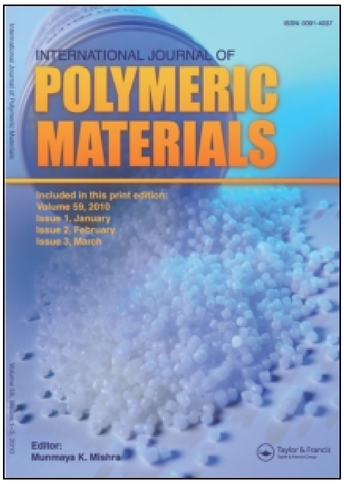

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Mechanical Properties of Isotactic Polypropylene/Wood Flour Composites

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Tensile and impact properties of wood flour filled isotactic polypropylene composites are studied at 0–70 wt% filler levels. Tensile modulus showed an increase whereas tensile strength and breaking elongation decreased with increasing filler content. Restriction in molecular mobility or deformation of polypropylene by wood flour particles accounted for the modulus increase and elongation decrease while increased amorphization and introduction of stress concentration points explained the decrease in tensile strength. Izod impact strength increased up to a critical filler concentration beyond which the value registered a slow decrease. Surface treatment of wood flour with a titanate coupling agent LICA 38 modifies further the mechanical properties. Better dispersion of filler particles upon surface treatment was indicated by SEM studies.

KEY WORDS Mechanical properties, interphase modification, coupling agent, stress concentration, amorphization, impact strength.

1. INTRODUCTION

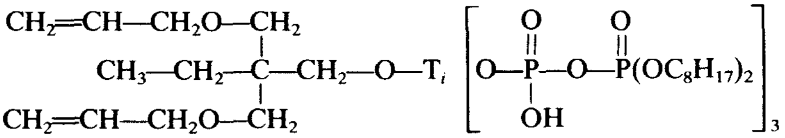
Use of wood flour (WF) as an inexpensive filler of organic origin in thermoplastic as well as thermoset compositions is reported in the literature.[1–5] Its use in thermoplastic composites is increasing since the ease in processability is retained while accompanied loss in strength properties is only moderate. Increase in modulus, izod impact strength and decrease in tensile strength with increase in filler content was observed in isotactic polypropylene (i-PP) based composites.[6] Ishihara *et al.*[2] reported decreased molding shrinkage, increased stiffness and creep resistance with WF concentration in i-PP. Since the interphase of two phase systems plays a decisive role in controlling the mechanical properties,[7] surface treatment of the filler with coupling agents is recommended[8–10] to improve the interaction of the matrix and the inclusion. The coupling agents are believed to enhance wetting of the filler by the polymer so that better dispersion is achieved. Modification of rheology and improvement in processability is also reported.[9,10]

In this paper we report the effect of addition of WF on the tensile and impact properties of i-PP at WF concentrations 0–32 vol%. Tensile data were analysed on the basis of some theoretical predictive equations. Scanning electron microscopic (SEM) studies were performed to analyse the state of dispersion of WF in i-PP. Effect of surface modification of WF by a titanate coupling agent LICA 38 on the above composite properties has also been studied.

2. EXPERIMENTAL

2.1. Materials

Isotactic polypropylene (i-PP) Koylene MOO30 (MFI 10 and density 0.89 cm^{-3}) supplied by Indian Petrochemicals Corporation Ltd. was employed in this work. Wood flour (WF) of Kail Wood (Botanical name—Pinus Excelsa) which is a class of soft wood of Northwestern Himalayan origin[11] was differentiated by sieving and particles of size 150–300 μm (density 1.32 $g \cdot cm^{-3}$) were used. The coupling agent[9] used to modify WF surface was neopentyl (diallyl) oxy, tri(dioctyl pyro-phosphato) titanate, LICA 38 of chemical structure



supplied by Kenrich Petrochemicals Inc. USA.

2.2. Method of composite preparation

Vacuum dried WF was surface treated in a high speed mixer for 10 min using 0.4 wt% of LICA 38. WF (both untreated and treated) was incorporated into i-PP on a two roll mill at 433 ± 2 K for 10 minutes to prepare thin sheets of composites containing 5–70 phr (3–32 vol%) WF. Sheets 1 mm and 3 mm thick were compression molded using several plies of these primary sheets at 493 ± 2 K and 27.6 MPa pressure for 5 minutes. Tensile specimens were machined from molded sheets 1 mm thick, impact specimens were cut from sheets of 3 mm thickness.

2.3. Measurements

Tensile properties of the dumb-bell shaped specimens were estimated on an Instron (Model 1121) at an initial crosshead separation of 5 cm and crosshead speed 20 mm/min. following ASTM D-638 test procedure. Izod impact strength values were measured using notched samples on an FIE instrument (Model IT-0.42) according to ASTM D-256 test procedure.

Cryogenic fractured surfaces of the composites were used for Scanning Electron Microscopic (SEM) studies using a Cambridge Stereoscan (Model S4-10) electron microscope. The fracture surfaces were sputter coated with gold prior to scanning.

3. RESULTS AND DISCUSSION

3.1. Tensile properties

Tensile properties such as tensile modulus, breaking strength and elongation-at-break, were evaluated from the stress-strain curves (not shown). The tensile parameters are presented in Figures 1–7 as the variation of the ratio of the property of the composite (subscript c) to that of the unfilled i-PP (subscript p) against WF concentration ϕ_F (volume fraction).

The relative elastic modulus E_c/E_p of the composites varies with ϕ_F in a manner shown in Figure 1. The modulus of composites with untreated WF

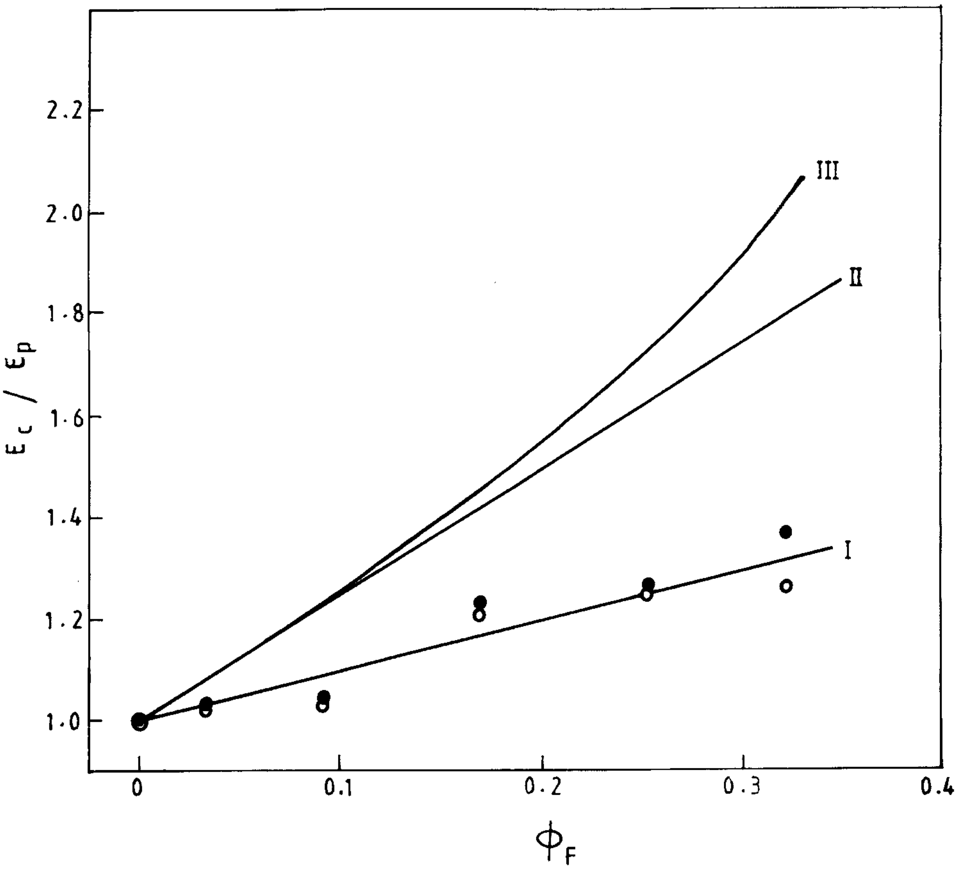


FIGURE 1 Variation of relative tensile modulus (E_c/E_p) of i-PP/WF (0) and i-PP/WF/LICA 38 (●) composites versus WF volume fraction ϕ_F. Curves I, II and III represent Einstein equation without adhesion, Einstein equation with adhesion and modified Kerner equation, respectively.

increases with ϕ_F. Surface treatment of WF with the coupling agent increases the modulus values marginally. Figure 1 also presents comparison of the modulus data with some of the predictive models of two-phase composites which take into account the shape, packing fraction, and adhesion between the filler and the matrix polymer. Curve I is the plot of Einstein equation without adhesion[12,13] [Eq. (1)], curve II shows Einstein equation with adhesion[12,13] [Eq. (2)], and curve III represents modified Kerner equation[12,14] [Eq. (3)], for filled polymer composites with Poisson

$$E_c/E_p = (1 + \phi_F) \tag{1}$$

$$E_c/E_p = (1 + 2.5\,\phi_F) \tag{2}$$

$$E_c/E_p = \frac{15(1-\gamma_p)}{8-10\,\gamma_p} \cdot \frac{\phi_F}{1-\phi_F} \tag{3}$$

ratio γ_p of i-PP assumed as 0.35. The data for both untreated and treated WF-filled composites fit the Einstein equation without adhesion, while Einstein's model with adhesion and Kerner's equation exhibited higher values. The increase in modulus of the untreated WF-filled composites indicates that addition of WF increases rigidity of i-PP through introduction of restrictions in the mobility of the polymer molecules. This restriction is, however, not very high due to the absence of any specific chemical interaction between polar WF and nonpolar i-PP. Surface treatment of WF with LICA 38 improves the interaction only marginally.

Relative elongation-at-break ($\varepsilon_c/\varepsilon_p$) showed a significant decrease with increase in ϕ_F, Figure 2, the decrease being quite rapid up to $\phi_F \simeq 16\%$ and quite slow beyond this point. This declining trend of elongation is maintained in the composites with surface treated WF, the values are, however, higher than the former case up to $\phi_F = 16\%$. The prediction by Nielsen model[7,12] [Eq. (4)] for elongation with perfect adhesion

$$\varepsilon_c/\varepsilon_p = 1 - \phi^{1/3} \tag{4}$$

shows slightly higher values than the data with untreated WF. In the composites with treated WF the values lie above the theoretical curve upto $\phi_F = 0.16$ and values beyond lie below. The decrease in elongation of i-PP in the presence of WF once again indicates hinderance by WF to molecular mobility or deformability of i-PP. In the treated WF-filled composites a plasticizing/lubricating effect by the coupling agent accounts for the lesser reduction of deformability of i-PP. Similar results were reported by other works also.[10,15,16]

The modulus data indicated increase in rigidity of i-PP/WF composites which may be due to the restriction imposed by WF on molecular mobility of i-PP. Surface treatment of WF by LICA 38 marginally enhances this rigidity. Elongation data also confirms the hinderance to deformability of i-PP by WF whereas lubricating/plasticizing type of behaviour was shown by the coupling agent in the surface treated WF composites. In the particulate filled composites[7,12] due to poor stress transfer at the filler-polymer inter-phase discontinuity is created which generates weak structure. Agglomeration of the filler particles and dewetting of the polymer at the interphase aggravate the situation by creating stress con-

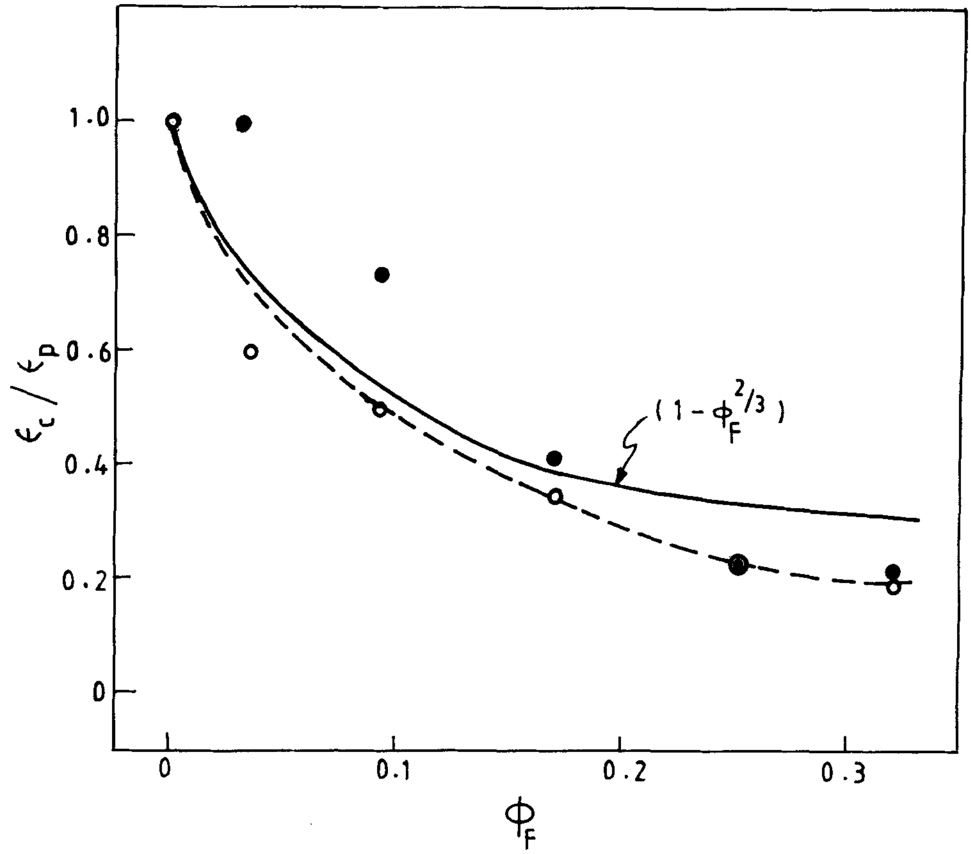


FIGURE 2 Plot of relative strain-at-break ($\varepsilon_c/\varepsilon_p$) of i-PP/WF (0) and i-PP/WF/LICA 38 (●) composites against ϕ_F. Dotted curve represents variation of experimental data. The solid curve represents predicted behaviour according to Eq. (4).

centration points which account for the weakness in the composite. For any significant contribution in the composite properties the dispersed phase should meet the important requirements of maintenance of continuity in the structure and/or interfacial adhesion. The subsequent analysis throws more light into these aspects.

3.2. Analysis of tensile stress data

In order to evaluate the weakness in the structure generated through discontinuity of stress transfer in these two-phase composites the tensile strength data were compared with some of the most commonly used theoretical predictive models, Eq. (5)–(8). A comprehensive review is given in references 16 and 17. In these expressions

$$\sigma_c/\sigma_p = (1-\phi)S \tag{5}$$

$$\sigma_c/\sigma_p = (1-\phi^{2/3})S' \tag{6}$$

$$\sigma_c/\sigma_p = (1 - K\phi^{2/3}) \tag{7}$$

$$\sigma_c/\sigma_p = \exp(-a\phi) \tag{8}$$

σ_c and σ_p denote the tensile stress of the composite and the matrix polymer, respectively. Equations (5) to (7) are based on the relationship of area fraction to volume fraction of the dispersed phase[18,19] and describe no adhesion type of composite structure. The parameter S in the first power law expression [Eq. (5)] was used in analogy to Nielsen equation[20] [Eq. (6)] to describe stress concentration at the filler-polymer interphase arising out of discontinuity in stress transfer. For "no stress concentration effect" the value of S is the maximum which is unity; the lower the value of S the greater the stress concentration effect (or poorer the adhesion). The weightage factor K in Eq. (7) accounts for the adhesion quality between the continuous phase and the dispersed phase;[21] the lower the value the better the adhesion. The extreme case with $k = 0$ represents unfilled polymer. For spherical inclusions in the extreme case of poor adhesion[18] K assumes a value of

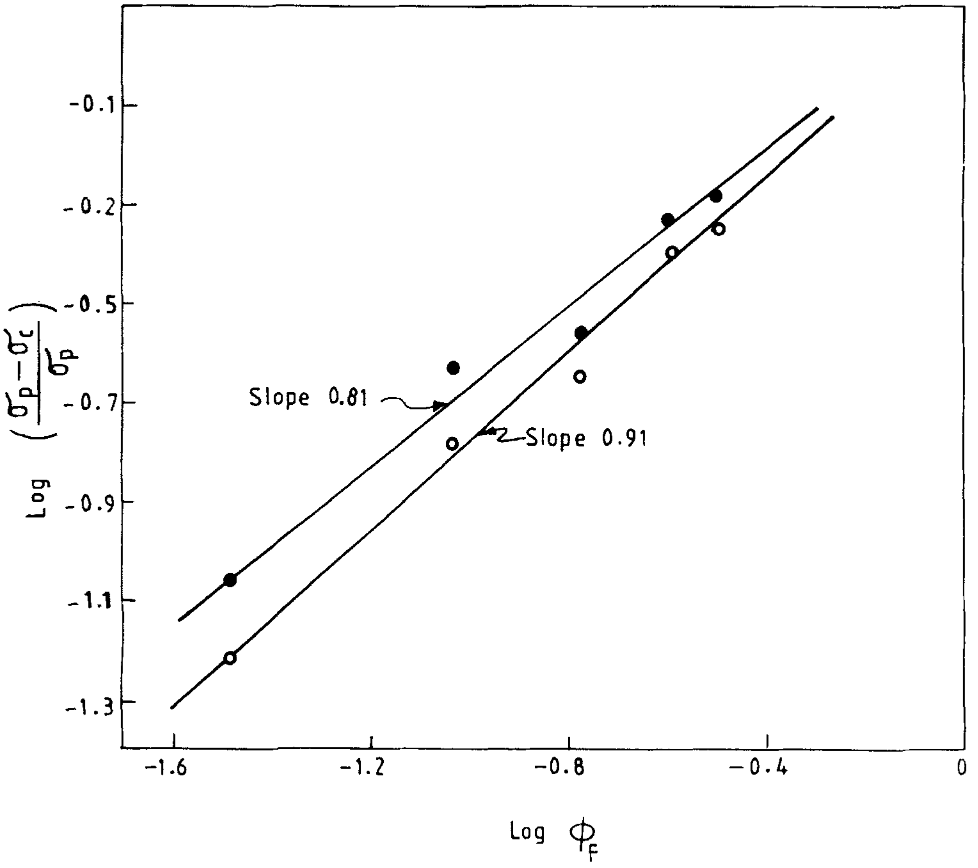


FIGURE 3 Variation of log $(\sigma_p - \sigma_c)/\sigma_p$ of i-PP (0) and i-PP/WF/LICA 38 (●) composites as a function of log ϕ_F.

1.21. The porosity model [Eq. (8)] assumes the dispersed phase equivalent to pores or voids in metals and ceramics[22] as well as in polymer blends[18] and composites,[23] which do not exhibit any influence on the composite mechanical properties due to nonadhesion at the interphase boundary. It was shown[18] that the parameter a describes stress concentration, the higher the value of a the higher the stress concentration.

Variation of log $[(\sigma_p - \sigma_c)/\sigma_p]$ vs. log ϕ obtained from tensile stress data is plotted in Figure 3 in order to assign first or two-thirds power-law to i-PP/WF system. The slope of this plot gives the value of the power-law exponent according to Eq. (5) or (6) in the absence of the parameter S or S'. It is noted that with untreated WF the slope was close to first power-law whereas upon surface treatment the slope was closer to two-thirds power-law, the values being 0.9 and 0.8 respectively. In the analysis of the strength[24] and yield stress[25,26] of composites better applicability of first power-law than the fractional power-law was observed. The latter was found to describe well the filled polymer composites.[4,7,27] The present analysis is thus subjected to further modification when the stress concentration parameters described in Eqs. (5)–(8) are included.

Some degree of agreement can be achieved between the tensile stress data and the first and fractional power-law models as well as the porosity model [Eq. (5), (6) and (8)] by using suitable values of S, S' or a, at each individual filler concentration, Table I. According to the first power-law model the stress concentration values are less than unity while according to the porosity model the value is greater than unity, both indicating significant weakness or discontinuity in the structure. The S' values according to Nielsen's model are, however, close to unity.

Figures 4–7 compare tensile stress data with the theoretical models. With the

TABLE I

Values of stress concentration parameters S [Eq. (5)], S' [Eq. (6)] and a [Eq. (8)] in i-PP/WF and i-PP/WF/LICA 38 composites

WF vol%	S^a	S'	a
0	—	—	—
3.3	0.9712 (0.9412)	1.047 (1.014)	1.911 (2.863)
9.3	0.9147 (0.8358)	1.043 (0.953)	2.014 (2.990)
16.9	0.9298 (0.8664)	1.113 (1.036)	1.525 (1.943)
25.4	0.7798 (0.6913)	0.970 (0.860)	2.134 (2.609)
32.2	0.7850 (0.7046)	1.003 (0.901)	1.958 (2.294)
Mean value	0.8523 (0.7745)	1.037 (0.953)	1.908 (2.539)

[a] Mean taken for the last four values i.e., for composites with WF content > 3 vol%.
Values in the parentheses are for surface treated WF/i-PP composites.

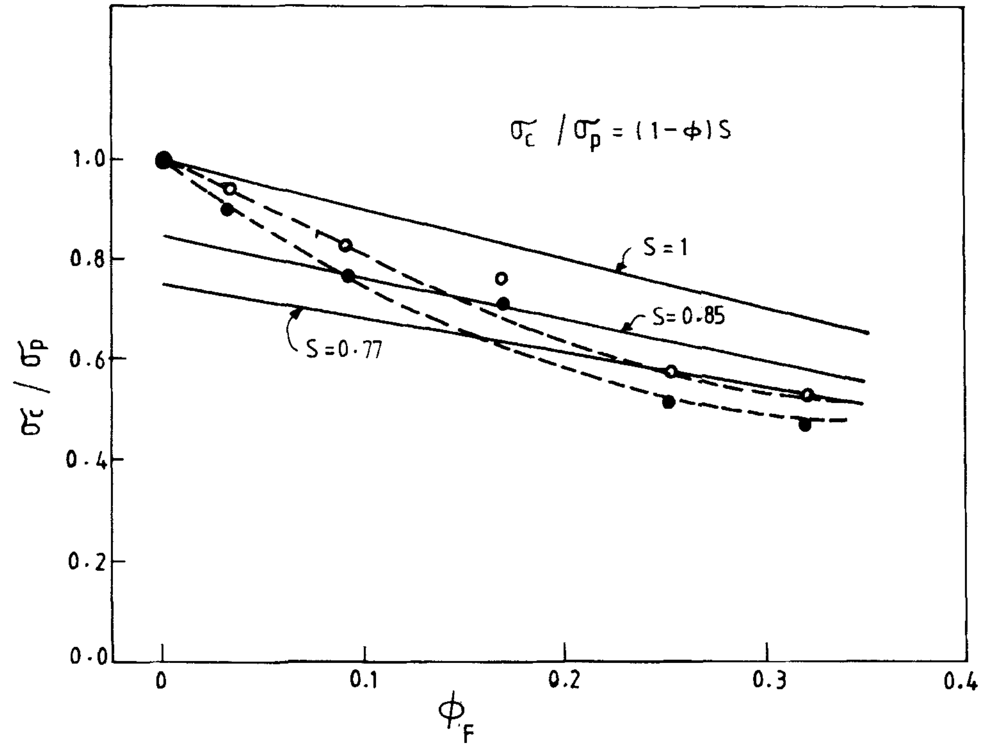


FIGURE 4 Relative tensile stress (σ_c/σ_p) of i-PP/WF (○) and i-PP/WF/LICA 38 (●) composites against ϕ_F. Dotted curves represent variation of experimental data. Solid curves represent predicted behaviour according to Eq. (5) with S values indicated.

first power-law expression [Eq. (5)], the data with untreated WF show poor fit with the theoretical curve, Figure 4, with $S = 0.85$ which is the mean of the values with more than 3.3 vol% WF, Table I. The value of $\phi_F = 3.3\%$ was close to unity. The overall implication is that the system changes from the state of "low stress concentration effect" to "significant stress concentration effect" beyond 3.3% WF. Upon surface treatment the tensile stress values were lower than in the previous case and the data lied scattered around the curve with $S = 0.77$, the mean of the values beyond 3.3% WF, implying also introduction of significant stress concentration.

Nielsen model, Eq. (6), exhibited good agreement with the data in the entire range of WF concentration studied, Figure 5. The mean value of S' for the untreated WF/i-PP composites was 1.04 while for the treated WF-filled composites the value was 0.95. This latter value, however, indicates inappreciable stress concentration in the surface treated WF/i-PP composites.

Nicolais and Narkis model, Eq. (7), shows lower values than the data with both untreated and treated WF, Figure 6, indicating a degree of better adhesion with the untreated fillers with $K = 0.85$ similar to other reports.[4,6] In the treated WF/i-PP composites the values are closer to the theoretical expression indicating

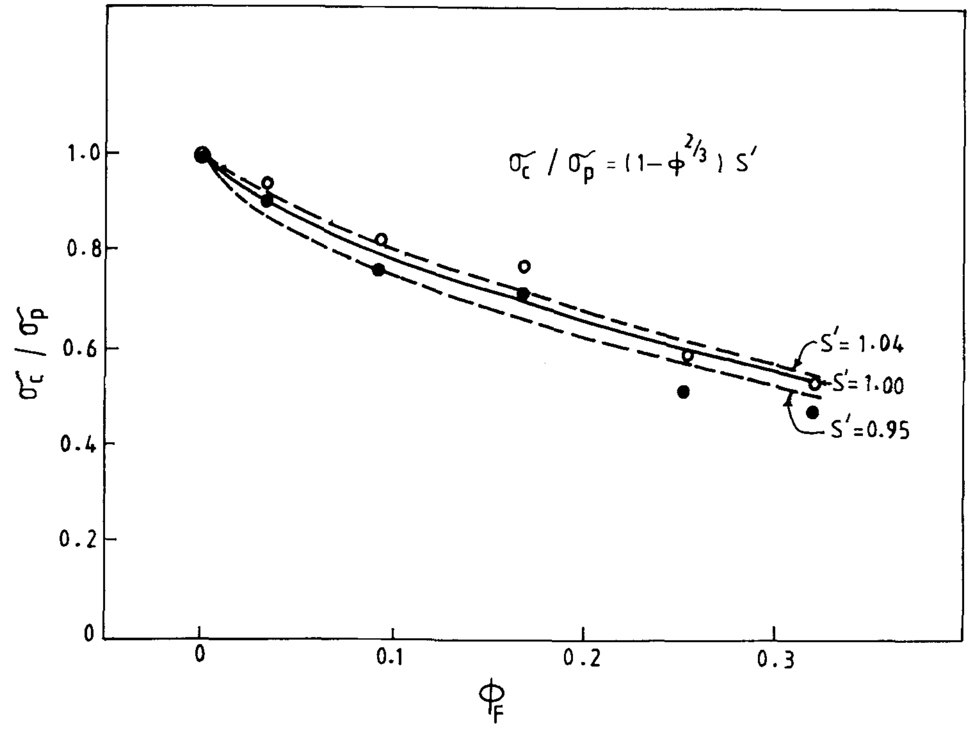


FIGURE 5 Variation of (σ_c/σ_p) of i-PP/WF (○) and i-PP/WF/LICA 38 (●) composites versus ϕ_F. Solid curve represents Eq. (6) with $S' = 1$, dotted curves represent the same with S' values indicated.

almost no adhesion ($K = 1.15$) due probably to the lubricating/plasticizing effect[10,15,16] of the coupling agent.

Finally, the data with both untreated and treated WF-filled composites showed good agreement with the porosity model, Eq. (8), with a values 1.90 and 2.54, respectively, Figure 7. Thus, stress concentration was observed in i-PP/WF composites, the value being greater in the treated WF-filled composites. Occurrence of stress concentration according to porosity model was reported in other works[16,17] as well.

It may be worthwhile to emphasize that i-PP is a hydrocarbon polymer with very low degree of intermolecular interaction forces. Isotactic polymer chains fit into helical type of crystal structure and the polymer derives its mechanical strength from crystallinity.[28] Presence of additives such as fillers, blending polymers, or miscellaneous other modifying agents impose restrictions on i-PP molecules in fitting into the crystal structure. The overall crystallinity may thus decrease which in turn also decreases the strength properties. If the decrease in crystallinity is compensated for by increased interaction of the additive with i-PP through any means, the strength of the neat polymer may be retained. Therefore, the nature of the interphase between the additive and i-PP could play an important role[7] in determining the strength properties of the filled i-PP composites.

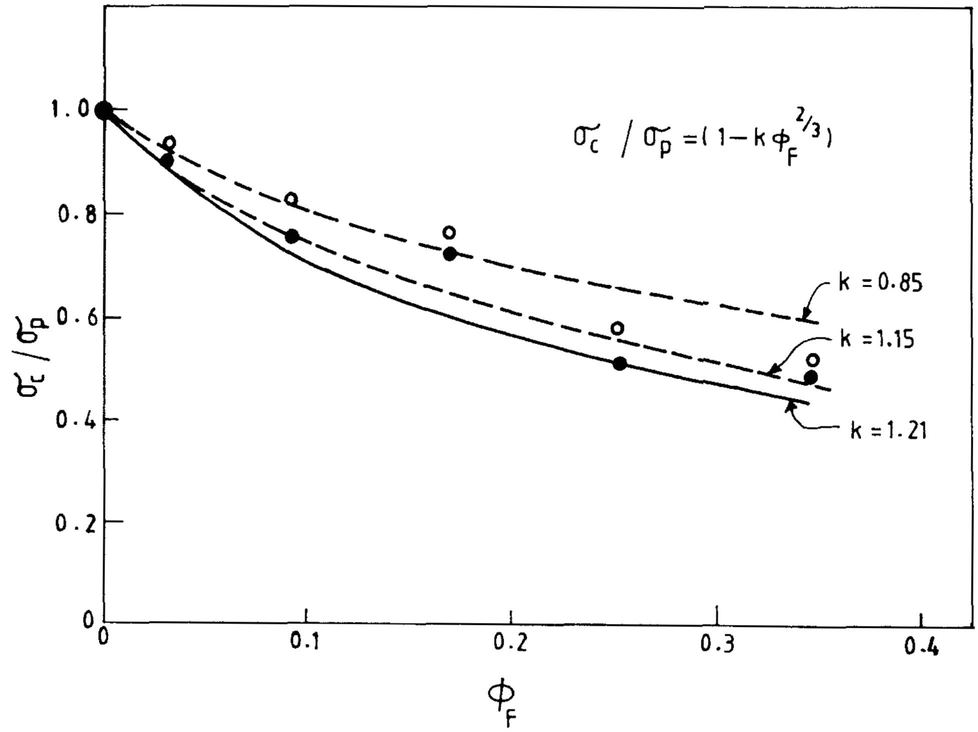


FIGURE 6 Plot of (σ_c/σ_p) of i-PP/WF (○) and i-PP/WF/LICA 38 (●) composites against ϕ_F. Solid curve is the predicted behaviour according to Eq. (7) with $k = 1.21$, dotted curves represent the same with k values indicated.

In the i-PP/WF composites the tensile strength and breaking elongation decreased while tensile modulus increased with WF concentration. WF particles were coarse (size 150–300 μm) and porous so that some degree of physical interaction with i-PP was possible. Presence of WF decreased the crystallinity of i-PP, Table II, which would tend to decrease the strength properties. The physical interaction, on the contrary, functions in enhancing strength properties of i-PP. The resultant of these two opposing factors will determine mechanical properties in these composites. Enhancement was registered in tensile modulus which is a property determined at low deformations which may remain insensitive to microflaws, if any, in the composite structure. Tensile strength, on the other hand, is determined at relatively high solid displacements which may magnify any weakness in the structure reducing the stress transfer. In that case despite some degree of physical interaction of i-PP with WF the overall effect is that the tensile strength showed a decrease. In addition, formation of increased amount of amorphous i-PP in the presence of WF, Table II, would also reduce the tensile strength. Similar results were reported in other works[16] also. Reduction in elongation of i-PP in the composites is due to increased rigidity in the presence of WF which restricts the deformation of i-PP.

Surface treatment of WF with LICA 38 marginally increases the modulus values due to enhanced i-PP-WF interaction which surpasses the decrease in

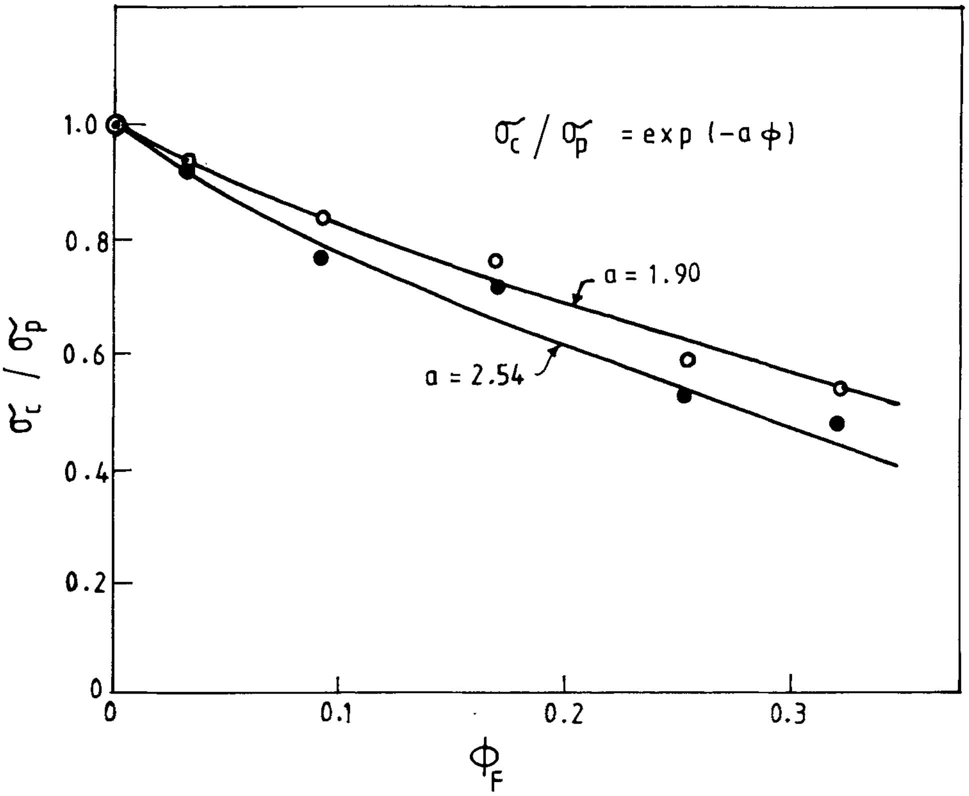


FIGURE 7 Variation of (σ_c/σ_p) of i-PP/WF (○) and i-PP/WF/LICA 38 (●) composites against ϕ_F. The curves are predicted behaviour according to Eq. (8) with a values indicated.

crystallinity brought about by this enhanced interaction, Table II. Tensile strength values decreased further due to this decrease in crystallinity. Furthermore, the lubricating/plasticizing type of effect by the coupling agent also is responsible in decreasing the tensile strength, at least, in part. The elongation values, although decreased with WF content, were slightly higher than the untreated WF-filled composites which substantiates the lubricating/plasticizing effect[10,15,16] of the coupling agent.

3.3 Impact strength

Variation of relative Izod impact strength values are shown in Figure 8. The strength increases up to $\phi_F = 3.3\%$ showing a maximum here, the value then decreases with ϕ_F, the decrease beyond $\phi_F = 10\%$ being quite small so that at the maximum WF content ($\phi_F = 32\%$) the value is 0.75 times that of unfilled *i*-PP. The increase in the impact strength at low ϕ_F may be attributed to the formation of smaller size of spherulites[29] of i-PP as well as ability of the polymer to absorb more energy by the increased extent of amorphous i-PP. Further increase in ϕ_F decreases the deformability of i-PP coupled with the formation of stress

TABLE II

Values of crystallinity (%) of i-PP in i-PP/WF composites estimated from X-ray diffraction measurements

Sample composition (vol% Filler)	X-Ray crystallinity[a] (%)	
	i-PP/WF	i-PP/WF/LICA 38
i-PP	72	72
i-PP/WF, 3.3%	67.1	62.8
i-PP/WF, 9.3%	62.1	58.1
i-PP/WF, 16.9%	65.0	52.0
i-PP/WF, 25.4%	47.3	27.3
i-PP/WF, 32.2%	29.9	21.3

[a] X-Ray Crystallinity data were taken from reference 29. The values were calculated following the method used by Sotton *et al.*[30]

concentration points at the WF-i-PP inter-phase. Similar results were observed in i-PP/$CaCO_3$ and i-PP/mica composites.[16,31] Composites containing surface treated WF exhibited similar trend of variation of impact strength, the values were, however, slightly higher. This can also be attributed to the lubricating/plasticizing effect of the coupling agent similar to other studies.[10,15,16]

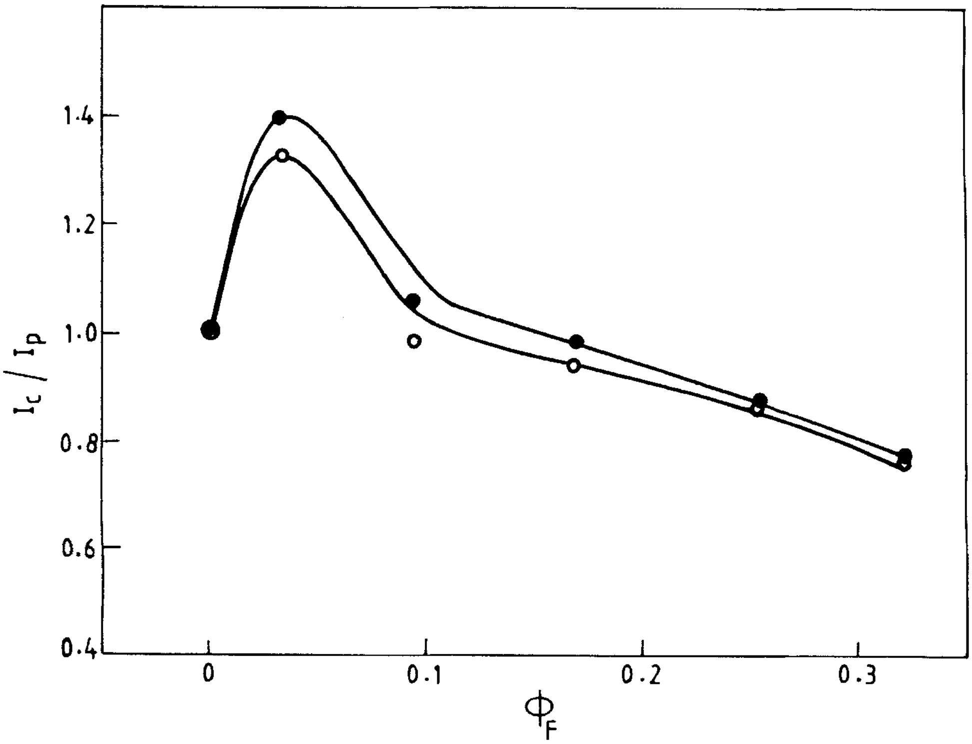
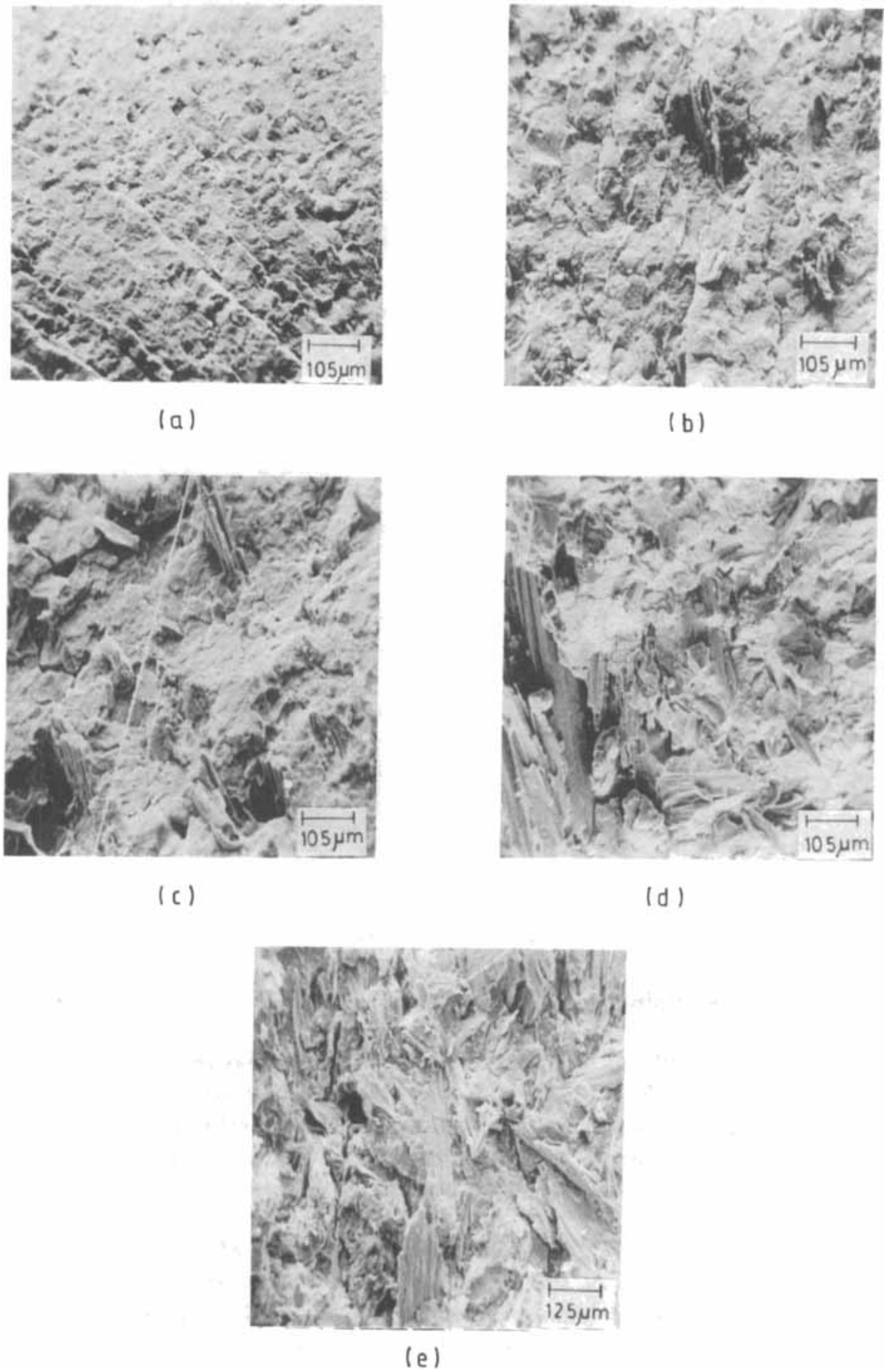


FIGURE 8 Relative Izod impact strength (I_c/I_p) of i-PP/WF (○) and i-PP/WF/LICA 38 (●) composites versus ϕ_F.

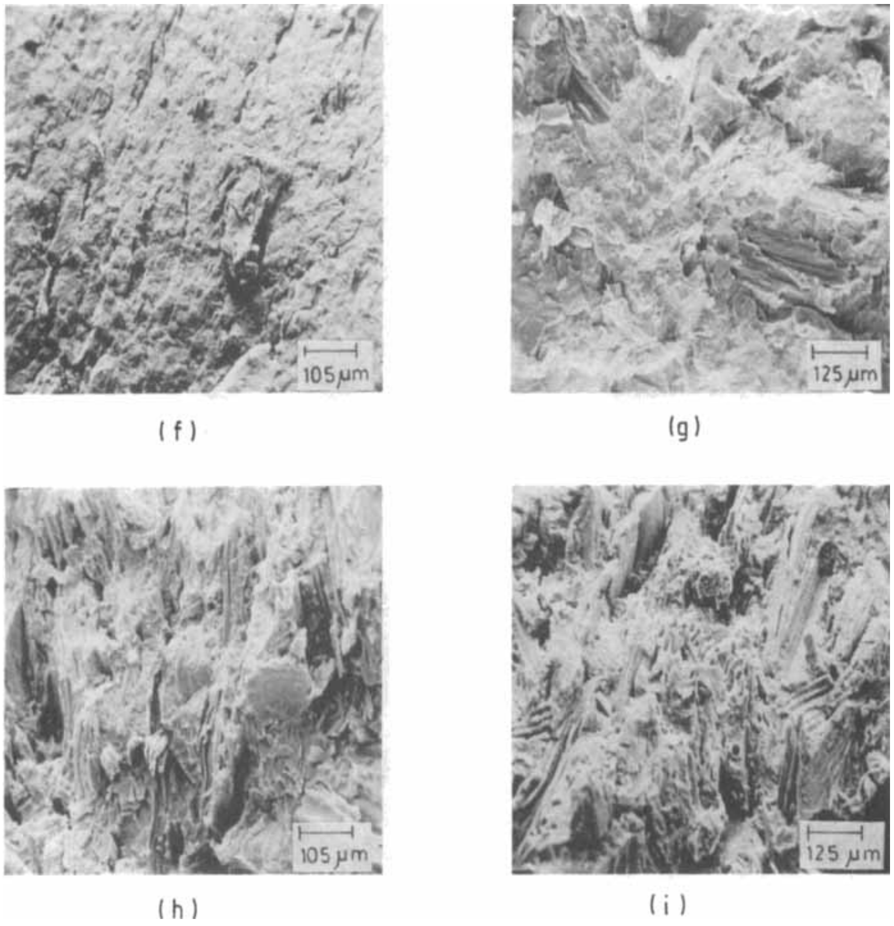


(a) (b)

(c) (d)

(e)

FIGURE 9 Scanning electron micrographs of i-PP (a) and i-PP/WF composites at varying ϕ_F: (b), 0.03; (c) 0.09; (d), 0.17; and (e), 0.32. The micrographs of i-PP/WF/LICA 38 composites at corresponding WF levels are shown in (f), (g), (h) and (i), respectively.



(f) (g) (h) (i)

FIGURE 9 (*Continued*).

3.4. State of dispersion

SEM photomicrographs of the i-PP/WF composites are shown in Figure 9(a)–(i). Poor dispersion of WF in i-PP matrix was observed with untreated WF, Figure 9(a)–(e), where the fracture surfaces were nonuniform and random with uneven voids. Better dispersion of WF in i-PP matrix with less voids together with uniform and smoother fracture surfaces were observed with treated WF-filled composites, in particular at low ϕ_F (i.e., up to 9.3 vol% WF), Figure 9(f) and (g). This indicates increased interaction of WF with i-PP which also modify the crystallization behaviour of the latter. At higher levels of ϕ_F, however, the fracture surface becomes nonuniform with uneven voids, Figure 9(h) and (i), similar to the composites with untreated WF. This may be attributed to the lack of wetting by i-PP of WF particles which were coarse and porous. The coupling agent in these filler levels would predominantly function as lubricating/plasticizing agents as stated earlier.

CONCLUSIONS

Incorporation of WF brings about significant modifications in the mechanical properties of i-PP. Tensile strength and elongation-at-break exhibited decreasing trends whereas tensile modulus increased with WF content. Izod impact strength showed an initial increase with a maximum at $\phi_F = 3.3\%$, the value decreased beyond this ϕ_F, the decrease being quite small compared to unfilled i-PP. At the highest filler level of 32.2 vol% (70 wt%) the impact strength was ~0.75 times that of unfilled i-PP. This implies the potentiality of WF as a very useful and inexpensive filler for i-PP with a possibility of going to still higher filler level without much lost to other useful mechanical properties. WF introduces stress concentration in i-PP beyond a critical $\phi_F = 3.3\%$.

Composite properties were further modified when WF surface was treated with a titanate coupling agent LICA 38. Interaction of WF with i-PP increased causing increased amorphization of i-PP. Tensile modulus showed inappreciable increase over the values with untreated WF. Tensile strength values continued to exhibit declining trend with WF content and the values were slightly lower than those of untreated WF filled composites due to increased amorphization of i-PP and/or lubricating/plasticizing effect of the coupling agent. Tensile strain-at-break and izod impact strength showed very similar trend as in the previous case although the values were somewhat higher. This substantiates the lubricating/plasticizing type of function by the coupling agent as stated earlier.

Better dispersion of surface treated WF into i-PP was exhibited by the SEM studies.

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