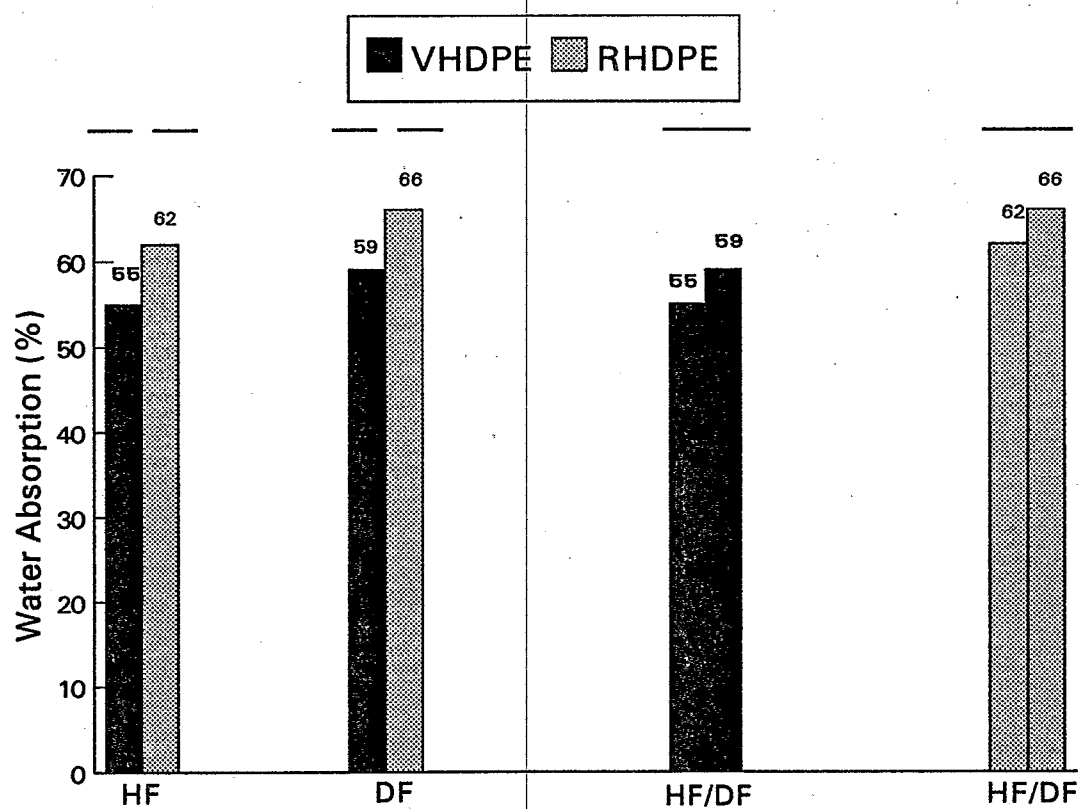


Figure 18. Task 3, AL Series 2. Water absorption as a function of virgin HF and recycled DF and HDPE.

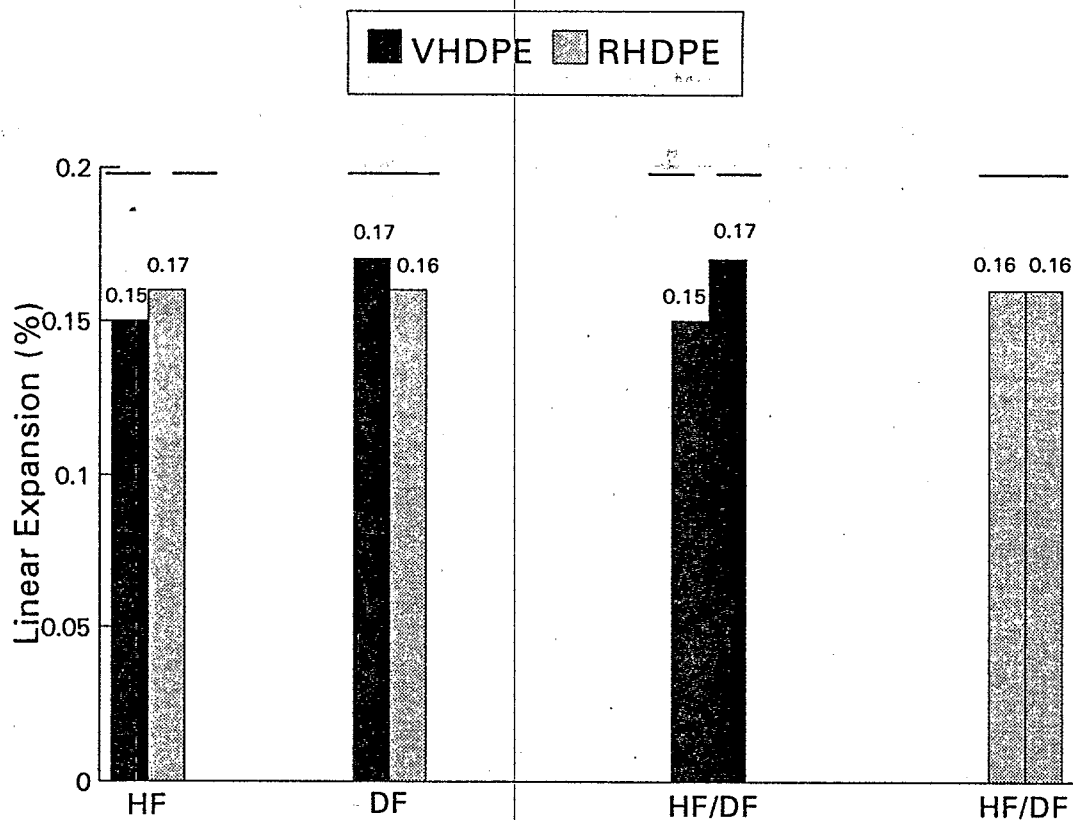


Figure 19. Task 3, AL Series 2. Linear expansion at 30 percent RH as a function of virgin HF and recycled DF and HDPE.

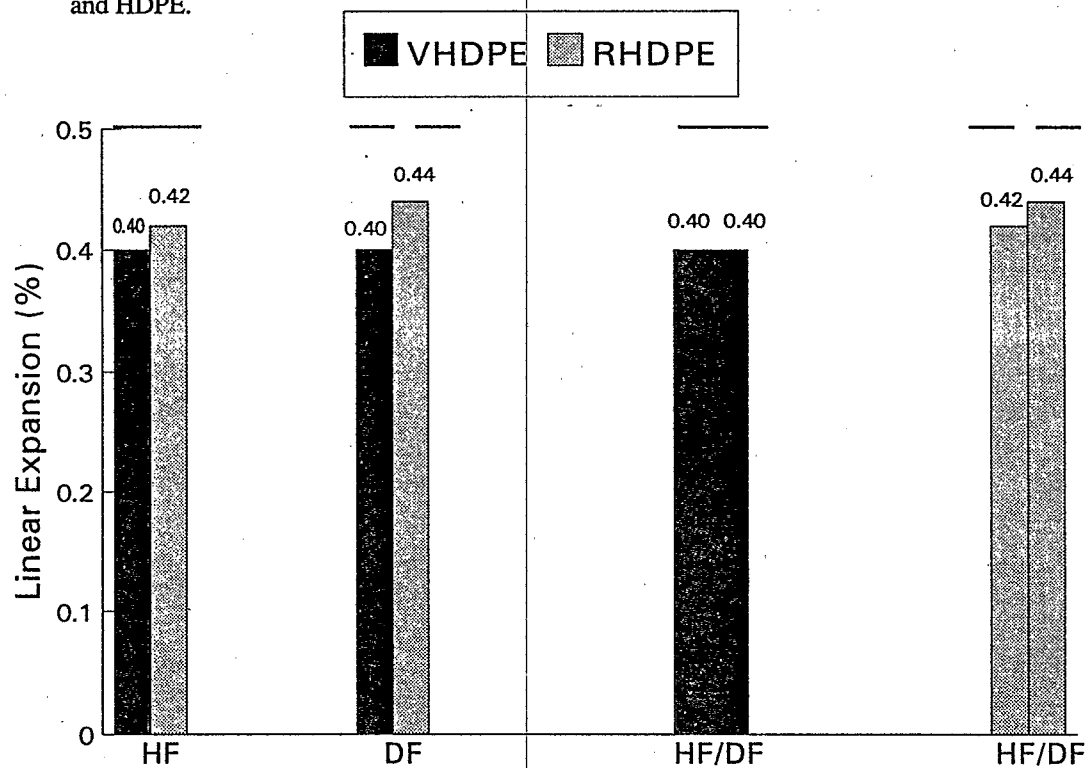


Figure 20. Task 3, AL Series 2. Linear expansion at 65 percent RH as a function of virgin HF and recycled DF and HDPE.

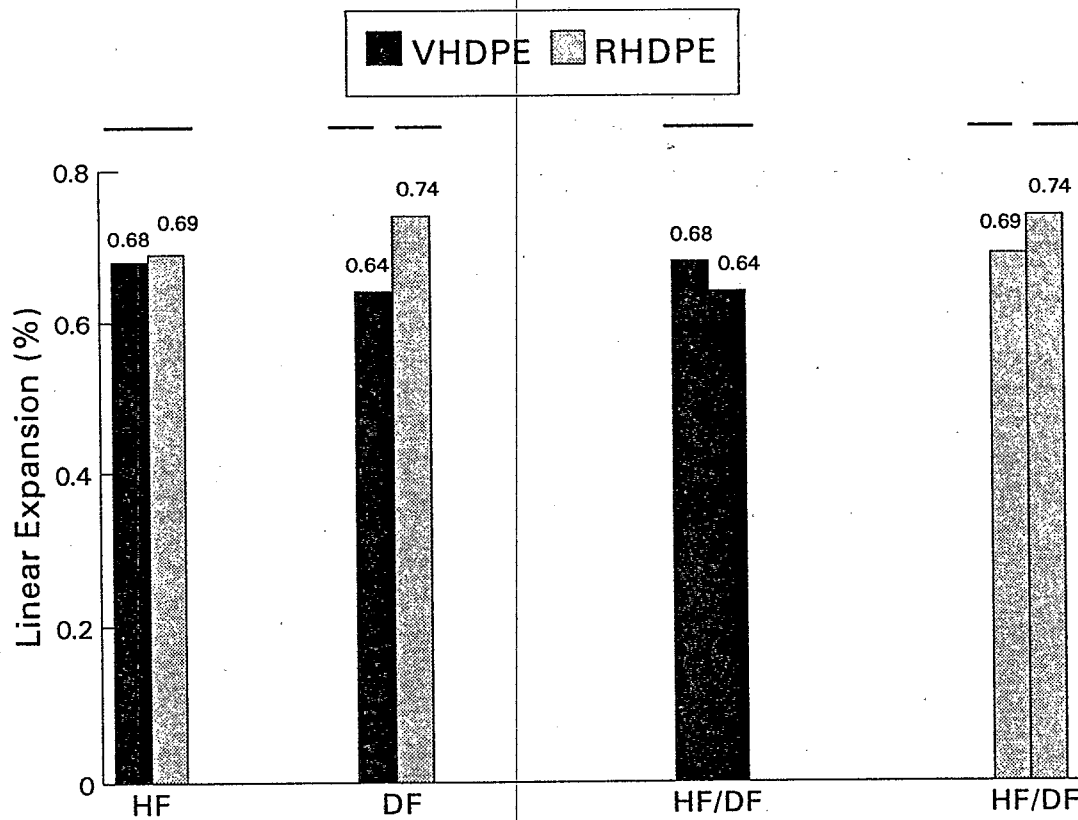


Figure 21. Task 3, AL Series 2. Linear expansion at 90 percent RH as a function of virgin HF and recycled DF and HDPE.

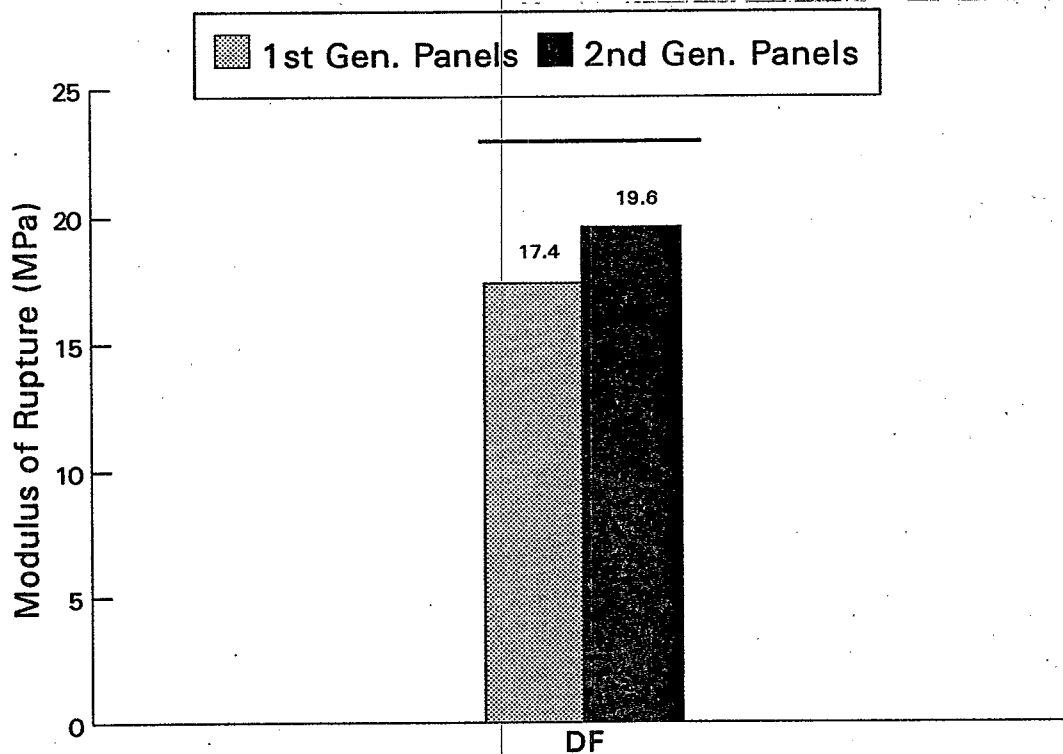


Figure 22. Task 4, Recyclability. Bending strength (MOR) as a function of recycled DF and HDPE and first-generation panels.

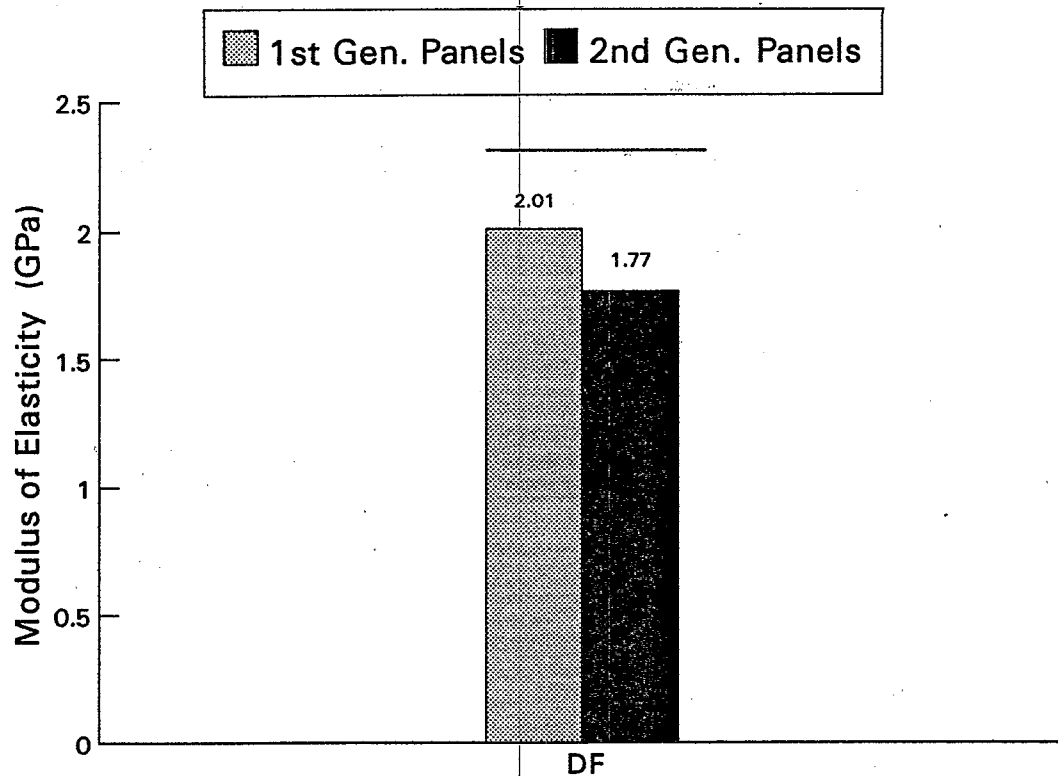


Figure 23. Task 4, Recyclability. Bending stiffness (MOE) as a function of recycled DF and HDPE and first-generation panels.

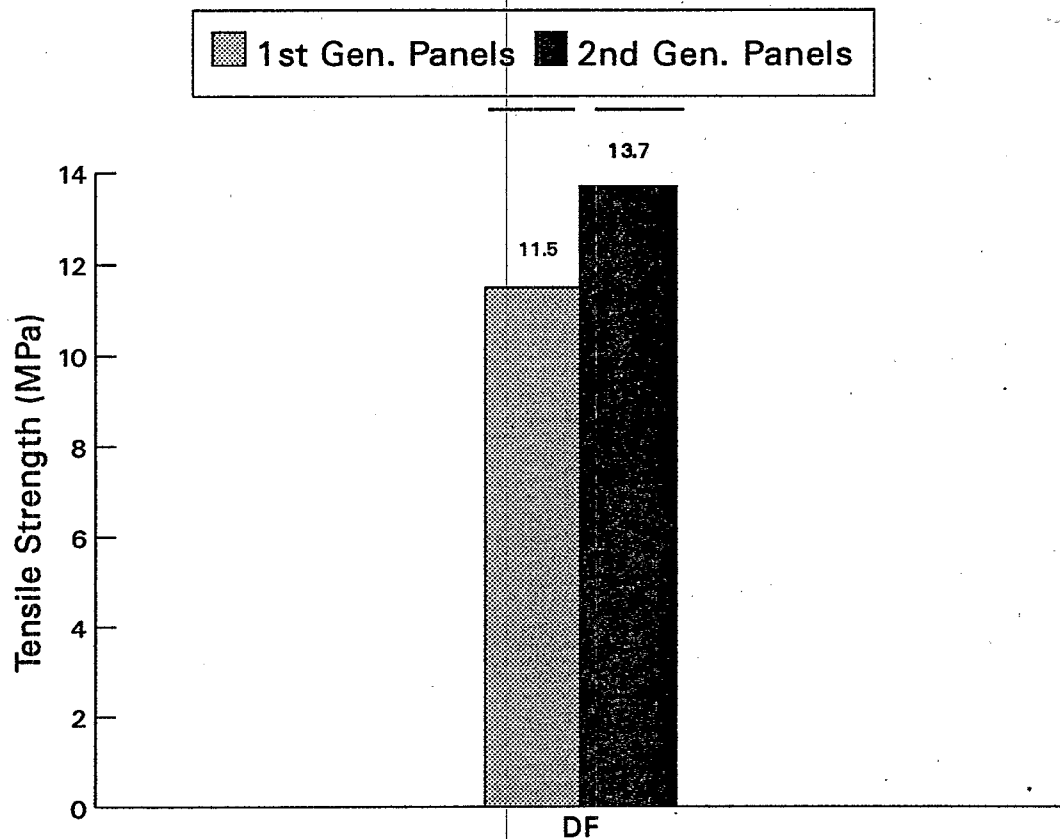


Figure 24. Task 4, Recyclability. Tensile strength as a function of recycled DF and HDPE and first-generation panels.

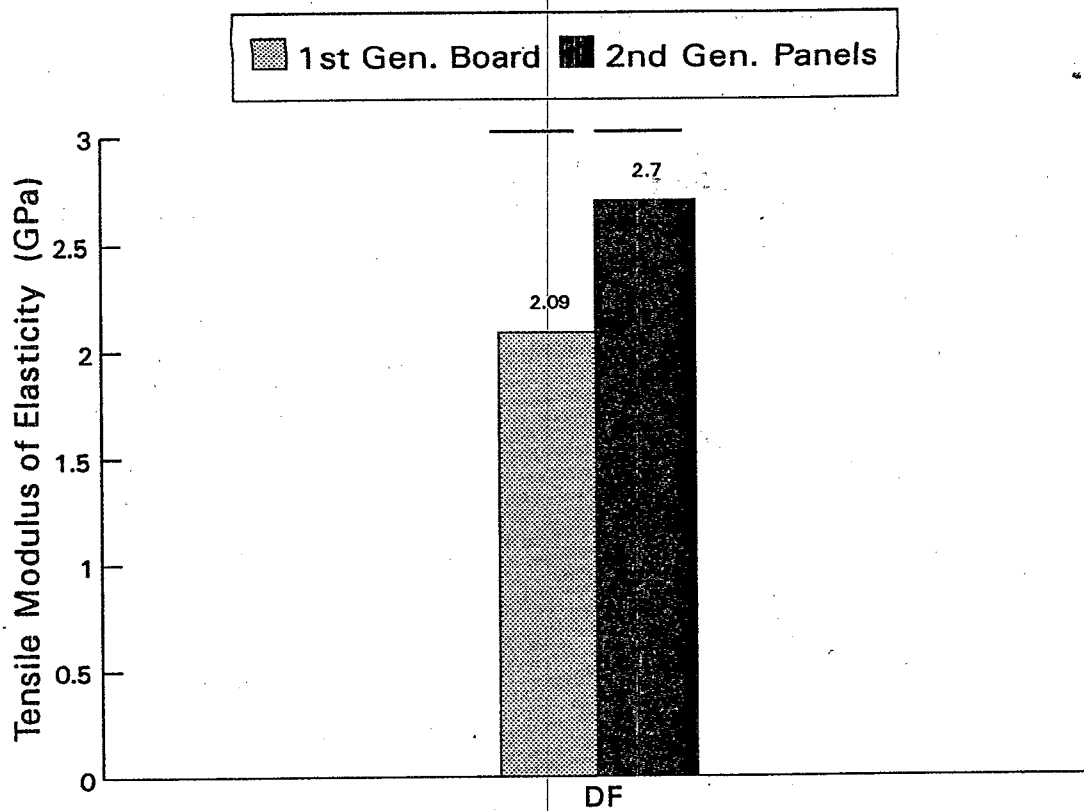


Figure 25. Task 4, Recyclability. Tensile modulus (MOE) as a function of recycled DF and HDPE and first-generation panels.

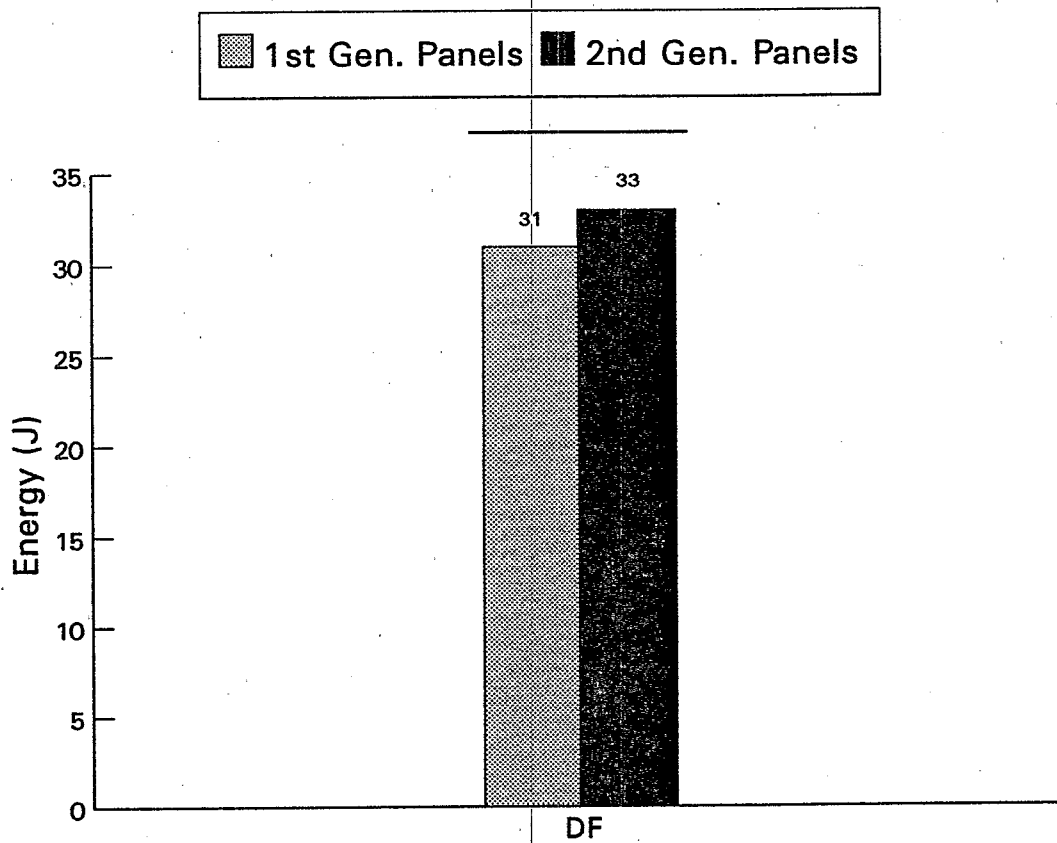


Figure 26. Task 4, Recyclability. Impact energy as a function of recycled DF and HDPE and first-generation panels.

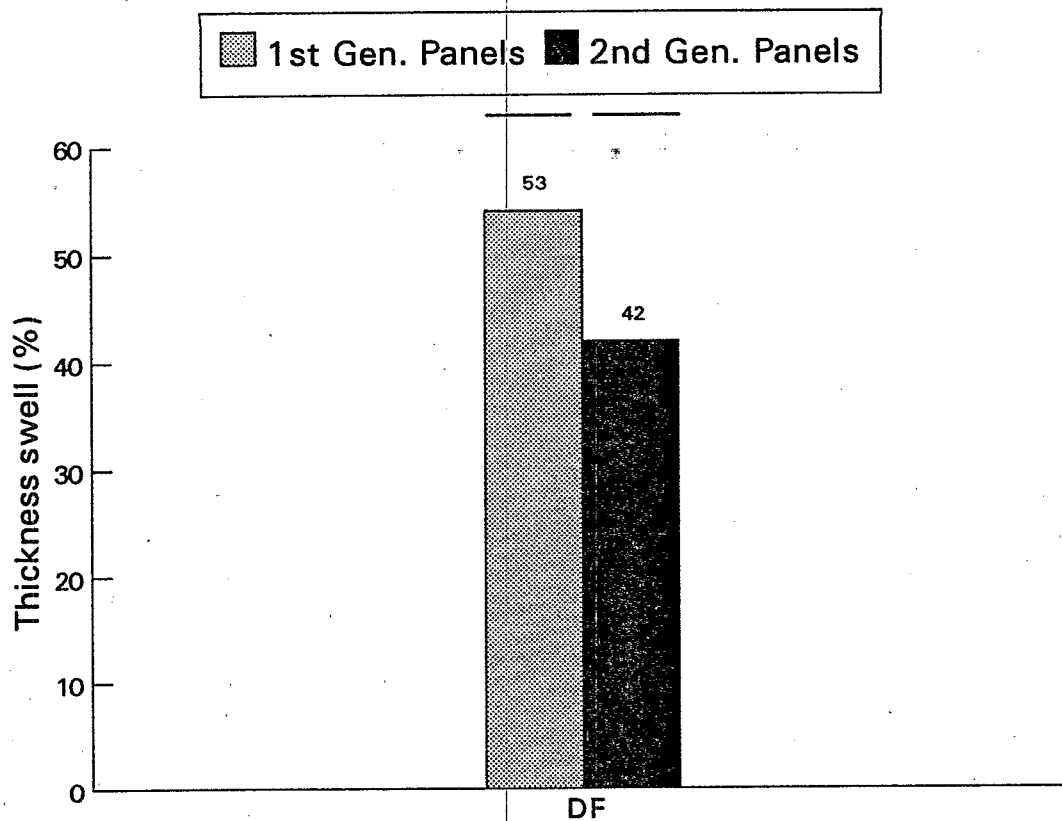


Figure 27. Task 4, Recyclability. Thickness swell as a function of recycled DF and HDPE and first-generation panels.

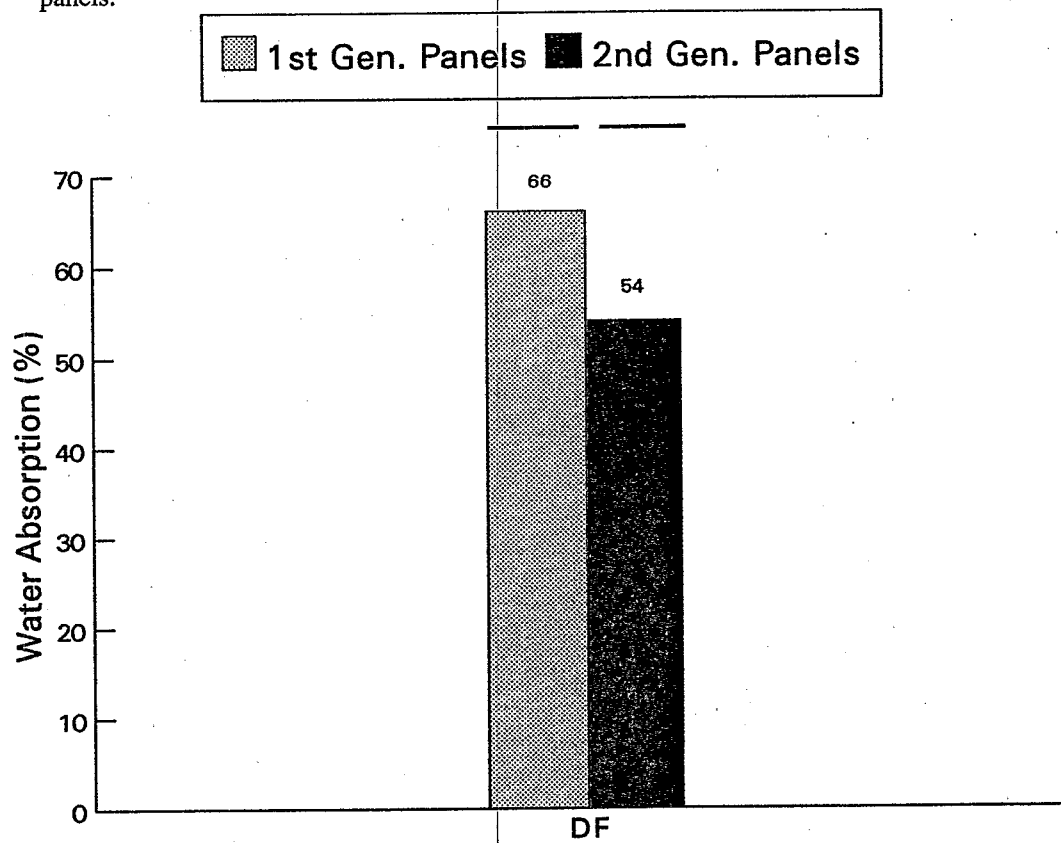


Figure 28. Task 4, Recyclability. Water absorption as a function of recycled DF and HDPE and first-generation panels.

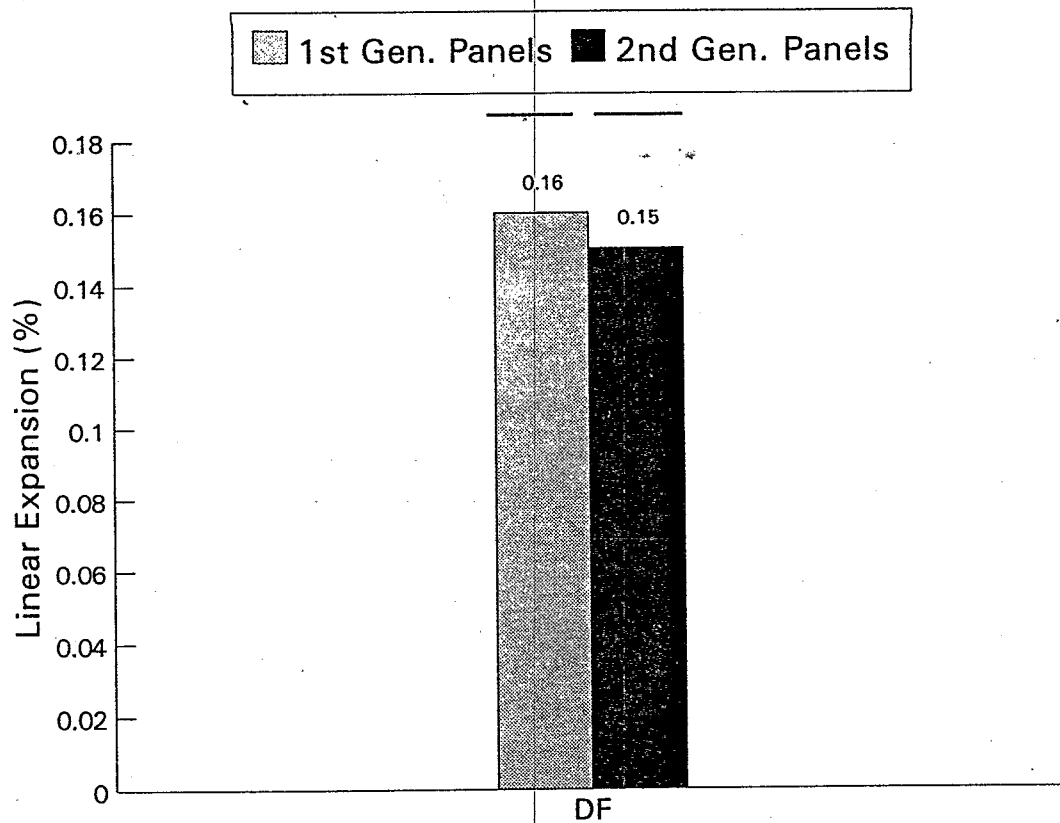


Figure 29. Task 4, Recyclability. Linear expansion at 30 percent RH as a function of recycled DF and HDPE and first-generation panels.

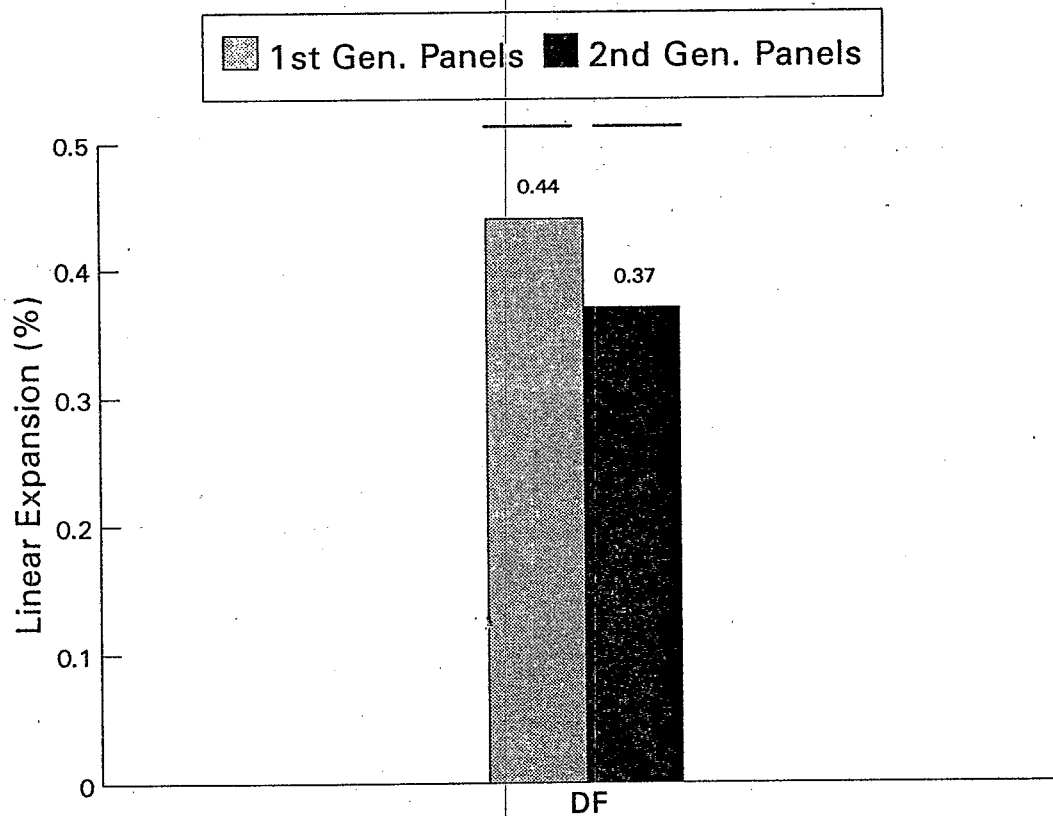


Figure 30. Task 4, Recyclability. Linear expansion at 65 percent RH as a function of recycled DF and HDPE and first-generation panels.

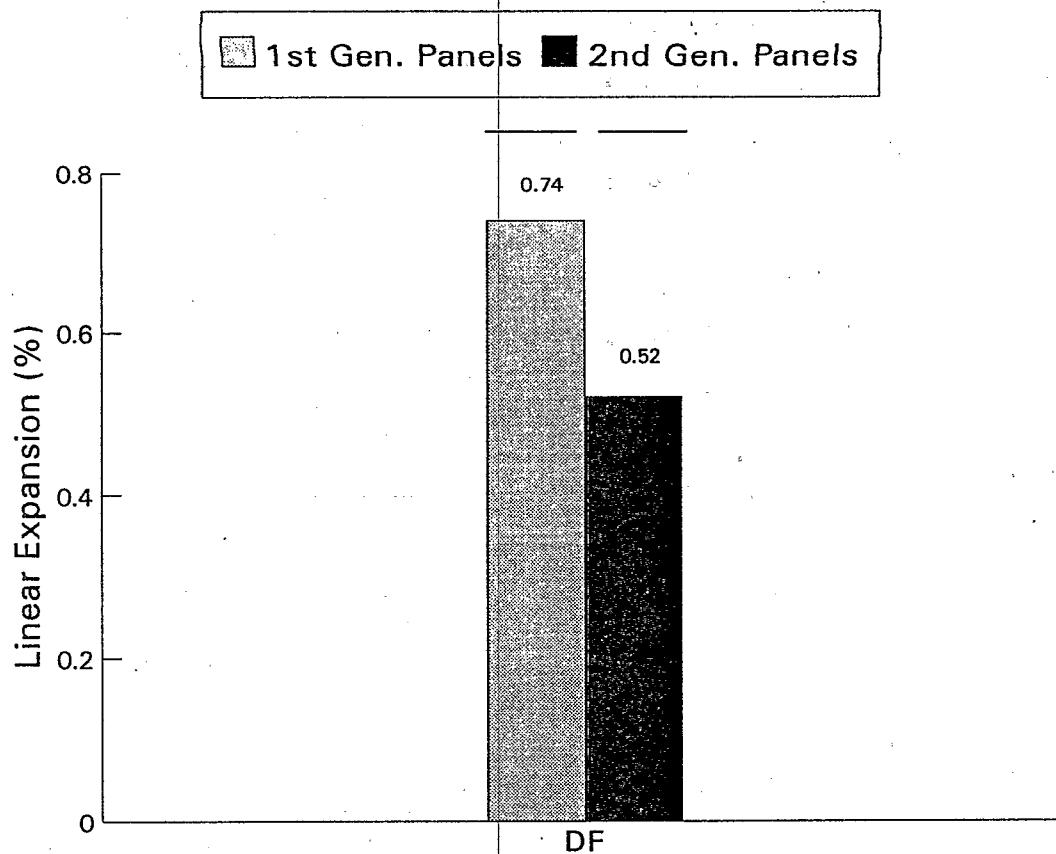


Figure 31. Task 4, Recyclability. Linear expansion at 90 percent RH as a function of recycled DF and HDPE and first-generation panels.

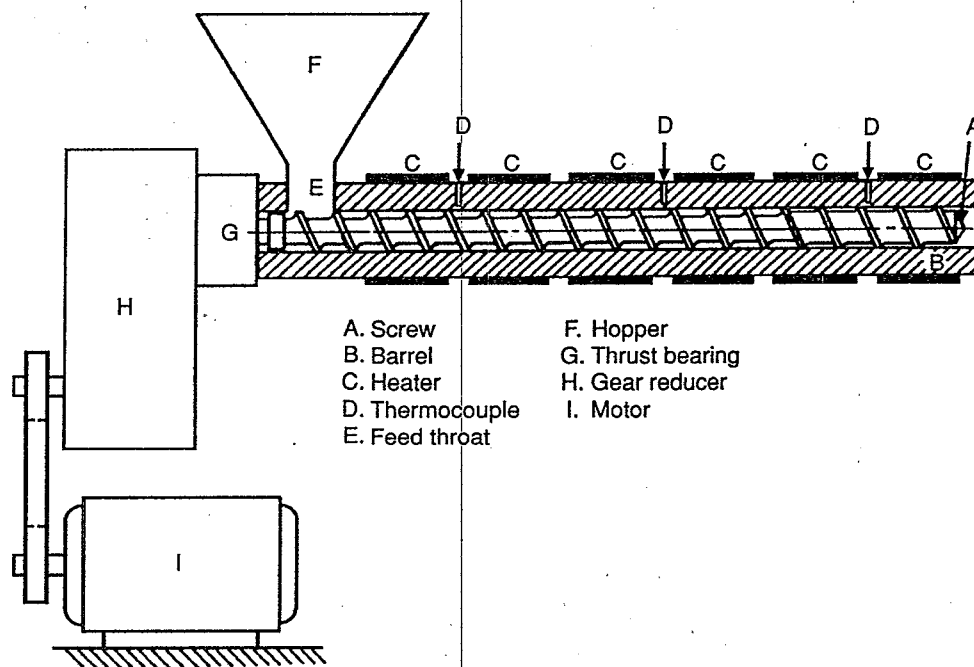


Figure 32. Schematic of single-screw extruder.

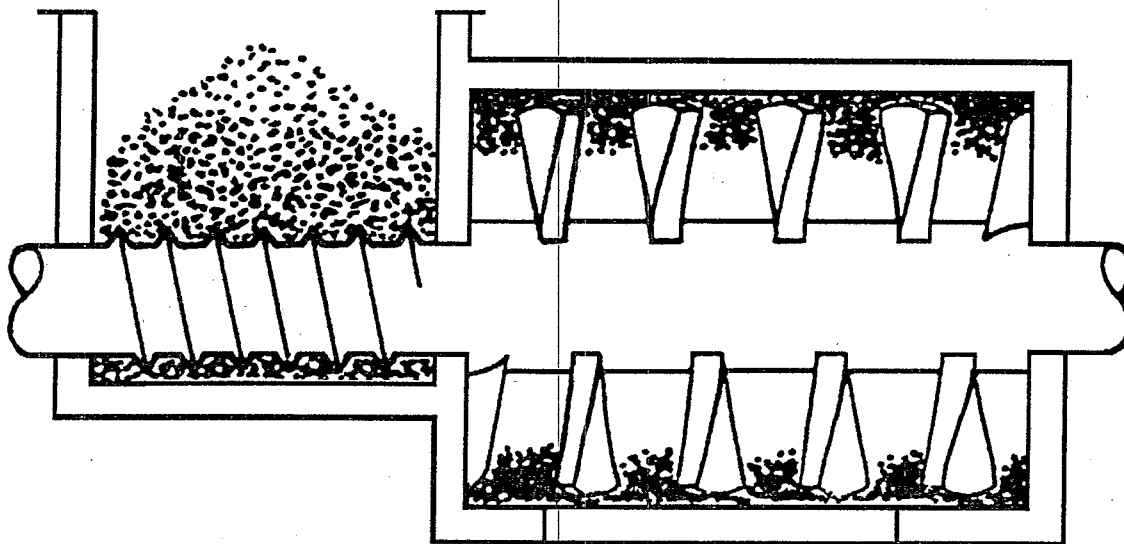


Figure 33. Schematic of K-mixer.

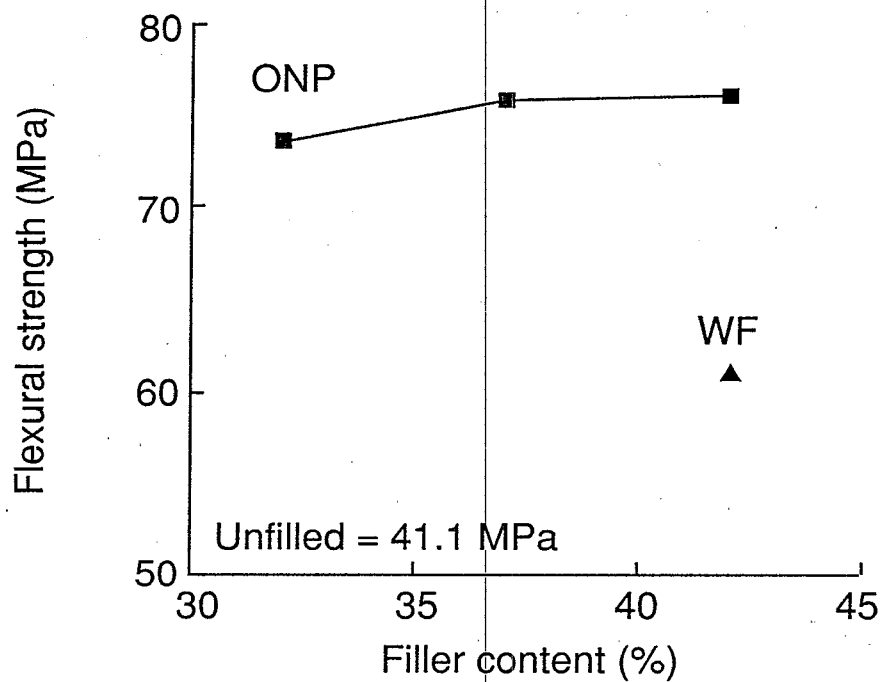


Figure 34. Study MB-1. Flexural strength as a function of filler content and type.

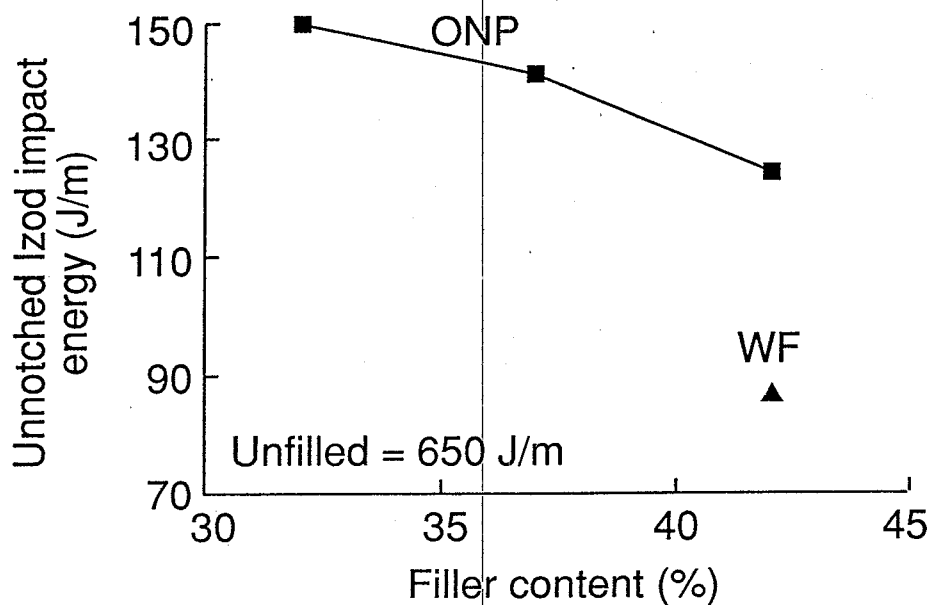


Figure 35. Study MB-1. Unnotched impact energy as a function of filler content and type.

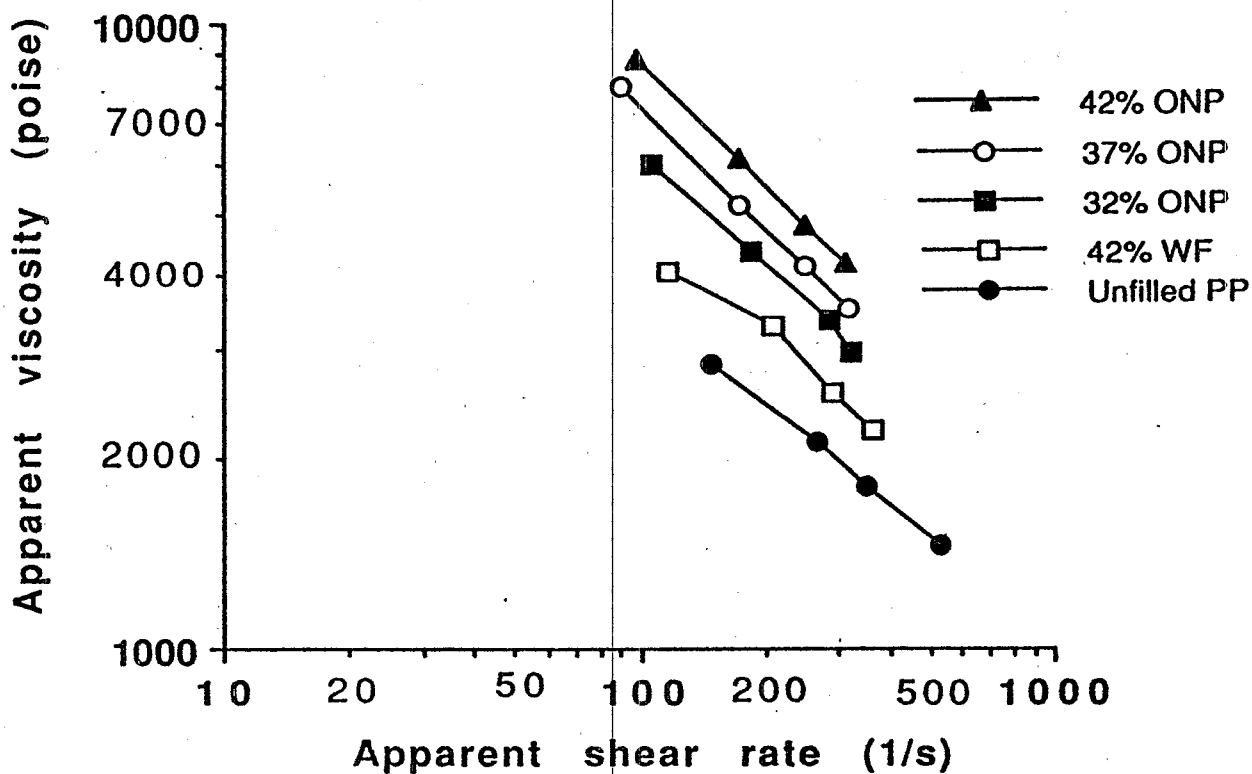


Figure 36. Study MB-1. Apparent melt viscosity of composite blends. ONP is old newspapers; WF, wood flour; and PP, polypropylene.

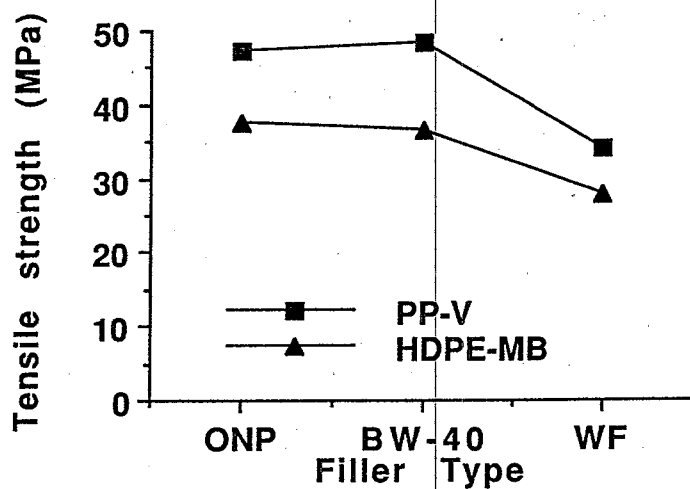


Figure 37. Study MB-2. Tensile strength as a function of filler and polymer types. PP-V is virgin polypropylene; HDPE-MB, melt-blended high density polyethylene.

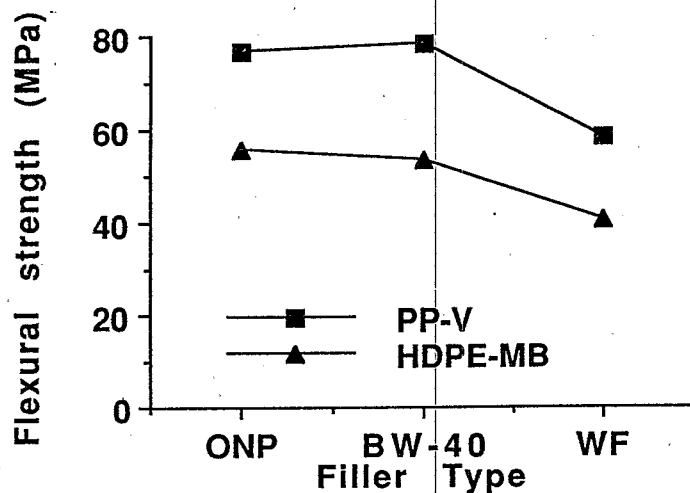


Figure 38. Study MB-2. Flexural strength as a function of filler and polymer types.

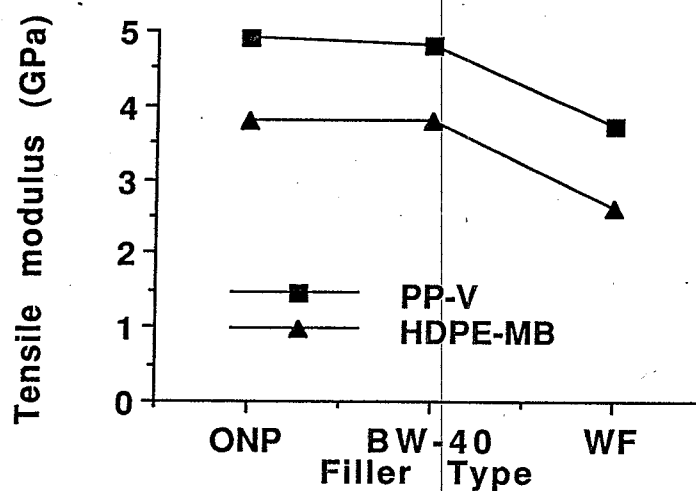


Figure 39. Study MB-2. Tensile modulus as a function of filler and polymer types.

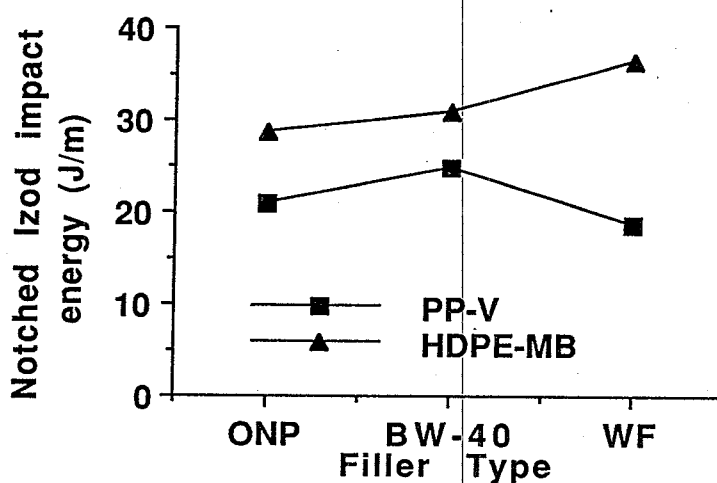


Figure 40. Study MB-2. Notched impact energy as a function of filler and polymer types.

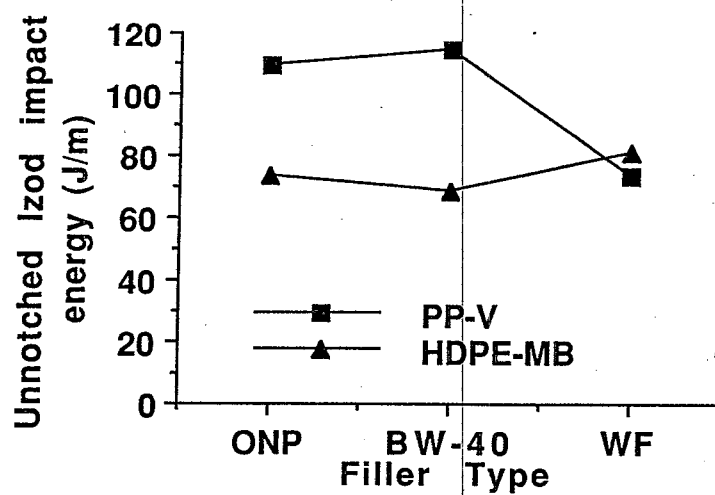


Figure 41. Study MB-2. Unnotched impact energy as a function of filler and polymer types.

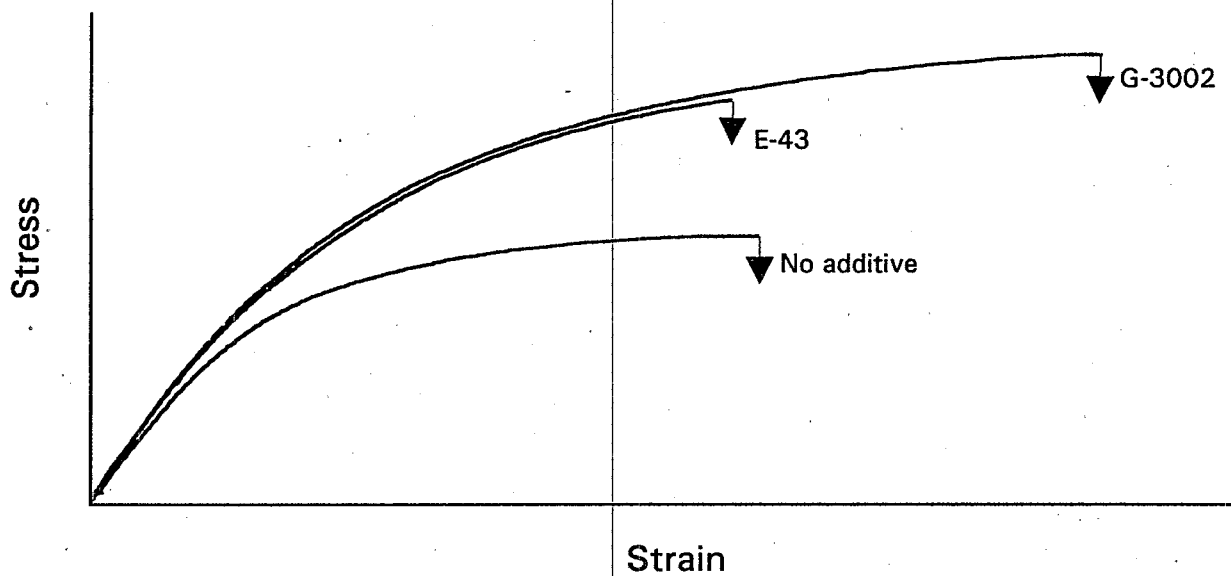


Figure 42. Study MB-3. Effect of coupling agent on tensile stress-strain curve of ONP/PP composites.

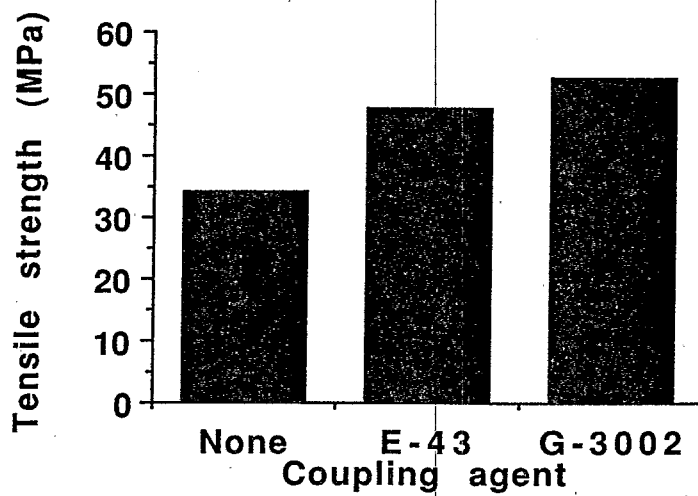


Figure 43. Study MB-3. Effect of coupling agent on tensile strength of ONP/PP composites.

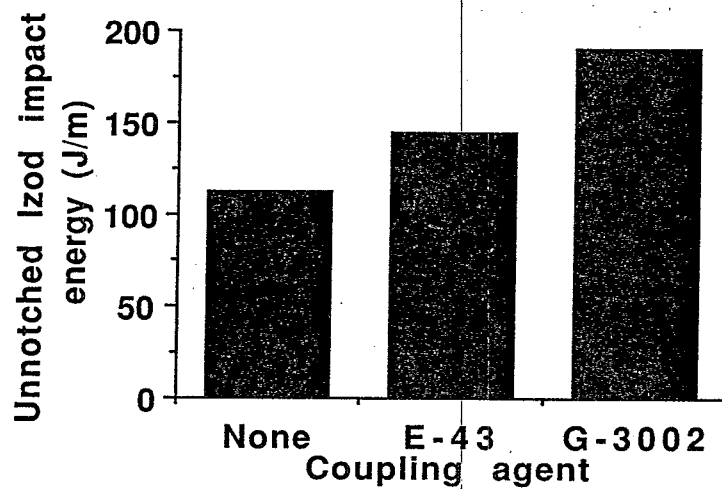


Figure 44. Study MB-3. Effect of coupling agent on impact energy of ONP/PP composites.

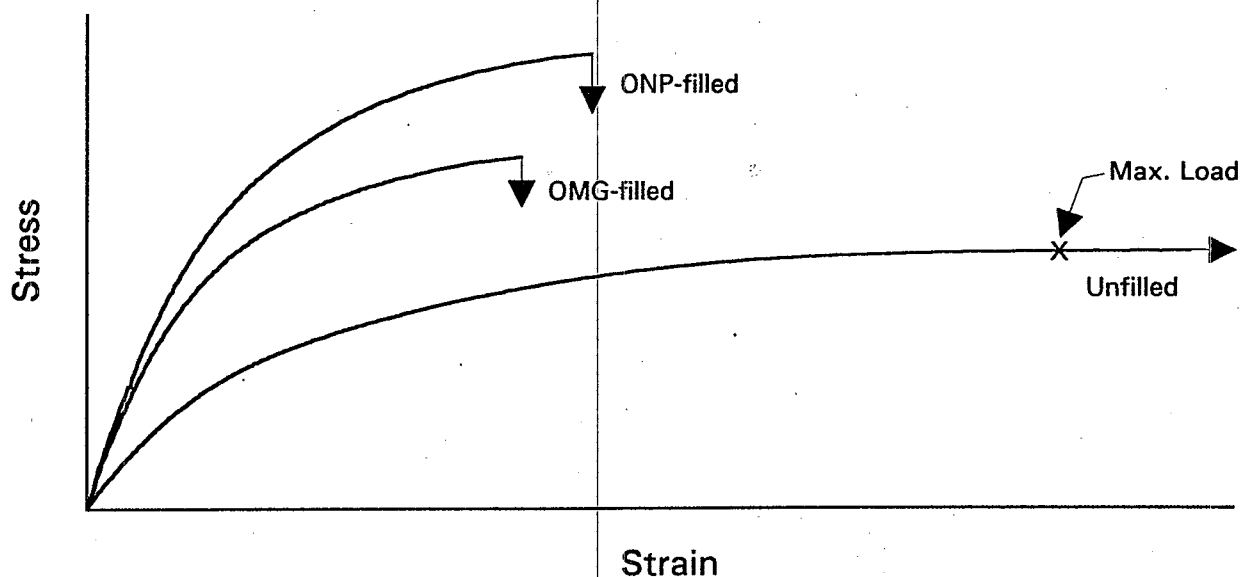


Figure 45. Study MB-4. Effect of wastepaper on tensile stress-strain curve of polypropylene from recycled auto battery cases (BPP). ONP is old newspapers; OMG, old magazines.

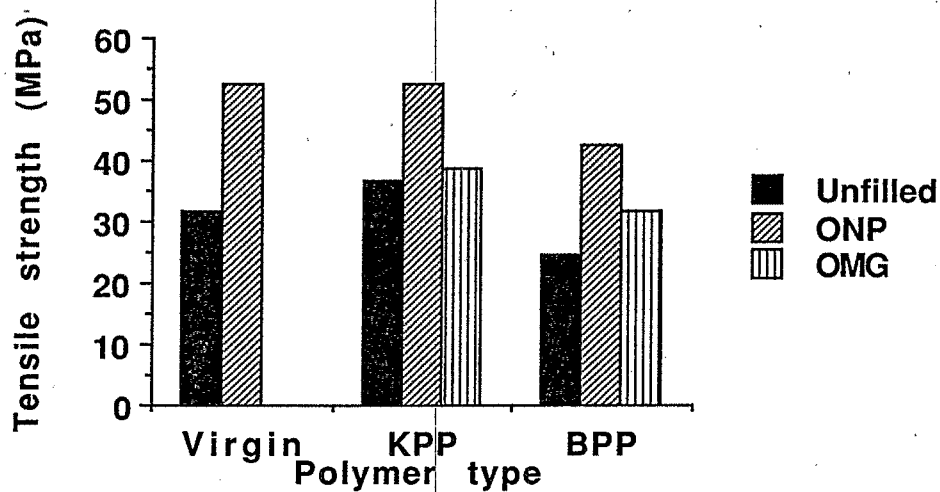


Figure 46. Study MB-4. Effect of polymer and waste paper types on tensile strength. KPP is recycled polypropylene from ketchup bottles.

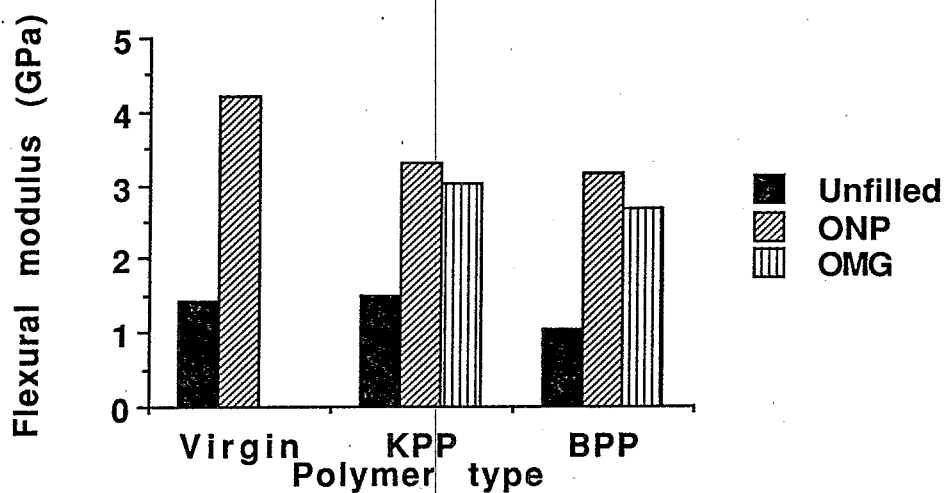


Figure 47. Study MB-4. Effect of polymer and wastepaper types on flexural modulus.

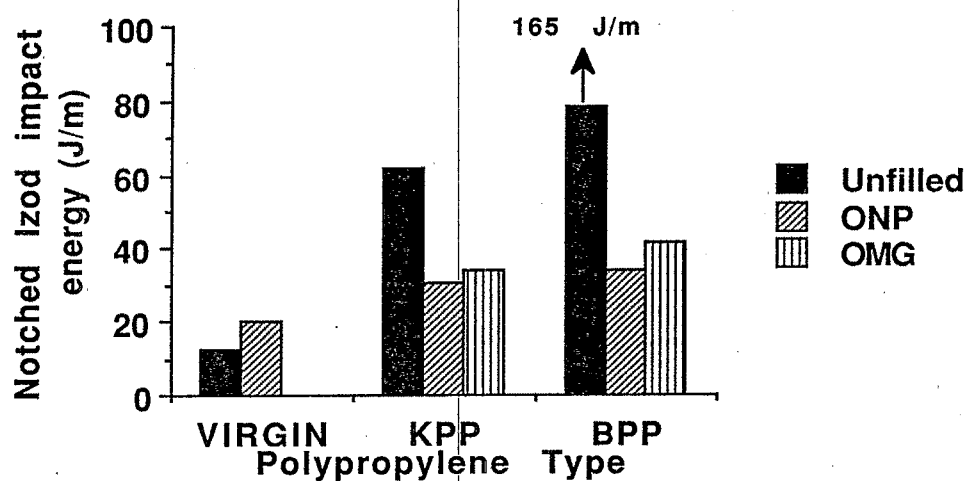


Figure 48. Study MB-4. Effect of polymer and wastepaper types on notched impact energy.

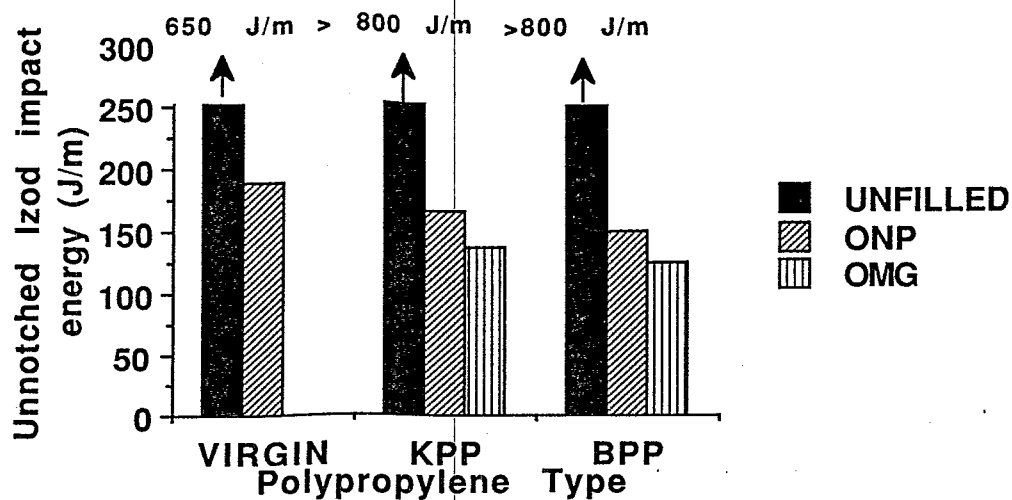


Figure 49. Study MB-4. Effect of polymer and wastepaper types on unnotched impact energy.

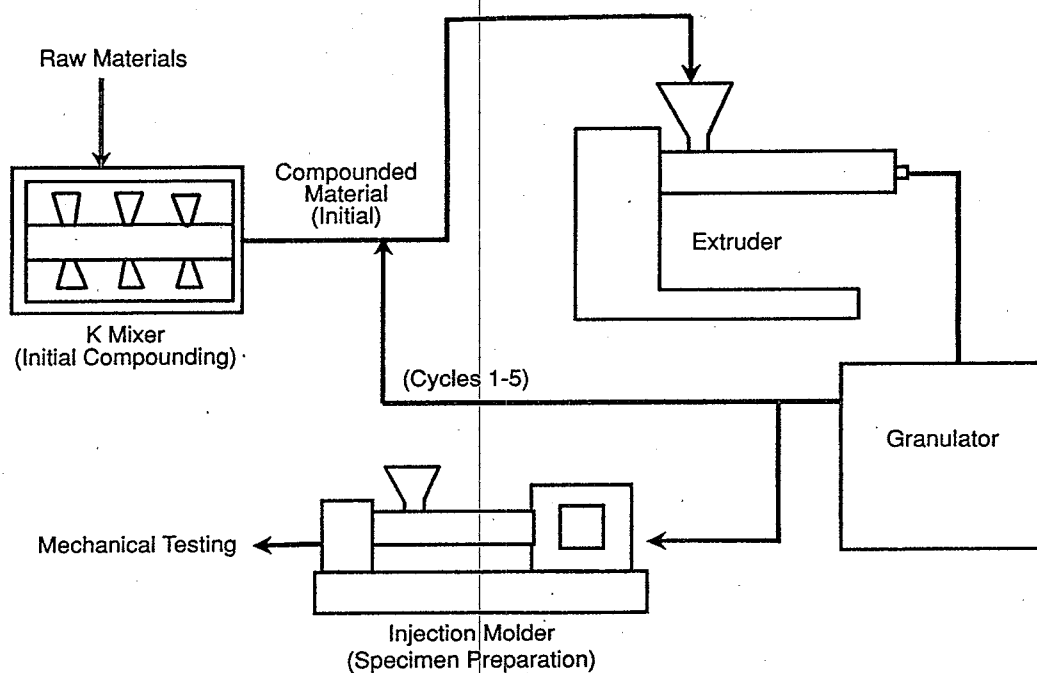


Figure 50. Study MB-5. Processing flow diagram for recycling study.

PROJECT SUMMARY

COMPOSITES FROM RECYCLED WOOD AND PLASTICS

John A. Youngquist, George E. Myers,
James H. Muehl, Andrzej M. Krzysik, and Craig M. Clemons

USDA Forest Service
Forest Products Laboratory
Madison, WI 53705-2398

Lisa Brown, Project Officer

Waste Minimization, Destruction, and Disposal Division
Risk Reduction Engineering Laboratory
Cincinnati, OH 45268

ABSTRACT

The ultimate goal of our research was to develop technology to convert recycled wood fiber and plastics into durable products that are recyclable and otherwise environmentally friendly. Two processing technologies were used to prepare wood-plastic composites: air-laying and melt-blending. Research was conducted in (1) developing laboratory methods for converting waste wood, wastepaper, and waste plastics into forms suitable for processing into composites; (2) optimizing laboratory methods for making composite panels from the waste materials; (3) establishing a database on the effects of formulation and bonding agent on physical and mechanical properties of composites; (4) establishing the extent to which the composites can be recycled without unacceptable loss in properties; and (5) reaching out to industry to provide education, to develop applications, and to extend the database. Overall, the program demonstrated that both air-laid and melt-blended composites can be made from a variety of waste wood, wastepaper, and waste plastics. The composites exhibit a broad range of properties that should make them useful in a wide variety of commercial applications. For air-laid composites, the waste materials were demolition wood waste and waste plastics from milk bottles (polyethylene) and beverage bottles (polyethylene terephthalate). Results showed that air-laid composites made from these waste ingredients possessed properties very similar to those of composites made from the virgin ingredients. In addition, air-laid

composites containing 20% reground panels possessed some properties that were superior to those of the original composites. For melt-blended composites, waste materials were wastepaper, polyethylene from milk bottles, and polypropylene from automobile battery cases or ketchup bottles. Waste magazines were slightly inferior to waste newspapers as a reinforcing filler; the properties of composites made from waste newspaper were better than those of composites made from wood flour, which is currently used in some commercial composites. Properties of wood-plastic composites were generally parallel to those of the plastics; thus, different balances in composite properties are possible from using waste plastic. Outreach activities included the organization and presentation of two international conferences on wood fiber-plastic composites, presentations at many conferences, publication of several papers, and several spin-off cooperative studies with industry. One major study with industry demonstrated the commercial feasibility of making melt-blended composites from old newspapers and polypropylene.

STUDIES ON AIR-LAID COMPOSITES

Air-laid (AL) web composites provide options for balancing performance properties and costs, depending upon the application under consideration. However, poor attraction and low interfacial bonding between the hydrophilic wood and hydrophobic polyolefin limit the reinforcement imparted to the plastic matrix by the wood component. We compared the mechanical and dimensional stability properties of flat panel composites made from virgin and postconsumer waste wood and plastics in two series of tests:

AL Series 1. PET systems

- Virgin hemlock wood fiber (HF), virgin polyester fiber (VPET), phenolic resin
- HF, recycled PET (RPET), phenolic resin
- Demolition waste wood fiber (DF), VPET, phenolic resin
- DF, RPET, phenolic resin

AL Series 2. HDPE systems

- HF and virgin high-density polyethylene (VHDPE)
- HF and recycled HDPE (RHDPE)
- DF and VHDPE
- DF and RHDPE

Proportions of components were based on oven-dry fiber weight. For AL Series 1, the proportions were 80% wood fiber/10% PET/10% phenolic resin. For AL Series 2, the proportions were 60% wood fiber/30% HDPE/5% PET/5% tackifier (E-10).

We also studied the recyclability of air-laid composites by testing the mechanical properties of second-generation panels made from AL Series 2 panels. Two formulations were tested. Each formulation consisted of 30% RHDPE, 5% VPET, and 5% E-10 tackifier. In addition, one formulation had 60% DF and no refiberized first-generation panels; the other formulation had 40% DF and 20% refiberized first-generation panels.

Methods and Materials

Experiments consisted of the following sequence of steps: (a) modifying the FPL air-forming equipment to ensure that uniform machine- and cross-machine direction webs could be produced routinely; (b) converting raw materials into forms suitable for use in this equipment; (c) producing air-formed webs; (d) selecting and stacking the webs to produce mats of a given weight; (e) consolidating the mats in a platen press to produce test panels; (f) cutting test specimens from panels; and (g) testing properties.

For both test series, each data set was tested for normality at the 95% confidence level using Shapiro-Wilk statistical analysis. An analysis of variance was performed and the means were compared at the 95% confidence level using Tukey's method of multiple comparisons.

The raw materials studied in the air-forming portion of this research program fell into three general classes: cellulosic fibers, plastics, and additives.

Cellulosic Fibers

Two basic types of wood fiber were used in the AL series of tests. The first was virgin western hemlock wood fiber (HF), which was produced in a pressurized single-disk refiner from 100% pulp-grade chips. The second was demolition wood fiber (DF), which was derived from waste wood from buildings that had been torn down in the Boston, Massachusetts, area.

Plastics

The virgin polyester (VPET) was 5.5 denier (6.1×10^{-7} kg/m), 38 mm long, and crimped, and had a softening temperature greater than 215°C. The recycled polyester fiber, which was spun from recycled soft drink containers, was 6.00 denier, 51 mm long, and crimped. For all the air-laid experiments, VPET served as a matrix to hold the fibers together within the mat.

The virgin high-density polyethylene (VHDPE) was a blow-molding polymer normally used as a feedstock for plastic milk bottles. The flakes were cryogenically ground to a (-)35 mesh size. The melt flow index of the recycled HDPE was 0.7.

Additives

Liquid phenolic resin was used as the binder for the AL Series 1 panels; it had a solids content of 51% to 53% and a pH of 9.5 to 10.0 at 25°C. The resin was sprayed on the wood fiber at 25°C at a level of 10% solids by weight as it rotated in a drum-type blender.

For the AL Series 2 boards, the wood fiber was blended with granulated HDPE. Previous work had showed that a tackifier was needed to retain the granule HDPE in the web during the web formation process. Preliminary testing had also indicated that the tackifier, a wax emulsion of oxidized low molecular weight polyethylene (E-10), did not have an adverse effect on the properties of the resultant test panels. The tackifier was applied to the wood fibers in a rotating drum blender with an air spray gun in a manner similar to the application of phenolic resin in the AL Series 1 studies.

Equipment Modifications and Additions

A 305-mm-wide, laboratory-scale Rando-Webber forming machine was used to make nonwoven mats for the air-laid composites. The equipment was modified to minimize the density gradient across the web, a problem encountered in preliminary experiments.

Panel Fabrication

Nonwoven webs were weighed, sorted, and stacked on the basis of weight and specific gravity. A steam-heated platen press was used to press the panels to a thickness of 3.2 mm and a specific gravity of 1.0. A cooling cycle was used to maintain target thickness.

The recyclability part of the study required that we determine the feasibility of recycling panels made for AL Series 2, which contained DF and RHDPE. We found

that we needed to recycle the boards through the pressurized refiner to be able to produce the desired fiber length and bundles.

Tests on Mechanical and Physical Properties

We evaluated the performance of panels made for AL Series 1 (PET systems), AL Series 2 (HDPE systems), and recyclability.

Results of Tests on PET Systems (AL Series 1)

In general, the mechanical, water resistance, and dimensional stability properties of panels made from recycled materials were equivalent to similar properties obtained from panels containing all virgin or virgin/recycled materials. Therefore, the recycled ingredients tested in AL Series 1 could replace virgin materials with minor consequences.

Mechanical Properties

Panels made with the HF/VPET formulation had the highest bending MOR value (50.6 MPa), although no statistically significant differences were observed for MOR values for either wood fiber or PET variations. The modulus of elasticity (MOE) values followed a different pattern. In both the HF and DF groups, the MOE values of boards containing RPET were significantly higher than those of boards containing VPET; total average increase for this property was 16% for both groups.

For the HF/VPET formulation, tensile strength was 33.0 MPa; tensile strength decreased by 14% for the HF/RPET formulation. However, when RPET or VPET fibers were used with DF fibers, no significant differences were noted. In contrast to tensile strength, the incorporation of RPET fibers increased tensile modulus (MOE) by 6% and 7% for HF and DF formulations, respectively, although these differences were not statistically significant.

Impact energy of specimens from the HF and DF formulations showed a consistent trend. Impact strength was respectively 20% and 10% higher for HF and DF formulations containing VPET fibers compared to formulations containing RPET fibers.

Table 1. Results of water soak and linear expansion tests for AL series^a

Composite ^b	24-h water soak		Linear expansion (%)		
	Thickness swell (%)	Water absorption (%)	30% RH	65% RH	90% RH
AL Series 1					
HF-80% VPET-10% PR-10%	25.2 (9)	43.4 (20)	0.19 (10)	0.42 (4)	0.61 (6)
HF-80% RPET-10% PR-10%	22.3 (9)	41.3 (25)	0.21 (13)	0.44 (8)	0.70 (7)
DF-80% VPET-10% PR-10%	29.8 (14)	48.2 (25)	0.20 (11)	0.43 (6)	0.64 (7)
DF-80% RPET-10% PR-10%	26.9 (8)	44.1 (16)	0.20 (12)	0.45 (6)	0.71 (6)
AL Series 2					
HF-60% VHDPE-30% VPET-5% E10 wax-5%	43.8 (14)	54.9 (17)	0.15 (9)	0.39 (5)	0.68 (7)
HF-60% RHDPE-30% VPET-5% E10 wax-5%	42.7 (21)	61.8 (19)	0.17 (12)	0.42 (12)	0.69 (12)
DF-60% VHDPE-30% VPET-5% E10 wax-5%	45.2 (13)	58.7 (13)	0.17 (5)	0.40 (7)	0.64 (9)
DF-60% RHDPE-30% VPET-5% E10 wax-5%	52.8 (15)	65.8 (16)	0.16 (7)	0.44 (6)	0.74 (6)

^aValues in parentheses are coefficients of variation (%).

^bDF is demolition wood fiber; E10, epolene-maleated polyethylene; HF, hemlock fiber; VPET, virgin polyester fiber; RPET, recycled polyester; RHDPE, recycled high-density polyethylene; and VHDPE, virgin high-density polyethylene.

Physical and Dimensional Stability Properties

Thickness swell values were significantly different for HF/RPET and DF/VPET specimens (Table 1). The HF/RPET specimens had the lowest thickness swell value (22.3%), and the DF/VPET specimens the highest value (29.8%). Water absorption values were not significantly different.

Linear expansion values at 30% RH were statistically equivalent (Table 1). At 65% and 90% RH, the HF/RPET and DF/RPET formulations had slightly higher values and were statistically different from the HF/VPET and DF/VPET formulations.

Results of Tests on HDPE Systems (AL Series 2)

Panels containing virgin and recycled wood fiber/polyethylene had equivalent mechanical and physical properties. Therefore, as in AL Series 1, the recycled materials used in AL Series 2 could replace virgin materials with minor consequences.

Mechanical Properties

The DF/VHDPE panels had the highest bending MOR value at 19.1 MPa, followed by 18.7 MPa for the HF/RHDPE panels. Generally no statistically significant differences were observed for MOR values for either wood fiber or HDPE variations. In contrast, the HF/RHDPE panels had the highest bending MOE value (2.13 GPa) and the DF/VHDPE panels the lowest MOE value (1.75 GPa); however, these results were not statistically significant.

For tensile strength, the highest value (12.4 MPa) was observed for the DF/VHDPE formulation; tensile strength of the DF/RHDPE panels was 7% lower (11.5 MPa). For both wood fiber variations, the use of either virgin or recycled HDPE did not significantly influence tensile strength values. The HF/VHDPE panels had the highest tensile MOE (2.81 GPa); incorporation of RHDPE lowered tensile MOE by 21% (2.23 GPa), a significant change. Tensile MOE values of DF formulations were about equal, averaging 2.11 GPa.

Type of wood fiber and formulation did not significantly affect impact energy.

Physical and Dimensional Stability Properties

Thickness swelling of DF specimens was an average of 22% higher than that of HF specimens; the highest value (53%) was observed for the DF/RHDPE formulation (Table 1). Particularly notable is the fact that RHDPE had a significant effect on only the DF formulation and not the HF formulation. Thickness swelling was lowest in the HF/RHDPE panels (43%).

The formulation had a consistent influence on water absorption (Table 1). Incorporating RHDPE with either type of wood fiber produced a statistically significant

increase in this property (average 13% increase). The HF/VHDPE and DF/VHDPE formulations showed the lowest water absorption values.

Linear expansion values for all formulations at 30% RH ranged from 0.15 to 0.17% (Table 1). At 65% and 90% RH, the HF/RHDPE and DF/RHDPE formulations had slightly higher values.

Results of Tests on Recyclability

In general, the mechanical, water resistance, and dimensional stability properties of second-generation panels made from recycled materials were essentially equivalent to or better than properties obtained from first-generation panels. Therefore, the second-generation composites, or possibly higher generation composites, can be produced using recycled materials without the consequence of reduced property values.

Mechanical Properties

The MOR of second-generation panels was higher than that of first-generation panels (19.6 MPa compared to 17.4 MPa). On the other hand, the bending MOE values of first-generation panels were higher than that of second-generation panels (2.01 vs. 1.77 GPa). These differences were not statistically significant. The tensile strength of second-generation panels was 19% higher than that of first-generation panels. Similar results were obtained for tensile modulus. The higher properties of the second-generation panels indicate that wood fiber/RHDPE composites can benefit by the addition of refiberized material from first-generation panels.

Impact energy values of specimens made from first-generation panels and second-generation panels were nearly equal and not statistically different.

Physical and Dimensional Stability Properties

In the 24-h water-soak tests, first-generation panels showed 53% thickness swell (Table 2). Incorporating first-generation panel fibers into second-generation panels improved this property by 21%. Similar trends were observed for water absorption values (Table 2). Water absorption of second-generation panels was 18% lower than that of first-generation specimens. These differences were statistically significant. The results suggest that the additional HDPE from refiberized first-generation panels further encapsulated the wood fibers, thus limiting water uptake by the wood fibers.

Linear expansion values were similar for both formulations at 30% RH, although the differences were statistically significant (Table 2). The incorporation of recycled panels into second-generation panels significantly decreased linear expansion at both 65% and 90% RH.

The positive influence of incorporating 20% first-generation panels into the second-generation panels may be the result of several factors. The incorporation of 20% refiberized first-generation panels reduced the percentage of wood fiber (DF) from 60% to 40%. The actual amount of total wood fiber was reduced from 60% to 52%, and the total amount of RHDPE was increased from 30% to 36%. Likewise, the percentage of PET and E-10 each was increased by 1%. The increase of these components, particularly the HDPE, and the decrease of the wood fiber may be a direct cause for some improvements in property values. More wood fiber was able to be encapsulated by plastic, thereby reducing exposure of the wood to moisture.

Table 2. Results of water soak and linear expansion tests on recyclability specimens^a

Composite	24-h water soak		Linear expansion (%)		
	Thickness swell (%)	Water absorption (%)	30% RH	65% RH	90% RH
First-generation panels ^b	52.8 (15)	65.8 (16)	0.16 (7)	0.44 (6)	0.74 (6)
Second-generation panels ^c	42.0 (12)	54.3 (12)	0.15 (11)	0.37 (9)	0.52 (11)

^aValues in parentheses are coefficients of variation (%).

^bDF-60%, RHDPE-30%, VPET-5%, E10 wax-5%. DF is demolition wood fiber; RHDPE, recycled high-density polyethylene; VPET, virgin polyester fiber; and E10, epolene-maleated polyethylene.

^cDF-40%, recycled panel-20%, RHDPE-30%, VPET-5%, and E10 wax-5%

Conclusions

- The Rando-Webber air-forming equipment can be adapted to handle both long and short synthetic and natural fibers as well as powder. Nonwoven air-laid webs can be produced that have excellent uniformity in both the machine- and cross-machine directions.
- Recycled and granulated HDPE can be used in the FPL air-forming equipment to produce an air-laid web that can be subsequently made into flat panels or shaped sections.

- Pressure refining techniques can convert postconsumer demolition wood or construction waste into fiber bundles that can be processed very successfully in the FPL air-forming equipment and subsequently pressed into flat panels or shaped sections.
- Panels made with recycled materials compare favorably to those made of virgin materials. Mechanical and physical properties of panels made with recycled polyester fiber or high-density polyethylene and demolition waste wood are similar to those of panels made with virgin materials.
- Second-generation composites, or possibly higher generation composites, can be produced using recycled materials without the consequence of reduced property values. Mechanical and physical properties of second-generation panels made from recycled materials were essentially equivalent to or better than properties obtained from first-generation panels.

STUDIES ON MELT-BLENDED COMPOSITES

Melt-blending is an inherently low cost, high production rate process in which wood and/or paper are mixed with molten plastic. These blends can then be formed into products using conventional plastics processing techniques such as extrusion and injection molding. The plastic acts as a means to convey the wood/paper during processing and the wood/paper fiber bears the load in the final composite, offering an effective balance between processability and strength of end product. With melt-blending techniques, wood fiber provides several advantages as reinforcement in thermoplastic composites. These include economy on a cost per unit volume basis, desirable aspect ratios, flexibility (hence less fiber breakage), and low abrasiveness to equipment. Composites can be produced containing up to 50 weight percent wood fiber and are low cost, thermoformable, and relatively insensitive to moisture.

Methods and Materials

In laboratory investigations of melt-blended composites, experimental operations generally proceed through the following sequence of steps: (a) conversion of raw materials into forms suitable for preparing dry mixtures quantitatively and feeding those mixtures into the melt-blending apparatus, (b) quantitative dry mixing, (c) melt-blending by either an extruder or a K-mixer, (d) injection molding of test specimens, and (e) measurement of properties.

Five major studies were undertaken to investigate the effects of a number of variables on mechanical and physical behavior of wood-fiber-reinforced thermoplastics. The studies were statistically designed and analyzed and all comparisons were made at a 95% confidence level. However, for the sake of brevity, the results will be presented as a whole, centering on the larger effects of the variables because of their greater impact on composite performance. Cellulosic fibers, plastics, and coupling agents are described in Table 3.

Table 3. Materials used in melt-blending studies

Material	Abbreviation	Description ^a	Source
Plastic			
Virgin polypropylene	VPP	Fortilene 9101, 1602, 1902; nominal MFI 3, 12, 30.	Solvay Polymers, Inc., Deer Park, TX
Recycled polypropylene from auto battery cases	BPP	Cleaned chips; nominal MFI 10	Gopher Smelting and Refining Co., Eagon, MN
Recycled polypropylene from ketchup bottles	KPP	Cleaned chips; nominal MFI 3	Wheaton Plastic Recycling Co., Milleville, NJ
Recycled polypropylene from milk bottles	HDPE-MB	Cleaned chips; nominal MFI 0.7	Recycle Worlds, Madison, WI
Cellulosic filler			
Wood flour	WF	Western pine; -40+80 mesh	American Woodfiber Co., Schofield, WI
Cellulose	BW40	Pure cellulose fiber; mean length 60 μ m	James River Corp., Hackensack, NJ
Waste newspaper	ONP	Over-production issue	Milwaukee Journal/Sentinel Inc., Milwaukee, WI
Waste magazine	OMG	Representative sample of Madison waste stream	Madison Recycling Center, Madison, WI
Coupling agent			
Epolene E43 powder	E43S	Powdered maleated polypropylene; M = 4,200	Eastman Chemical Co., Kingsport, TN
Epolene E43 emulsion	E43E	Emulsified potassium salt	Eastman Chemical Co.
Epolene G3002 powder	G3002	Powdered maleated polypropylene; M = 11,000	Eastman Chemical Co.

^aMFI is melt flow index. M is molecular weight.

Plastics

Plastics act as the matrix in the composite. In this program, a baseline matrix plastic, virgin polypropylene (VPP) homopolymer, was compared to three waste plastics: polypropylene from auto battery cases (BPP), recycled ketchup bottle polypropylene (KPP), and recycled high-density polyethylene from blow-molded milk bottles (HDPE-MB). These plastics were chosen on the basis of low melting temperature (necessary for use with wood/paper fiber), cost, performance, and availability.

Cellulosic Fibers

Celluloses act as the reinforcing filler in the composite. We used wood flour (WF) as the primary baseline filler because it is currently used commercially with polypropylene in extruded sheets for automobile interior panels. We included relatively expensive (several times the price of WF) pure cellulose fibers (BW40) as another baseline filler for comparing against recycled fibers—waste newspaper (ONP) and old magazines (OMG).

Both plastics and additives can be readily used in traditional plastics processing machinery. However, wood or paper fibers present difficulties during the melt-blending step. Cellulosic filler must be in a form that can be completely dispersed into the molten plastic by the shear forces exerted during melt-blending. Wood flour was readily disaggregated and fully dispersed into individual particles in the plastic using a simple laboratory single-screw extruder. Although difficult to disperse with an extruder, a usable blend of BW40 cellulose fibers was also obtained in this way. Wastepaper fibers were much more difficult to handle because of their low bulk density. For melt-blending in a high intensity K-mixer, the paper was milled or ground into approximately 4- to 8-mm flakes using a small granulator. For melt-blending in an extruder, the paper was first reduced into fibers by a small modified hammermill.

Coupling Agents

Additives aid the dispersion of filler into the matrix plastic and/or enhance the bonding (act as a "coupling agent") between filler and matrix. In this program, we restricted ourselves to Eastman Epolenes (E43, G3002)—maleated polypropylene "waxes" in which the small degree of maleation provides polar groups capable of bonding to the cellulose while the polypropylene segments, in theory, offer compatibility with the polypropylene matrix.

Mechanical and Physical Properties

Materials were compounded in either a 38-mm single-screw extruder or a 1-L K-mixer. Extrusion parameters were temperature profile 170°C to 190°C, screw speed 15 rpm, and residence time 1 to 2 min. The K-mixer parameters were 4500 to 5500 rpm and discharge temperature 171°C to 200°C. In all studies, test specimens were prepared by injection molding. At least five specimens of each blend were tested for each mechanical property. After molding, the specimens were stored over dessicant for at least 3 days before testing.

Mechanical properties were measured on the dry specimens at approximately 23°C. Specimens and test methods followed ASTM D256, D638, D747, and D790. Apparent melt viscosities and shear rates were calculated from measured volumetric throughput rates and pressure drops across the die during steady-state extrusion in a single-screw extruder at 190°C.

Selected properties from the various studies are summarized in Table 4. This table shows the larger effects of some more-sensitive variables. Because the conditions of each study varied somewhat, care must be taken when comparing data. General trends rather than actual values will be emphasized in the following discussion.

Reinforcement Effects

Addition of 30%–40% of any of the wood/paper reinforcing fillers to the plastics resulted in a composite with higher modulus and strength but lower impact energy and percentage of elongation and energy to maximum load. These effects are not surprising and are typical of reinforced thermoplastics in general. Table 5 summarizes property changes with addition of 40% ONP to several virgin and recycled polypropylenes.

Most major changes in composite performance occurred at filler contents below 30%. For example, aside from an approximate 10% increase in modulus, no major changes in mechanical performance were found over the rather narrow range of 32% to 42% ONP in VPP.

Table 4. Selected mechanical property data from various studies

Plastic	Filler	Coupling agent	Tensile modulus (GPa)	Tensile strength (MPa)	Izod impact energy Notched (J/m)	Izod impact energy Unnotched (J/m)
PPV	None	None	—	31.5	23.8	650
	40% ONP	E43	4.89	47.1	20.8	109
	40% BW40	E43	4.80	48.2	24.7	114
	40% WF	E43	3.72	34.1	18.7	72
HDPE-MB	40% ONP	E43	3.80	37.6	28.6	73
	40% BW40	E43	3.79	36.6	30.8	68
	40% WF	E43	2.61	27.8	36.4	81
PPV	40% ONP	None	4.97	34.0	20.8	113
	40% ONP	E43	4.90	47.4	19.8	144
	40% ONP	G3002	4.56	52.3	20.4	190
KPP	None	None	1.62	36.5	62.0	>800
	40% ONP	G3002	4.03	52.3	30.6	167
	40% OMG	G3002	3.55	38.9	34.2	138
BPP	None	None	1.32	24.5	165.0	>800
	40% ONP	G3002	3.98	42.5	34.3	150
	40% OMG	G3002	3.44	31.8	41.8	125

^aCoupling agent was added at a level of 3%–5% of fiber weight depending on the study.

Typically, the more fibrous fillers (ONP and BW40) resulted in composites with superior mechanical properties but were more difficult to process when compared to WF. However, the presence of clays and other impurities in the OMG made the material more difficult to disperse and interfered with bonding, resulting in decreased mechanical performance when compared with other fibrous fillers.

Figure 1 demonstrates some effects on viscosity at low shear rates. Viscosity measurements showed dramatic increases when WF was replaced with ONP and as fiber content was increased. This increase in viscosity demonstrated the effects of both aspect ratio and filler contents on composite melt behavior. Even at a fiber loading of 32% ONP, PPV composite melts were significantly more viscous than a 42% WF-filled PPV.

Table 5. Change in property after addition of 40% ONP to plastic

Property	Filled/Unfilled		
	KPP	BPP	VPP
Tensile			
Modulus	2.49	3.00	—
Strength	1.49	1.66	1.66
Energy	0.65	0.73	—
Elongation	0.47	0.46	—
Flexural			
Modulus	2.29	3.01	2.94
Strength	1.52	1.91	2.00
Impact			
Notched	0.49	0.21	0.86
Unnotched	<0.21	<0.19	0.29

Matrix Effects

Changing the melt flow index (MFI) of VPP had a significant effect on viscosity but little effect on mechanical performance over the range studied (3–30 g/10 min). This result suggests that MFI could be used to compensate for increased viscosity of the higher performance fibrous composites.

The polypropylene composites were stronger and stiffer than the HDPE–MB composites but had lower notched impact energy values. The unfilled plastic composites showed the same trend; i.e., composite properties qualitatively followed those of the matrix polymers. Similar trends were also seen for composites made with BPP and KPP. For example, the use of BPP (a block copolymer with higher notched impact strength) instead of VPP resulted in composites with higher notched impact strength, although the difference in this property was not nearly as great as that in the unfilled polymers. In other words, although choice of plastic can affect composite properties, addition of reinforcement can affect the mechanisms by which the plastic achieves its performance.

Analysis of the results of both the filler and plastic effects can lead to some interesting conclusions. For example, the mechanical properties of a recycled ONP/HDPE–MB system were at least as good as those of the current commercial WF/polypropylene system. This may offer some practical utility if problems associated with the high viscosity of the recycled system can be overcome.

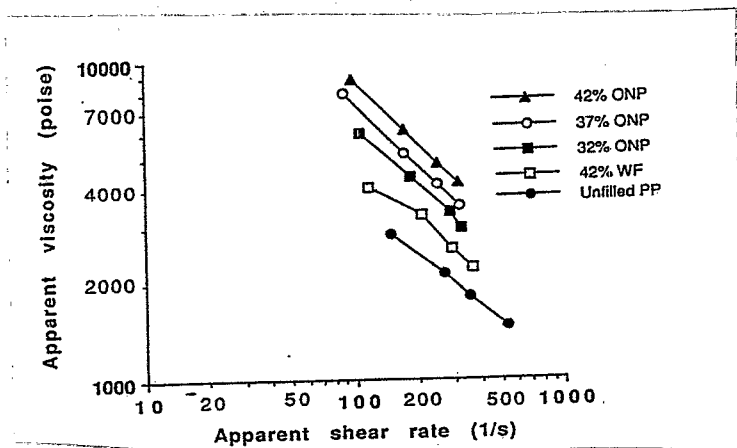


Figure 1. Apparent melt viscosity of composite blends.

Coupling Agent Effects

Coupling agents may be incorporated into composites in different ways to concentrate the material at the interface where it is active. In our studies, adding solid coupling agent (E43) to the melt or applying emulsified E43 directly to the fiber had little effect on mechanical performance of composites. Concentrating the emulsified additive at the fiber/matrix interface by precoating the fibers was apparently counteracted by the much lower chemical reactivity of the potassium salt with the cellulose compared with the reactivity of the anhydride with cellulose. Perhaps the E43 functions more to enhance dispersion of the cellulosic fillers than to bond the cellulose to the polymer.

The higher molecular weight maleated polypropylene, G3002, had a very beneficial effect on composite strength, energy absorption, and unnotched impact behavior. Initial moduli were approximately the same but ultimate strength and area under the curves (a measure of toughness) were much greater for systems with coupling agent (especially G3002). The fact that unnotched rather than notched impact energy was increased indicates that G3002 probably enhances dispersion of the fibers and reduces aggregates that would act as failure loci. For example, unnotched impact energy was more affected by removal of failure loci than was notched impact energy because the notch itself constitutes a failure locus. The apparently greater interaction of G3002 with polypropylene relative to E43 may be attributable to the higher molecular weight of G3002.

Processing Effects

Little preference was shown for compounding in a K-mixer or a single-screw extruder with WF or cellulose fiber. However, those fillers are relatively easily dispersed, and the ONP could not be melt-blended with a single screw. Moreover, the small interaction terms between filler and compounder in the statistical analysis also indicated some differences in dispersibility between WF and cellulose fiber.

We investigated the effects of re-extrusion on the mechanical and rheological performance of three different composite blends. For almost all mechanical and rheological properties, very little change occurred as the material was recycled over five cycles. The length and therefore the aspect ratio of the ONP fiber decreased, but these decreases were apparently not great enough to result in any large reductions in composite performance. The WF was reduced in both thickness and length, as smaller bundles of wood fibers were sheared off the larger particles. These changes in

dimension resulted in no overall change in aspect ratio and, ultimately, in composite performance.

Conclusions

- Melt-blended composites cannot be prepared with wastepapers as reinforcing filler using a conventional laboratory single-screw extruder. However, these composites can be prepared with a laboratory high-intensity K-mixer, an industrial-scale K-mixer, or a twin-screw extruder that employs a properly designed feeder for the fiber.
- Old newspaper (ONP) as reinforcing filler provides better properties than wood flour, which is currently used as filler in commercial composites. Old magazines (OMG) can also be used as a filler, but they are less easily dispersed into the matrix plastic and result in somewhat lower properties than those of composites containing ONP.
- With the same filler, substituting recycled milk bottle polyethylene for VPP leads to lower strength, stiffness, and unnotched impact energy, but higher notched impact energy.
- Use of recycled high density polyethylene from milk bottles and recycled polypropylene from battery cases as a matrix in composites with ONP results in improved impact performance when compared with that of composites made from VPP and ONP.
- Addition of the coupling agent Epolene G3002 at 3 weight percent of filler results in very useful increases in composite properties, probably as a result of improved fiber dispersion.
- Select composite systems showed little or no loss in mechanical properties when repeatedly reprocessed (re-extruded and injection molded).

COMMERCIAL IMPLEMENTATION

The research program led to many cooperative studies with industry and academia, all with the ultimate goal of commercializing composites made with waste materials.

Commercial Feasibility of Waste Newspaper-Thermoplastic Composites

The program was partially funded by the Forest Products Laboratory (FPL) and the Wisconsin Department of Natural Resources (DNR) and by in-kind contributions from the eight cooperating companies. Laboratory experiments demonstrated that old newspapers could be dispersed as fibers into thermoplastics by melt-blending,

resulting in substantial improvements in some properties compared with the unfilled plastic or plastic filled with wood flour. Major conclusions are as follows:

- Old newspaper/polypropylene (ONP/PP) composites can be compounded on a commercial scale using either the K-mixer with ONP flakes as feed or using a twin-screw extruder with ONP fibers fed separately from the plastic.
- An ONP/PP sheet containing 42 weight percent ONP can be prepared by extrusion on a commercial scale. This sheet meets existing specifications for automobile panels and can be thermoformed into a variety of shapes.
- Given proper design of melt processing equipment, a wide variety of other commercial products could be manufactured from ONP/PP composites with similar ONP content.
- Firm estimates of production costs for ONP/PP composite products must await
 - additional examination of compounding methods to define optimum balance of dispersion ability, throughput rate, and cost
 - improvement in methods to deliver wastepaper in a form and at a cost acceptable to a compounder or a manufacturer of plastic products

Waste LDPE Program

A consortium of companies is investigating the use of waste LDPE "contaminated" by residual fiber from a hydropulping operation that scavenges wood fiber from coated paper stock. The program involves raw material processors, compounders, plastics processors, and research institutions and is being coordinated by the FPL. Major hurdles in this program are the residual moisture in the raw material from the hydropulping process and product applications.

Waste Jute-Polyester Panels As Reinforcing Filler

In response to interest expressed by a U.S. company in the possibility of recycling panels produced by impregnating jute fibers with thermosetting polyester, we granulated the panels and investigated the ability to use the resultant mixture as reinforcing filler in melt-blended composites with a polypropylene matrix. Overall, this waste material produced composite mechanical properties approximately equivalent to those of similar composites containing wood flour as the reinforcement.

Waste Kenaf Core As Reinforcing Filler

This program resulted from a request by the Agrecol Corporation (Madison, Wisconsin) to determine whether kenaf core material could be useful as a reinforcing filler in plastic composites. We granulated the core material and successfully melt-blended the -40 mesh fraction with polypropylene. The composite properties were approximately equivalent to those of similar composites containing wood flour. Therefore, where kenaf core is readily available at low cost, it could very likely substitute for wood flour as a reinforcing filler.

Wastewood Composite As Reinforcing Filler

The University of Tennessee extension requested an evaluation of waste wood composite as a reinforcing filler in thermoplastic composites. Although such solid waste is available in large quantity; it contains wood with cured thermoset adhesives that might cause problems in melt-processed composites because the adhesives do not melt at processing temperatures. We granulated the plywood and successfully melt-blended the -40 mesh fraction with polypropylene. The composite properties were approximately equivalent to those of similar composites containing wood flour. This waste material could therefore substitute for wood flour as a reinforcing filler in melt-processed composites.

RECOMMENDATIONS

Research

Additional research is needed on both air-laid and melt-blended composites made from recycled wood fiber and plastics to improve properties and processing and to thereby increase potential applications.

1. Evaluate the potential for making composite materials with other major components of the waste stream, including low-density polyethylene, polystyrene, and mixed waste plastics.
2. Verify the recyclability of composites made with reground first-generation ingredients.
3. Improve melt-blending processes to achieve better fiber dispersion with minimal fiber breakage.
4. Improve bonding between wood fiber and plastic matrix to enhance physical and mechanical properties.
5. Improve impact energy and creep resistance (decreased deflection under long-term load), currently the limiting properties of these composite systems.
6. Determine the resistance of these composite systems to relatively extreme environments and develop means to enhance resistance to moisture, biodegradation, and fire.

Commercialization

1. Continue extensive outreach to industry to acquaint companies with these types of composite systems, to develop applications, and to cooperate in product development.
2. To obtain commercial acceptance of melt-blended composites containing wastepaper fiber,
 - a. improve methods for converting wastepaper at costs acceptable to industrial users; costs must at least approach current cost of wood flour (about \$0.22/kg)
 - b. improve methods for melt-blending fiber and plastics on a commercial scale at costs acceptable to melt fabricators (extruders, injection molders, etc.)