Use of Some Plant Wastes as Fillers for Polypropylene

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ABSTRACT: Palm kernel shell and the palm fruit pulp fiber were used as additives for polypropylene. The mechanical properties of the polymer, namely, tensile strength, impact strength, surface hardness, and flexural modulus were remarkably enhanced. This was attributed to the fact that these solid wastes functioned as reinforcing fillers through bonding interactions and stress-sharing mechanisms. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 90: 1447–1452, 2003

Key words: fillers; poly(propylene)

INTRODUCTION

Fillers give body, strength, and other desirable properties to plastics, elastomers, papers, paints and textiles.[1] Essentially, they are used for nondecorative purposes, although some may incidentally impart color or opacity to compositions. They modify existing physicochemical characteristics but also invariably reduce cost, improve workability during processing, and develop new properties not present in the original resin.[2] The use of fillers dates back to the early Roman era when artisans incorporated ground marble into lime plaster, frescoes, and pozzolanic mortar.[3] In the paper industry, the application of fillers started in the mid-19th century when silica, diatomite, and titanium dioxide were used in bond papers.[3] Functional fillers, which introduce new properties instead of modifying existing ones, were commercially developed early in the 20th century as carbon black was added to rubber, and phenol formaldehyde plastics were formulated with wood flour.[4] This treatment of polymerics with fillers became necessary, as it was recognized that most of these materials could not successfully be processed or used in their pure or native forms without these additives.[5,6]

Nearly every aspect of the oil palm plant is very useful to man, except perhaps the kernel shell (Ks) and the pulp fiber (Kf), which only find outlets as local firewood and lamp wicks, respectively. In this article, we present the results of incorporating these otherwise waste agricultural products into ELPROP, the polypropylene (PP) from the NNPC Plant at Eleme, Port Harcourt, Nigeria.

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EXPERIMENTAL

Materials

The two filler samples were obtained from the African oil palm, *Elaeis guineensis,* and had characteristics as shown in Table I. The characteristics of the PP received from the Eleme Petrochemical Plant, near Port Harcourt, Nigeria, are shown in Table II.

Methods

Filler preparation

The Ks was washed thoroughly with hot detergent solution, dried at 105°C for 2 h, crushed with an Atlas YL 112-4 (5 HP) electric grinder (Proda, Enugu, Nigeria), and finally milled with a manual Corona lever machine (model Landers and CIA, Medellin, Colombia). The powder was filtered with a filter cloth to further reduce its fineness and was then sieved with a Gilson automatic sieve (model SS-15, Washington).

The fruit pulp fiber was immersed in boiling water for 1 h to liberate the enmeshed oil. On removal, the sample was repeatedly washed in warm detergent solution until a grayish mass was obtained.

Characterization of the fillers

Specific gravities of the two powders were measured according to ASTM D 2840 with an air comparison pycnometer.

Color was evaluated with a Lovibond tinctometer (model E).

Solubility performances of the samples were examined in water, acetone, *n*-hexane, and dilute HCl.

The surface chemistry of the fillers was determined by the pH technique;[7] the pH of the aqueous slurry made by stirring 1 g of the sample in 10 cm^3 of boiled distilled water obtained with a Horiba pH meter (model M-8,

TABLE I
Some Physical Properties of the Fillers

Filler properties	Ks	Kf
Specific gravity (g/cm^3)	0.64	0.56
Charring temperature (°C)	180–240	180–200
Color	Dull: 1.5 yellow/4.2 green	Dull: 1.4 yellow/4.1 green
Particle size (μm)	150	150
pH of aqueous slurry	5.30	5.03
Solubility performance in water, ethanol, acetone, *n*-hexane, and dilute HCl	Insoluble	Insoluble

Tokyo, Japan). The mixture was cooled before readings were taken.

Charring temperature was determined by visual notation of the changes in color of the sample subjected to a heating program.

Metal contents of the powders. The sample was digested by the standard technique,[8] and K, Na, Ca, Fe, Cu, Zn, and Pb were assayed with an atomic absorption spectrophotometer (Buck Scientific, model 200A, New Jersey) under the following specifications: air–acetylene flame type, helium lamp type, 2600°C flame temperature, and 6 mm flame height.

In addition, the anionic contents, namely, SO_4^{2-}, NO_3^{-1}, Cl^{-1}, SiO_3^{2-}, Br^{-1}, and PO_4^{3-} ions were determined with a digital direct reading spectrophotometer (model DREL 5, Hach, Loveland, CO).

Preparation and characterization of filled PP composites

Exactly 200 g of ELPROP was mixed with varying weights of each filler sample. The weights used were 20, 40, 60, 80, and 100 g, corresponding to 10, 20, 30, 40, and 50 ppm; these were subsequently compounded separately in a single-screw injection molder (Model ATS FAAR, Metal-Fluid Engineering, Italy) at a pressure of 150 Nm^{-2} and a temperature of 190°C. Dumbbell-shaped bars were produced for each composition.

Impact strength measurement. This was carried out with an Izod digital impact tester (Milan, Italy) with a notch (Vis No. 250-351) and range selector of 2–25 J. Readings were taken when the impact mode on the test piece broke the piece.

Flexural modulus. This was determined with a three-point bending jig clamped to a universal tensometer under specifications complying with BS 2782. A standard load cell of 100 N was allowed to gradually impress the test piece. With a computer incorporated into the system, the average value of the flexural modulus and the bending pattern of each test composition were obtained.

Surface hardness. The resistance of the composites to surface attrition was investigated with a Rockwell testing machine, (model ATS FAAR, Italy) that is, the pin technique.

Tensile properties. The tensile yielding and breaking strengths of the samples were obtained with a computer-aided Instron tensile testing machine (ILAO TIEH model 4501, IX series, Manchester, UK) under the following specifications:

Specimen type: ASTM.
Sample width (mm): 12.83.
Sample depth (mm): 3.19.
Span (mm): 50.80.
Sample rate (points/sec): 5.00.
Crosshead speed (mm/min): 1.27.
Static load cell (kN): 5.00.

Softening temperature. A Vicat high digital temperature tester (model NP/S) was used to determine softening temperature.

RESULTS AND DISCUSSION

The results shown in Table I and in Figure 1(a,b) indicate very close similarities in the properties of the two fillers. This is hardly surprising because the two materials are from the same plant. It was necessary to use very fine powders (average 150 μm) because this enhances the effectiveness for most reinforcing powders. Exhaustive analysis of the fillers showed a complete absence of toxic ions such as Pb and Hg. Figure 2 shows high concentrations of sulphate ions and an even distribution of sodium ions in both fillers.

It was evident [see Fig. 1(a)] that incorporation of the two filters gradually increased the impact strength of PP from 3.96 kg/cm^2 for the native resin to 4.52 kg/cm^2 for Ks and 4.60 kg/cm^2 for Kf. This observation suggests that the two fillers were reinforcing in action; the higher values observed for Kf were attributable to its fibrous nature.[9] In similar observations [see Fig. 1(b,c)], the reinforcing actions of the two fillers with respect to flexural modulus and surface hardness were clearly evident. Here, too, for most compositions, Kf was clearly more effective than Ks,

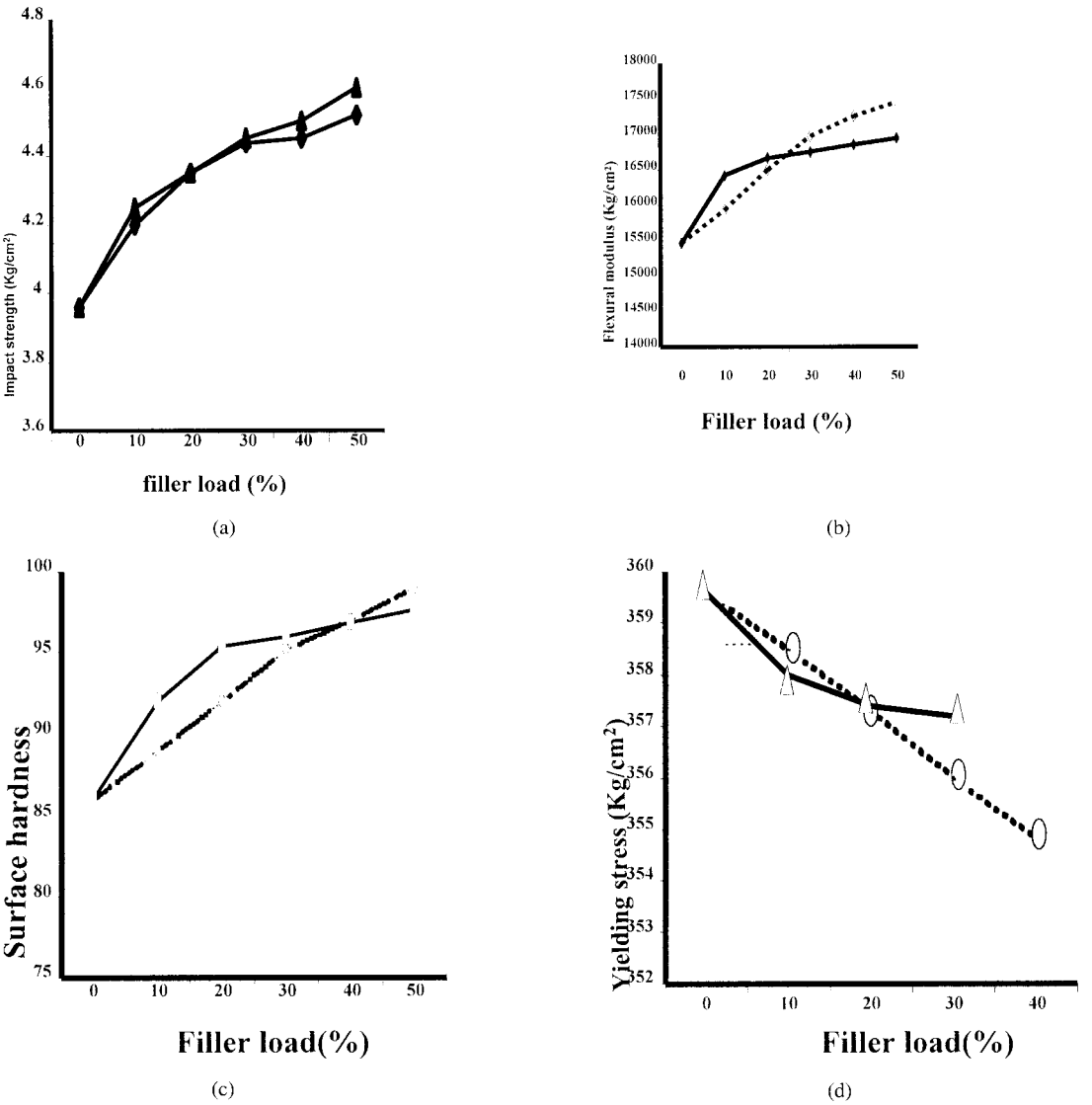
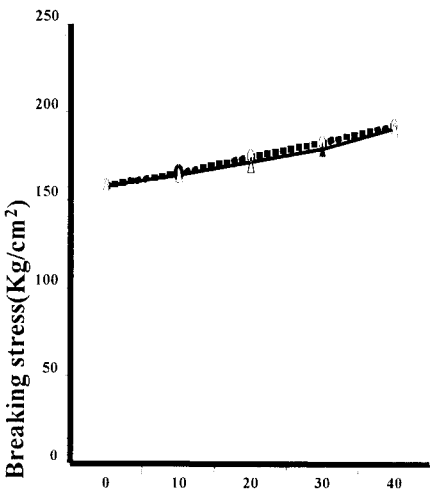


Figure 1 Effect of filler load on the following properties of PP: (a) impact strength [(◆◆◆) Ks and (▲) Kf], (b) flexural modulus [(◆) Ks and (◆◆◆) Kf], (c) surface hardness [(○) Ks and (□) Kf], (d) tensile (yielding) properties [(○) Ks and (△) Kf], (e) breaking properties [(○) Ks and (△) Kf], (f) strain percentage [(□) Ks and (△) Kf], (g) elastic modulus [(☒) Ks and (○) Kf], and (h) surface hardness [(○) Ks and (△) Kf].

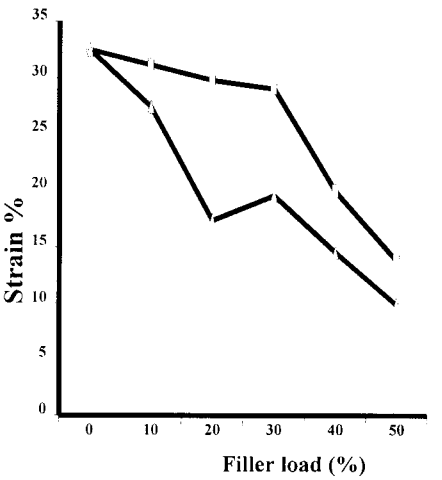
and these actions were more dramatic at lower filler concentrations; if anything, their effectiveness tapered at higher loadings. This was in agreement with the postulates of Billmeyer,[2] who maintained that this behavior of reinforcing filters was due essentially, to diluting action arising from increases in the partial volume of the fillers at higher pph values. The enhancement of surface hardness shown in Figure 1(c) was interesting as it indicated that these fillers upgraded the abrasion resistance of PP.

Because these fillers were reinforcing, it was expected that the tensile strength of PP would be remarkably improved by their presence. This expectation was borne out by the observations shown in Figure 1(d,e), especially for the breaking loads [Fig. 1(e)]. In fact, the observed decreases in yield strength [Fig. 1(d)] were understandable along the arguments of Bishop,[10] that is, that the material was at higher strength before the application of stress. As stress was applied to it, the material slacked in tension, with the yield strength decreasing.

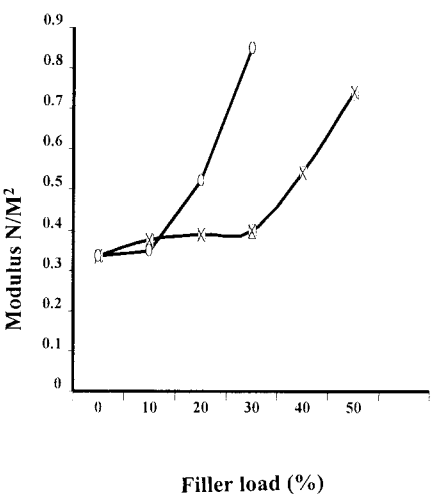
The reinforcing actions of these two filters were also evident in the observations depicted in Figures 1(f,g) and 3. As shown in Figure 1(f), for example, the reduction in flexibility as filler loading increased has been observed for similar systems.[7] In general, the enhancement in the tensile properties of PP observed in this investigation is explained, at least in part, by the postulates of Bueche[11] and Flemmert.[12] These authors believed that the filler particles, as it were, tie chain bundles together by filling interstitial voids, thereby restricting molecular slip-



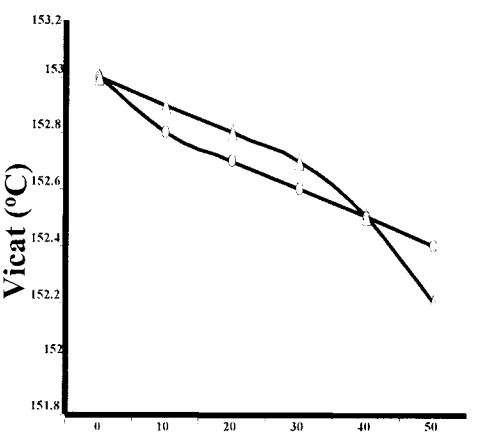
(e). Effect of filler load on the breaking properties of PP.



(f).Effect of the filler load on the strain percent



(g).Effect of filler load on the elastic modulus of PP



(h). Effect of filler load on the softening temperature of PP

Figure 1 (*Continued from the previous page*)

page on application of tensile force. Furthermore, the filler particles assist in the distribution of any induced stresses more equitably. This reduces the chances for craze propagation leading to failure. By this mechanism, elongation is hindered as filler loading is increased. Similar observations have been reported by Sydney and Dubois[13] and Ngene;[14] the former investigator arguing that each filler particle acts as a stress riser, thereby increasing the stress levels around itself. This leads to a premature failure in such a system.

We also feel, in addition to the arguments presented previously, that the filler actions of the two materials may have to do with surface effect. We feel that the filler surfaces form "platforms" for better alignment of chains, thereby increasing crystallinity and, hence, strength. Such chains on the filler surfaces interact more closely with the fillers by Van der Waal and dipole–dipole forces and such physical phenomena. It is also possible that the positive ions on the fillers also interact with the induced partially negative charges, along the PP chain by the +1 effect of the pendant CH_3 groups.

The observed decrease in the melting temperature of PP as filler loading increased, as shown in Figure 1(h), is difficult to explain by the arguments adduced previously. All that can be said is that the fillers appeared to be acting as impurities.

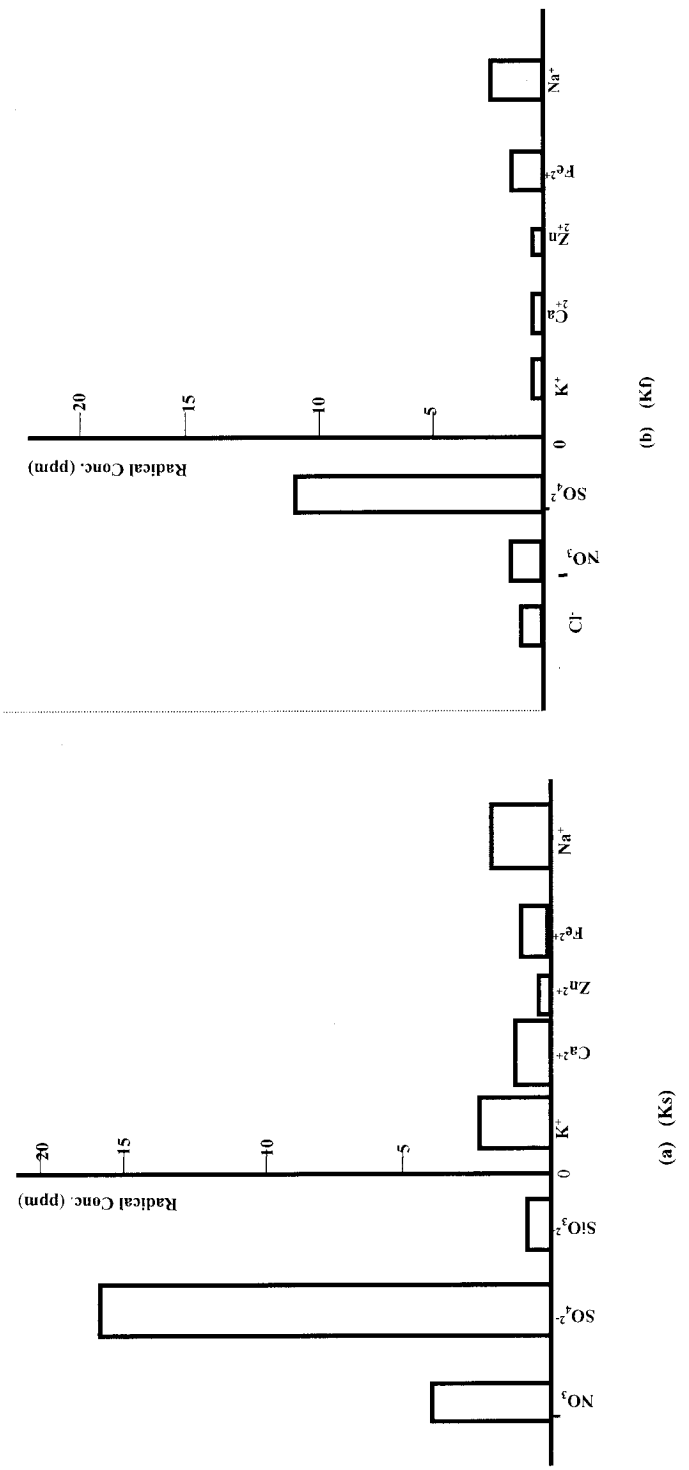


Figure 2 Chemical composition of the fillers.

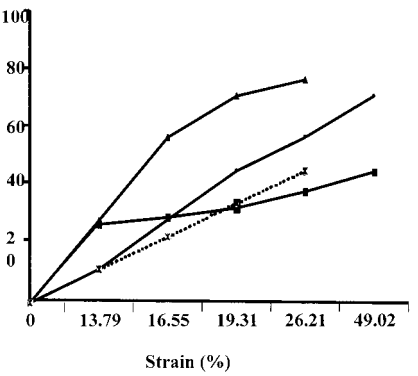


Figure 3 Some of the stress–strain data for PP-filled composites: (●) 0% Ks and Kf, (■) 30% Ks, (▲) 40% Ks, (◆◆◆) 50% Ks, and (☒) 10% Ks.

CONCLUSIONS

The following conclusions were drawn from this investigation:

- Ks and Kf function as reinforcing fillers for PP; impact strength, tensile strength at break, surface hardness, abrasion resistance, and flexural rigidity are dramatically enhanced.
- Incorporation of these fillers slightly reduces the heat distortion temperature of ELPROP.
- Thus, a useful outlet is established for otherwise useless solid waste.

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