

Recycled Newspaper Fibers as Reinforcing Fillers in Thermoplastics: Part I—Analysis of Tensile and Impact Properties in Polypropylene

A. R. SANADI* AND R. A. YOUNG

*Department of Forestry
1630 Linden Drive
Russell Laboratories
University of Wisconsin
Madison, WI 53706*

C. CLEMONS AND R. M. ROWELL**

*Forest Products Laboratory
1 Gifford Pinchot Drive
Madison, WI 53705*

ABSTRACT: Recycled newspaper fibers (ONP) are potentially outstanding nonabrasive reinforcing fibers with high specific properties. In this study, a high energy thermokinetic mixer was used to mix these fibers in a polypropylene (PP) matrix, and the blends were then injection molded in order to observe the tensile and impact strengths of the composites. A 40% (weight) of ONP in PP resulted in a tensile strength of 34.1 MPa and an unnotched Izod impact strength of 112 J/m. Small quantities of maleic anhydride-grafted polypropylene (MAPP) and acrylic acid-grafted polypropylene (AAPP) were used to improve the interaction between the hydrophobic PP and the highly polar fibers. The improvement in properties by using MAPP depended on the amount of maleic anhydride in the graft copolymer and the molecular weight of the copolymer. Tensile strengths as high as 57 MPa and an unnotched Izod impact strength of 212 J/m were achieved with the addition of one of the MAPP, while smaller improvements were found with AAPP. Reinforcing efficiencies and scanning electron microscopy have been utilized to aid in the analysis.

INTRODUCTION

A GREAT DEAL of interest has been generated in the use of wood fibers as reinforcing fillers in thermoplastic composites. The low densities of ligno-

*Author to whom correspondence should be addressed.

**Also: Department of Forestry, 1630 Linden Drive, Russell Laboratories, University of Wisconsin, Madison, WI 53706.

cellulosic fibers coupled with their low cost makes these fibers potentially attractive for non-structural composite applications [1]. An added advantage is the possible use of recycled wastepaper fibers in a recycled thermoplastic (HDPE or PP) matrix. However, the inherent polar and hydrophilic characteristic of the fiber gives rise to difficulties in compounding with nonpolar plastic matrices. This vastly different surface nature of the fiber and matrix also results in poor bonding between the fiber and matrix. A recent review by Maldas and Kokta [2] covers the complexities involved with the compatibilization of these materials and the different techniques used to understand the interfacial interaction.

Many factors contribute to the overall performance of a composite [3]. The fiber aspect ratio, uniformity of dispersion of the fibers in the matrix, and the ability of the interface to transfer stress to the fiber are some of the important factors that need to be studied to optimize composite properties. Several studies aimed at improving dispersion of the lignocellulosic fibers in the non-polar matrix and increasing the stress transfer efficiency of the interface have been conducted in various laboratories [1,4-9].

Lignocellulosic fibers, being of natural origin, have a wide range of properties depending on the type, source and age of the lignocellulosic. The pulping and processing conditions also affect the final newsprint fiber properties. It is thus difficult to obtain realistic fiber strengths for a batch of fibers and then estimate the theoretical strength of the composites for specific fiber lengths and fiber-matrix interfacial strengths. Furthermore, it is also difficult to attribute specific values to any of the above factors due to the complexities arising in defining the exact contribution of each of them.

In this article tensile and impact properties of 40% (by weight) of ONP in PP have been reported. In this article we will also discuss the use of reinforcing efficiencies, i.e., the efficiency of the fibers in strengthening the composite. Scanning electron microscopy (SEM) has been used to study the fracture surfaces.

Theory of Fiber Reinforcement

The tensile strength of fiber reinforced composites can be estimated using the modified rule of mixtures [10].

$$\sigma_c = V_m s_m^* + V_f \sigma_f K_1 K_2 \quad (1)$$

where

σ_c = the tensile strength of the composite

σ_ϕ = the tensile strength of the fibers

V_m = the matrix volume fraction

V_f = the fiber volume fraction

K_1 = orientation factor

K_2 = a factor dependent on the stress transfer between the matrix and the fibers

σ_m^* = the tensile contribution of the polymer matrix at the failure strain of composite

K_1 is an empirical factor that accounts for fiber orientation and has a value of unity for aligned fibers, 3/8 for planar random configuration and 1/5 for a three-dimensional random orientation. It is difficult to determine this factor for injection molded fiber-polymer systems due to the difference in fiber orientations at the skin and core of the specimens. The stress transfer Factor, K_2 , depends on the adhesion and interaction between the matrix and the fibers and the length of the fibers present in the composite.

The critical length (l_c) is inversely proportional to the fiber-matrix interfacial strength (τ_i) by the following equation [13]:

$$l_c = \frac{\sigma_f d}{2\tau_i} \quad (2)$$

where d is the fiber diameter and l is the fiber length.

If the fiber lengths are less than the fiber critical length, the fibers never reach their full reinforcing potential. For a short fiber composite, where the fibers never fail since they are all smaller than the critical length, K_2 is given by:

$$K_2 = l/2l_c$$

In the above case, K_2 has a maximum value of 0.5. If the fibers are equal to or longer than l_c , K_2 varies between 0.5 and 1.0, and the fibers can fail by fracture and,

$$K_2 = 1 - l_c/2l \quad (4)$$

In this case the composite failure is assumed to be at the failure strain of the fibers and σ_m is the stress taken up by the matrix at the failure strain of the composite, K_2 [Equations (3) and (4)] thus accounts for the fiber-matrix adhesion and the fiber length present in the system.

Quantifying the exact contributions from the various factors that govern composite properties is particularly difficult in short lignocellulosic-fiber injection molded composites due to the following reasons:

1. Orientation factors are difficult to determine due to differences in the skin and core of the injection molded specimens.
2. Interfacial shear strengths are difficult to estimate due to the uneven topography and twisted nature of the lignocellulosic fibers.
3. There is a wide distribution of fiber lengths present, with fiber both shorter and longer than the fiber critical length.
4. Fiber strength of natural fibers can vary a great deal depending on the source, age, and method of harvesting of these natural materials.

The volume of the polymer in these injection molded composites can be 50% or more, and the contribution from the matrix is thus significant. Furthermore, the addition of internal lubricants and processing aids may change the matrix

properties considerably. Information gained by comparing tensile strengths of the composites may be masked by changes in matrix contribution to the composite strength. The reinforcing efficiency (ϕ) is defined as the product of K_1 and K_2 and is a function of the fiber orientation, the fiber length, and the fiber-matrix adhesion [$\phi = f(K_1, l, \tau)$]. The contribution of fibers to the tensile strength of the composite (σ_{cf}) is equal to $V_f \sigma_f K_1 K_2$, and is determined by subtracting the contribution of the matrix from the tensile strength of the composite. Matrix contribution is determined experimentally from the stress-strain of the polymer and knowledge of the failure strain of the composite. Estimating reinforcing efficiencies can provide insights into the effectiveness of the fibers in reinforcing composites and has been used effectively by other researchers [11]. This also permits the comparison of the effectiveness of the fibers while using different matrices, varying volume fractions and using different processing techniques. Ideally, a third phase, the interphase should be taken into account; however, our present knowledge of the interphase properties is limited and we have neglected it here.

EXPERIMENTAL METHODS

Materials

The matrix polymer was a polypropylene (PP), Fortilene 1602 (Solvay Polymer, Houston, TX) with a melt flow index of 12 g/10 min (ASTM D-1238). Three maleic anhydride grafted polypropylene modifiers. Epolene E-43 and Epolene G-3002 [12] (Eastman Chemical Products, Kingston, TN) and BPMA (PolyBond-3002, BP Chemicals, Hackettstown, NJ) of different molecular weights and acid numbers were used as coupling agents. An acrylic acid grafted polypropylene (AAPP) (PolyBond-1001, BP Chemicals) was also tested as a coupling agent. Data on the matrix and coupling agents are given in Table 1. The fibers used for compounding were air-dried recycled newsprint fibers (ONP).

Methods

Old newspapers were run through a Sterco mini granulator (Sterling Inc., Milwaukee, WI) with a 5/16" screen to reduce the size of the paper to approx-

Table 1. Data on materials used.

<i>PP</i> : tensile strength = 34 MPa, tensile modulus (secant at 1% strain) = 965 MPa.
<i>E-43</i> : maleic anhydride-grafted polypropylene, $MW_{wt} = 10,000$, $MW_H = 4200$, 6% (wt.) of maleic anhydride in copolymer.
<i>G-3002</i> : maleic anhydride-grafted polypropylene, $MW_{wt} = 40,000$, $MW_H = 11,000$, 6% (wt.) of maleic anhydride in copolymer.
<i>BPMA</i> : maleic anhydride-grafted polypropylene. $MW_{wt} > 100,000$, <0.5% (wt.) maleic anhydride in copolymer, melt index = 7 (ASTM D1238).
<i>AAPP</i> : acrylic acid-grafted polypropylene, melt index = 40, 6% (wt.) of acrylic acid in copolymer, melt index = 40 (ASTM D1238).

imately 5 mm² flakes. The flakes, PP and coupling agent were compounded in a high intensity kinetic mixer (Synergistics Industries Ltd., Canada) where the sole source of heat is the kinetic energy of high speed blades.

Two methods of mixing were used in our experiments.

- 1 Single stage method: in the first 90 g of PP, 60 g of ONP (the moisture content of the fibers was accounted for to obtain 60 grains of fibers) and a coupling agent (3% of the fiber weight) were blended in the K-mixer at 4600 rpm for 2 min for adequate fiberization of the newspaper. During this time the temperature of the blend was between 115 to 135°C. After this step, the rpm was increased to 5500 to melt the PP, the flux the mixture, and disperse the fibers. The mixture was automatically discharged at 200°C and the total residence time of the mixing operation was about 140 seconds.
- 2 Two stage method: in the second, the fibers were first mixed with the coupling agent at 4600 rpm for one minute. The blender was then switched off and the PP added into it. This was then mixed at 5500 rpm until the mixture was discharged at a dump temperature of 200°C. In both techniques, we observed steam being released through a vent prior to the discharge of the mix.

The resulting mixtures were granulated and dried at 105°C. Test specimens were prepared using a Frohring model SP50 (Newbury Industries Inc., Newbury, OH) plunger type injection molder at 204°C. The residence time was approximately 1 min, ram pressure 7.5 to 9.0 MPa and the mold temperature at 30°C.

The specimens were stored in a dessicator for 3 days before testing. At least five specimens were tested for each composite blend and property. Tensile tests were conducted following ASTM D 638-90, and Izod impact conducted using ASTM D 256-90 standard. Scanning electron microscopy (JEOL 840) was used to observe the fracture surface of the notched Izod impact samples.

RESULTS AND DISCUSSION

Tensile Properties

Table 2 shows data on composite tensile and impact properties. Use of the E-43 and G-3002 resulted in considerable improvement in the tensile strength over control specimens (without any coupling agent). Improvements with AAPP and BPMA were more meager.

The stress-strain curves of the uncoupled, E-43 and G-3002 composites are shown in Figure 1. The nonlinearity in the curves is mainly due to the plastic matrix deformation. However, the distribution of fiber lengths present in the composite can also cause the slope of the stress-strain curve to decrease with increasing strain [13]. This is because the load taken up by the fibers and the efficiency of the fibers decreases as the strain increases. A detailed explanation of this phenomenon is beyond the scope of this article but is described elsewhere [13]. The difference in the curves of E-43 and G-3002 systems, particularly the higher failure strain of the latter, is noteworthy. The E-43 systems failed at a strain of about 2.2%. At this strain there were enough fibers in the G-3002

Table 2. Properties of composites.

Coupling Agent	Tensile		Izod Impact (J/m)	
	Modulus (GPa)	Strength (MPa)	Izod Impact (J/m)	
			Notched	Unnotched
none	4.42 (0.2)	34.1 (1.1)	20.8 (0.5)	112(18)
E-43	4.41 (0.3)	47.4 (2.6)	19.7(1.6)	144(11)
G-3002	4.12 (0.2)	52.3 (2.1)	20.4 (0.9)	190(25)
AABP	—	38.8 (0.5)	21.7 (0.2)	117(24)
BPMA	—	39.8 (0.6)	23.5 (2.3)	107(13)
G-3002(*)	—	57.0 (0.7)	21.6 (0.1)	212 (24)
AABP(*)	—	40.2 (0.5)	17.6 (1.0)	123 (4)
BPMA(*)	—	41.7 (0.3)	18.6 (1.9)	136 (8)

*Indicates two stage mixing technique, all others were single stage mixing. Standard deviations in brackets.

system (which has a lower critical length because of enhanced fiber-matrix bonding) to take up additional load; failure occurred at a strain of 3.5% when the fibers in the G-3002 system were unable to take up any additional load. The tensile moduli of the three systems measured were approximately equal.

The higher tensile strength of the G-3002 composite system over the E-43 system is probably due to improved *interphase* properties. The longer molecular chains of G-3002 results in greater molecular entanglement that strengthens the interphase. It is important to recall that as the degree of polymerization (DP) increases, the mechanical properties of a polymeric material improve until a critical DP, beyond which no significant effect on polymer mechanical properties occurs. Furthermore, the influence of molecular weight on the interphase morphology and crystallization of PP onto the fibers are an important consideration

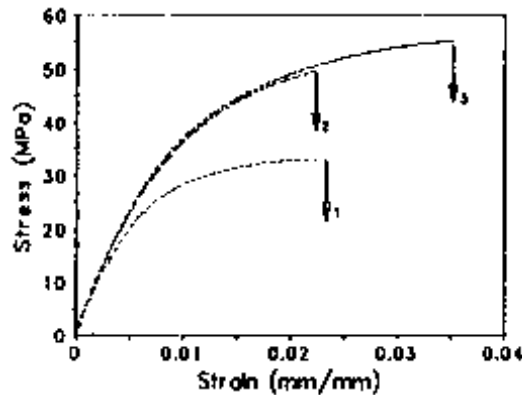


Figure 1. Stress-strain curves of the composites using 40% (wt.) of newspaper fibers in PP: (1) uncoupled. (2) E-43 as the coupling agent and (3) G-3002 as the coupling agent.

[14,2]. Felix and Gatenholm [14] report that the higher molecular weight of maleated PP compatibilizers result in a higher number of nuclei formed on the fiber surface. It is difficult to postulate if any further increase in molecular weight of the maleated PP will further improve interphase properties and result in improved composite properties. Excessively long chain lengths may reduce the potential of anhydride-hydroxyl covalent bond formation due to steric hindrance. Furthermore, the possibility of different molecular orientations at the interphase region complicates the situation. The lower strength properties obtained using BPMA as compared to both E-43 and G-3002 is due to the lower graft content (less than 0.5% by weight) and limited covalent/acid-base interaction. It is interesting to note that even a low graft content of the BPMA resulted in better properties than the AAPP system which had about 6% acrylic acid.

The anhydride groups present in the copolymer can covalently bond to the OH groups of the lignocellulosic fiber surface. It is important to point out that the presence of moisture in the fibers will hydrolyze the anhydride to its acid form [15] and covalent bonding through ester linkages is not likely to occur. Reversal of the acid to its anhydride form during the mixing operation is also possible under dehydrating conditions and sufficient temperature. We feel that the improved properties in the MAPP systems are due to a combination of some formation of covalent linkages (due to reconversion to the anhydride form) and enhanced acid-base interactions between the fibers and MAPP.

Property improvements using AAPP are less than with MAPP. With the processing condition used, it is unlikely that ester linkages can be formed between the acrylic acid of AAPP and the fiber surface. Improvement using AAPP is primarily due to acid-base interaction between the acrylic acid and the fiber surface. If the maleic anhydride is in its acid form, the possibility of acid-base interactions between the fiber surface and the two neighboring carboxylic groups of maleic acid should be considerably greater than with the single carboxylic group in the AAPP. In-depth chemical and spectroscopic analysis is difficult due to the presence of binders and inks present in the recycled newspapers.

Two different techniques were used during the mixing operation to obtain information on whether the presence of PP, in a single stage processing, restricts or hinders the migration of the AAPP or MAPP to the surface of the fibers. The difference between the two methods of mixing were not significantly different to dramatically impact the composite properties, although improvement in tensile and unnotched impact strengths were observed using the two stage process. In the one-step method, the fibers, AAPP or MAPP, and the PP were added simultaneously and it is likely that the MAPP/AAPP migrates and gets localized near the fiber surface. The polar MA and AA groups can thermodynamically segregate to the highly polar fiber surfaces during the high shear mixing operation. Evidence of MAPP localization in a cellulose-PP system has been obtained by transmission electron microscopy coupled with energy dispersive X-ray analysis [16]. There is no reason to doubt that the AAPP also migrates to the fiber surface.

In the second, two-stage technique, the fibers were first mixed for one minute with the coupling agent before the addition of PP. A slight improvement in both tensile strengths and unnotched Izod impact strengths were observed in all the

systems when using the two stage technique. Enhanced interactions at the inter-phase may be the cause of the slight property enhancement, though there is no conclusive evidence since this improvement can also be due to reduced fiber attrition with tile reduced processing time in the high shear mixer.

Strengthening Efficiencies

Table 3 shows estimated values of the contribution of the fibers to composite strength (σ_{cf}), where $\sigma_{cf} = V_f \sigma_f \phi$, and is estimated by subtracting the contribution of the matrix, $V_m \sigma_m^*$, from the experimental values of composite strength. V_f of the composites was estimated by assuming that the fibers had an apparent density of 1.5 g/cc. σ_m^* is the stress taken up by the matrix at the failure strain of the composite and is approximately equal to 23.8 MPa in all cases. Comparing the ratio of σ_{cf} for the modified system to that of the uncoupled system gives a direct comparison of ϕ , since σ_f and V_f are equal in all cases and cancel out.

The improvements in tensile properties due to the addition of MAPP or AAPP or by changing the mixing technique are also observed in the comparison of the contribution of the fibers to composite properties. A significant increase in σ_{cf} from 10.3 MPa for the uncoupled system to 33.2 MPa for the G-3002 system using the two stage blending process was observed.

The ratio of $\phi/\phi_{\text{uncoupled}}$ is a measure of the improvement in reinforcing efficiency with the use of the various additives. The improvement using the two stage method is more clearly seen here. Direct reinforcing efficiencies can only be estimated if the fiber strengths are known. Assuming a fiber tensile strength of 750 MPa, a reasonable estimate [17], the reinforcing efficiencies vary from 0.034 for the uncoupled composite system to 0.111 for the G-3002 coupled composite (Table 3). The ϕ for glass fiber reinforced injection molded PP has been reported to be as high as 0.19 [11]. The ϕ for a 2-dimensional randomly oriented fiber system for a well bonded system can have a maximum value of 0.33, while a 3-dimensional well bonded system can reach a maximum ϕ of 0.2. The low ϕ for the systems

Table 3. Estimated contribution of fibers to composite strength and reinforcing efficiencies.

Coupling Agent Used	σ_c (MPa)	σ_{cf} (MPa)	$\phi/\phi_{\text{uncoupled}}$	ϕ ($\times 10^2$)
uncoupled	34.1	10.3	1.00	3.4
E-43	47.4	23.6	2.29	7.8
G-3002	52.3	28.5	2.77	9.5
AABP	38.8	15.0	1.45	5.0
MABP	39.8	16.0	1.55	5.3
G-3002(*)	57.0	33.2	3.22	11.1
AABP(*)	40.2	16.4	1.59	5.5
MABP(*)	41.7	17.9	1.74	6.0

*Indicates two stage mixing method, all others were single stage mixing. Standard deviations in brackets.

we have studied, even for the G-3003 systems which has good fiber-matrix bonding, suggest that the very short fibers lengths strongly influence composite properties. As pointed out by Bigg [11], fiber lengths of about ten times the critical length are needed to achieve good reinforcing efficiencies.

Impact Properties and SEM Analysis

The impact strength of the composites depend on the testing procedure, i.e., whether the samples were notched or unnotched. There is little difference between the notched Izod impact strengths of the three composites evaluated in this study (Table 2). This suggests that there is little difference in the crack propagation energy, but considerable differences in the crack initiation energies. The fibers were too short to result in significant energy absorption through the fiber pull-out mechanisms. Although some pull-out was observed in the uncoupled system (Figure 2)—lateral contraction of these natural fibers can reduce the contribution through the fiber pull-out mechanism.

On the other hand differences were observed for the unnotched impact strengths; incorporation of coupling agents improves the fiber-matrix interaction and the work of fracture of the interphase. This enhanced toughness of the interphase increases the resistance to crack initiation and there is a subsequent increase in the unnotched impact strengths. This improvement in unnotched impact



Figure 2. SEM of the fracture surface (notched Izod test) of the uncoupled composite showing some fiber pull-out and topographical uneven surface.

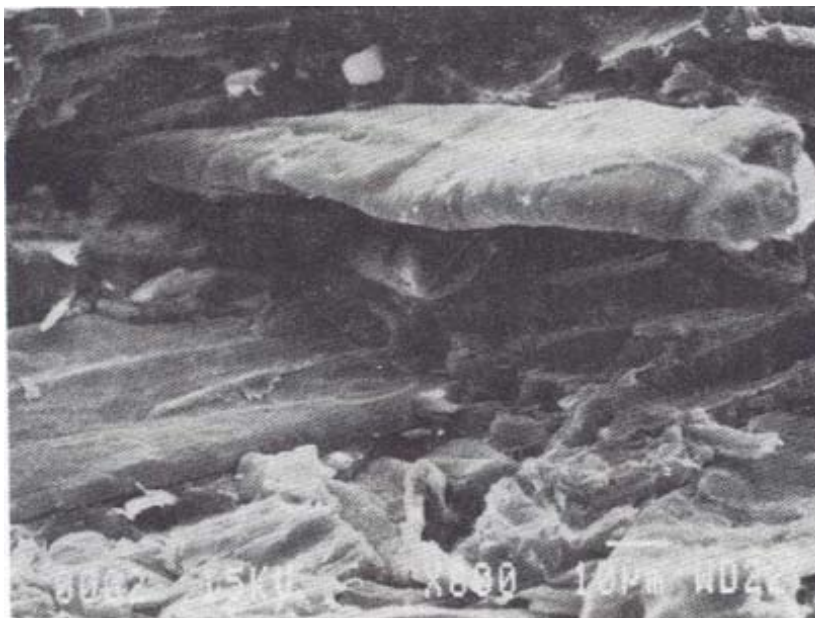


Figure 3. SEM of fiber pulled out of the fracture surface of the uncoupled composite showing fairly clean fiber surface suggesting poor fiber-matrix bonding.

strengths is seen with fibers with lengths below their critical lengths, and has also been observed in glass fiber-PP systems [18]. The high shear mixing using the K-mixer causes a great deal of fiber attrition and it is possible that a large proportion of the fibers are below their critical lengths. A more detailed study of this phenomenon is underway and initial results suggest few fibers longer than the estimated critical length remain in the final composite. In general, the maximum toughness is attained when all fibers are equal to the critical length and energy absorptions due to fiber pull-out and debonding are at a maximum [10]. Thus the slight increase observed when using the two stage technique may be due to enhanced fiber-matrix interactions and/or increases in fiber lengths due to reduced fiber attrition.

SEM observations of the fracture surface of notched Izod specimens indicate there is considerable difference in the fiber-matrix interaction between the uncoupled and coupled composites. Uncoupled composite fracture surfaces show some fiber pull-out (Figure 2) and fairly clean fiber surfaces (Figure 3). Composites prepared using E-43 and G-3002 (Figure 4 and Figure 5, respectively) show little fiber pull-out and a more topographically even surface. Any losses in the debonding and pull-out energy due to decrease in fiber pull-out in the E-43 and G-3002 systems are compensated for by a tougher interphase. The improved bonding is clearly seen in Figure 6 for the E-43 system where the fiber has pulled out from the matrix but a fair amount of polymeric residue remains on the fiber.



Figure 4. SEM of the fracture surface (notched Izod test) of the E-43 coupled composite showing little fiber pull-out.

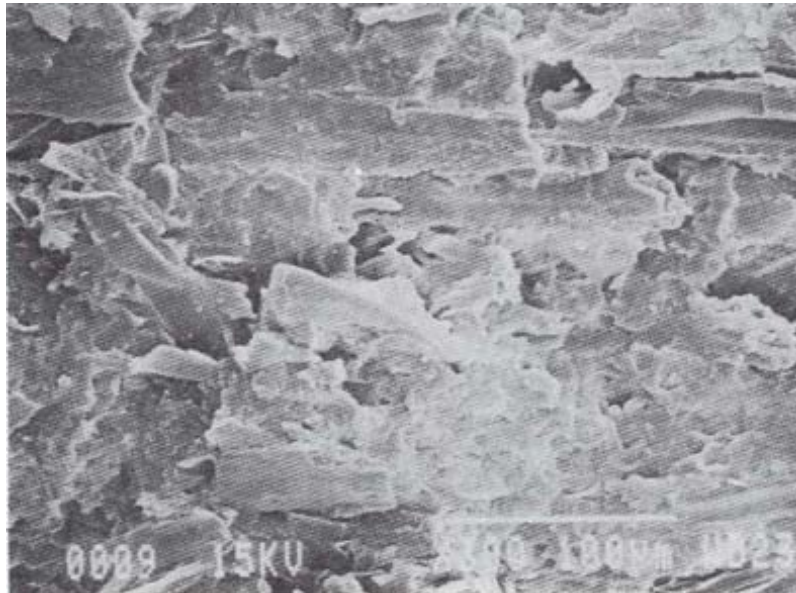


Figure 5. SEM of the fracture surface (notched Izod test) of the G-3002 coupled composite showing no fiber pull-out and topographical even surface.

The loci of the failure (Figure 6) suggest good fiber surface-interphase bonding, but the treatment has resulted in a weak boundary layer in the interphase. As mentioned earlier, a minimum degree of polymerization (DP) is necessary for good mechanical integrity and it is possible that the DP of E-43 is below the critical value needed for the high strengths. The fibers seen on the fracture surface of the G-3002 coupled system seem to have undergone some tearing and plastic deformation (Figure 7) resulting in an additional energy absorbing mechanism [19]. SEM of AABP and BPMA did not show the indications of improved bonding of the E-43 and G-3002 systems, although some improvement over the uncoupled systems was observed.

CONCLUSIONS

The high energy kinetic mixer helps improve the dispersion of newspaper fibers in the PP matrix and the use of the coupling agents results in improved composite properties. The improved dispersion of the fibers by using this mixer compensates for the loss in efficiency of the composites due to the presence of low aspect ratio fibers in the composites. Good adhesion between the polar fibers and maleated polypropylene matrix has been observed using SEM of the impact fracture surface of the composite. The low strengthening efficiencies observed.

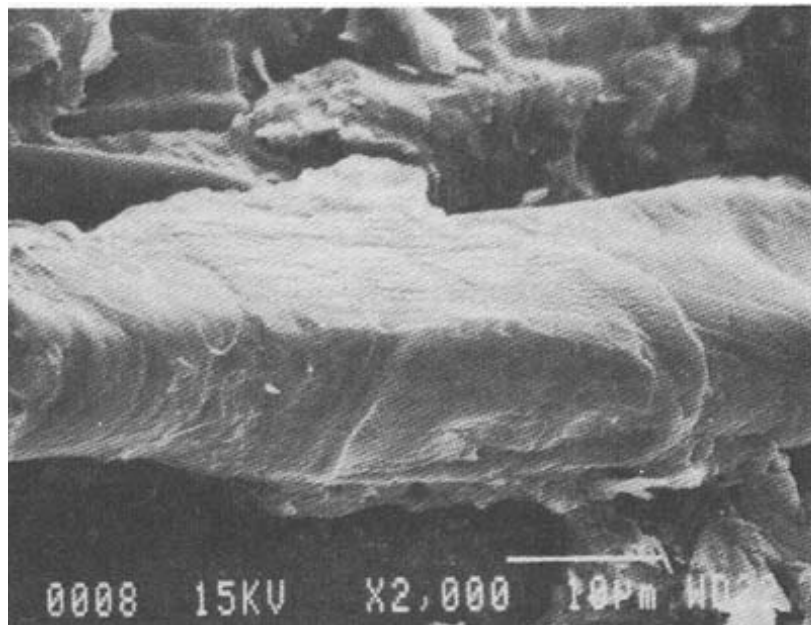


Figure 6. SEM of a pulled out fiber of the E-43 coupled composite showing a considerable amount of polymer adhering to the fibers which indicates good fiber-matrix adhesion. The loci of failure may indicate failure in the interphase region.

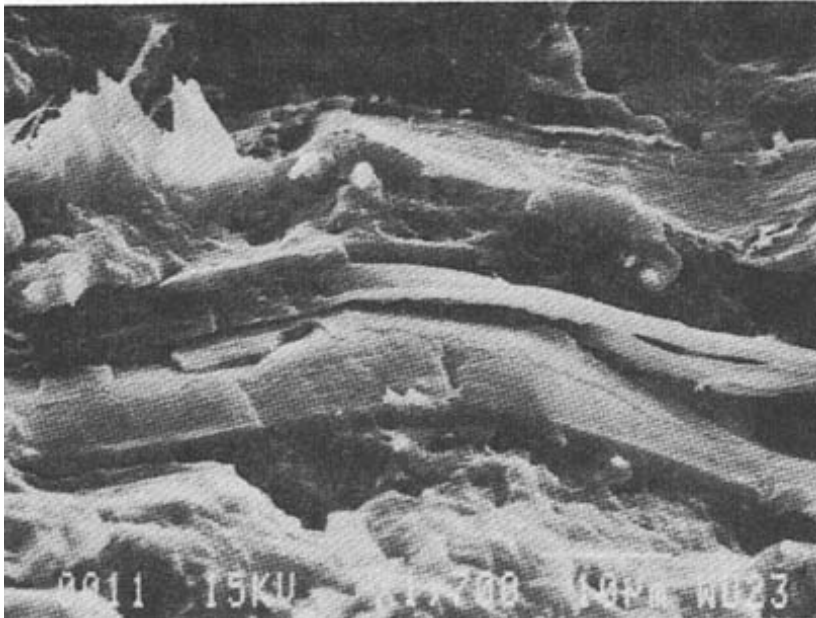


Figure 7. SEM of fibers split in the fracture process in the G-3002 composite revealing fibril-fibril debonding.

even for the well bonded systems, suggest that the short fiber lengths may be the primary reason for this loss in efficiency. Prevention of extensive fiber attrition through the use of lubricants and suitable processing equipment, improved bonding with maleated polypropylenes, and the use of impact modifiers should result in significant improvements in composite properties.

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