Composite Materials of Radiation-Treated Coconut Fiber and Thermoplastics

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Synopsis

Following our previous work using fibrous biomass byproducts for radiation-treated composite materials, polypropylene and two different kinds of PVC have been compounded with chopped coconut fiber (coir). Preirradiation of coir has been applied together with some crosslinking additive to achieve chemical bond between thermoplastics and fibrous biopolymer. Effect of addition of 10–50% coir to PVC and PP on the processability was monitored by Brabender plastograph. Dynamical mechanical analysis (DMA) data as well as tensile and impact strength of these coir composites have not been found superior to that of the starting thermoplastics. Considering, however, coconut fiber as a cheap filler, composites with acceptable tensile and impact strength could be produced with coir content as high as 30%.

INTRODUCTION

Composite making is an alternative way of better utilization of fibrous biomass. Hard cellulosic fibers are seen today as potential component of fiber-reinforced plastics.[1] Significant effort is made to develop composite plastics using natural hard fibers such as henequen,[2] jute,[3-6] and similar ones. Radiation grafting is applied to modify preferably these hard cellulosic fibers.[4,5]

In our previous works we produced thermosetting plastic composites using coconut fiber in phenol–formaldehyde and unsaturated polyester resins.[7,8] It has been found that with coconut fiber reinforcement the best properties of glass-fiber-reinforced plastics cannot be achieved, above all because of the big difference between the modulus of elasticity of the glass and natural fibers. Very good BMC (bulk moulding compound) type press material can be prepared, however, in which a great part of the chopped glass fiber is exchanged for coconut fiber. On the other hand, we observed promising results on polypropylene (PP) filled with pretreated wood fiber.[9]

In a followup of these works we aimed to apply chopped coconut fiber in typical thermoplastics like plasticized PVC, hard PVC, and PP.

EXPERIMENTAL

Materials

Coconut fibers have been imported from Sri Lanka in form of coarse fiber rope. Elemental fibers were 10–20 cm long, their diameter being 0.28 ± 0.17 mm, tensile strength 160 ± 80 M N/m^2, modulus of elasticity 2.40 ± 0.62

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G N/m^2. These hard fibers were cut into short ones (1–2 mm) and fractionated by passing through sieves of 1.6, 1.0, and 0.6 mm meshes.

Two sorts of PVC have been applied, both being suspension-type products of Borsod Chemical Works (BVK), Hungary: an impact-resistant PVC, Ongrovil S 5058 and another for plasticized PVC, Ongrovil S 5070, having K values 58 ± 1 and 70 ± 1, respectively.

The polypropylene applied in our experiments was Tipplen H 601, product of the Tisza Chemical Works (TVK), Hungary. It has been used in powdered form, its MFI (melt flow index) being 2.0 g/10 min at 230°C, 2.16 kp load.

As reactive additive, an unsaturated polyester resin type POLIKON R (made by Nitrokemia Works, Hungary) has been applied. Its viscosity was 450–550 mPa s at room temperature, its acid number 26 mg KOH/g. Other ingredients used: ULTRAMOLL III, a saturated polyester type plasticizer of Bayer AG, BCD-10, a Ba–Cd stabilizer of BVK, and Irganox 1010, a conventional stabilizer of CIBA [pentaerythrityl-tetrakis-3/3,5-di-*tert*-butyl-4-hydroxyphenyl(-propionat) as well as TBPB: *tert*-butyl-perbenzoate of FLUKA, all used in their original commercial form].

Methods

The radiation treatment of chopped coconut fiber consisted of a preirradiation process in air at room temperature by a Co-60 gamma source, with 10^4 Gy dose at a dose rate of 200 Gy/h.

The processability of the thermoplastic composites was evaluated in a Brabender plastograph type Plasticorder PLE 651. In the preheated kneading chamber—type W 30 H—of 30 mL volume, the torque can be registered as a function of time, together with the actual temperature and the number of revolutions/min.

Tensile strength and impact-tensile strength of the composites has been measured on standard testing machines equipped with electronic sensors (INSTRON 1195 and CEAST Fractoscope). A DuPont Dynamical Mechanical Analizer 981 has been used for the determination of dynamical mechanical data, such as the real component of the complex modulus of elasticity (E') and transition temperature (T_g).

RESULTS

Coconut Fiber in Plasticized PVC

The plastograph, which records the torque during plastic kneading, gives valuable information about the energy required in mixing and molding of thermoplastics. In Figure 1, a series of Brabender plastograms are shown as recorded during the kneading of plasticized PVC samples containing various amounts of coconut fibers without any particular treatment. After putting in the preheated (170°C) kneading chamber, the composite "dry blend" absorbs quite a high amount of energy ($M_{max} = 10 + 20$ N m) before getting molten. The torque after 6 min of kneading (M_6) can be considered as significant and close to equilibrium energy required for plastic transformation. These torque values (M_{max}, M_6) are shown in Table I, together with the composition of samples. Beside the saturated polyester type plasticizer (Ultramoll III) and

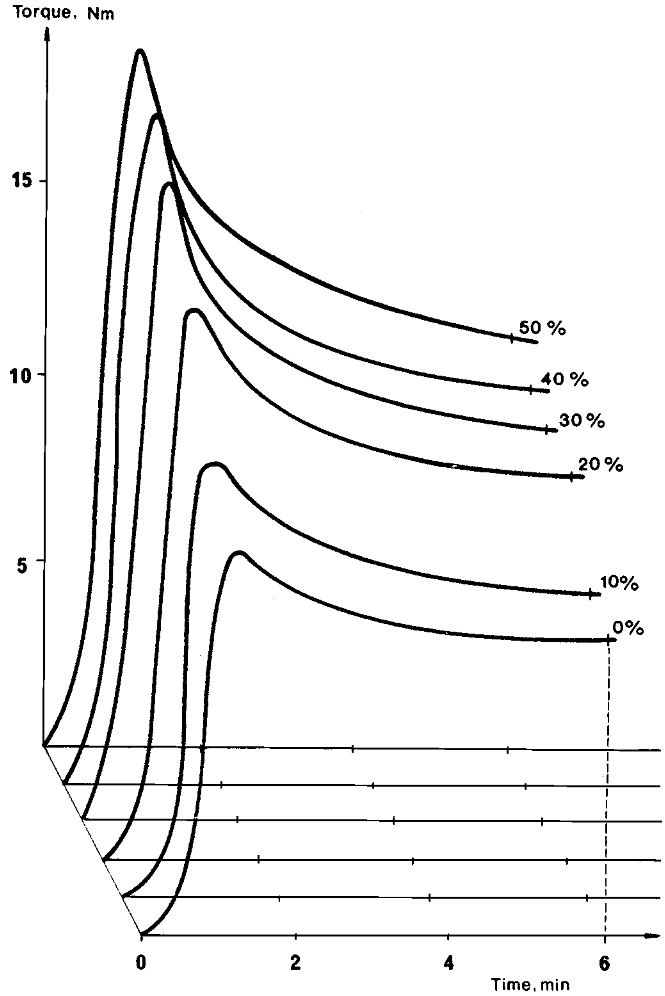

Fig. 1. Brabender plastograms of coconut-fiber-filled plasticized PVC. Fiber content of the compositions on curves: 0, 10, 20, 30, 40, and 50% (mass), respectively.

conventional stabilizers, samples contained a reactive additive, the Polikon R, which was intended to serve as reactive adhesive between main components. In the first half of the series in Table I, *t*-butyl-perbenzoate (TBPB) has been applied to the initiation. In the second half of experiments the initiation was expected to occur through peroxy groups and radicals created by gamma preirradiation of coconut fiber.

It is seen in Table I that, as the fiber content grows, M_{max} and M_6 torques are increasing. In case of irradiated fiber, however, the increase of these torque values is significantly less (Fig. 3 as compared to Fig. 2), which means a better workability of such composites. Mechanical properties of plasticized PVC–coconut fiber composites are seen in Table II—measured in somewhat bigger quantities of new samples. Tensile strength and impact tensile strength of current (unmodified) plasticized PVC is quite high. Modification of such PVC with coir results in a filler effect, but no reinforcement has been found. The

TABLE I
Torque Rheometry (at 170°C) of Coconut-Fiber-Filled Plasticized PVC

	Composition (g)									Torque (N m)	
No.	PVC S 5070	Coconut fiber	Irrad. coconut fiber	Ultramoll III.	Polikon R	TBPB	BCD-10	Irganox 1010	Epox. soya	M_{max}	M_6
326	23.44	0	0	4.69	0	0	0.47	0.10	1.17	9.7	7.1
325	20.27	0	0	4.05	4.05	0.20	0.41	0.10	1.01	10.2	7.9
320	18.99	1.90	0	3.80	3.80	0.19	0.38	0.10	0.95	11.8	8.2
321	17.86	3.37	0	3.57	3.57	0.18	0.36	0.10	0.89	14.8	10.3
322	16.85	5.06	0	3.37	3.37	0.17	0.34	0.10	0.84	17.0	10.5
323	15.96	6.38	0	3.19	3.19	0.16	0.32	0.10	0.80	17.8	10.6
324	15.15	7.58	0	3.03	3.03	0.15	0.30	0.10	0.76	18.3	11.0
356	23.44	0	0	4.69	0	0	0.47	0.10	1.17	11.9	8.8
350	20.27	0	0	4.05	4.05	0	0.41	0.10	1.01	4.2	3.7
351	18.99	0	1.90	3.80	3.80	0	0.38	0.10	0.95	6.0	5.8
352	17.86	0	3.57	3.57	3.57	0	0.36	0.10	0.89	9.3	7.5
353	16.85	0	5.06	3.37	3.37	0	0.34	0.10	0.84	8.3	7.6
354	15.96	0	6.38	3.19	3.19	0	0.32	0.10	0.80	8.0	6.9
355	15.15	0	7.58	3.03	3.03	0	0.30	0.10	0.76	11.3	8.6

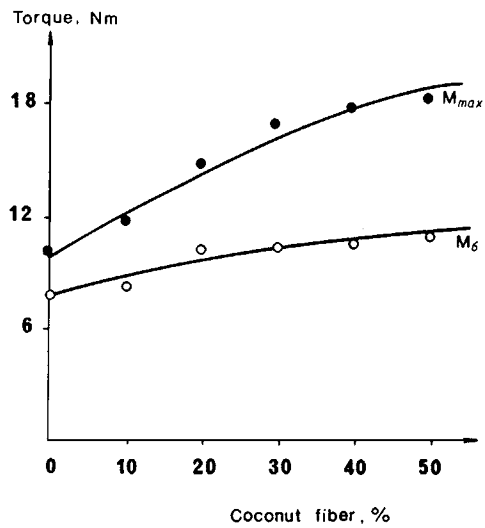

Fig. 2. Maximum torque M_{max} and torque in the 6th minute, M_6, of plasticized PVC, filled with untreated coconut fiber.

radiation treatment of the fiber produces however acceptable mechanical properties below 20% of fiber content.

Dynamic mechanical analysis of plasticized PVC–coconut fiber composites offers two kind of information: E', related to the modulus of elasticity in function of temperature, and E'', the maximum position of which indicates transition temperatures, very important from the point of view of practical temperature limits of application of any thermoplastics. Two typical sets of curves are shown in Figure 4. Evaluation of a series of such DMA curves are

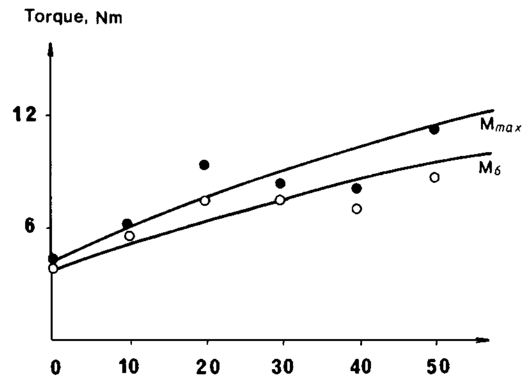

Fig. 3. Maximum torque M_{max} and torque in the 6th minute, M_6, of plasticized PVC, filled with radiation-treated coconut fiber.

TABLE II
Mechanical Properties of Coconut-Fiber-Filled Plasticized PVC

	Composition (g)[a]					Properties	
No.	PVC S 5070	Coconut fiber	Irrad. coconut fiber	Ultramoll III.	Polikon R	Tensile strength (N/mm^2)	Impact-tensile strength (kJ/m^2)
186	81.5	0	0	16.3	0	25.16	67.33
151	63.4	6.3	0	12.7	12.7	16.53	30.62
152	59.7	11.9	0	11.9	11.9	20.03	9.20
153	56.3	16.9	0	11.3	11.3	22.30	10.02
154	53.2	21.3	0	10.7	10.7	12.48	3.62
155	50.5	25.2	0	10.1	10.1	17.56	16.57
187	67.7	0	0	13.5	13.5	22.22	25.55
180	63.4	0	6.3	12.7	12.7	17.12	29.41
181	59.7	0	11.9	11.9	11.9	12.83	31.58
182	56.3	0	16.9	11.3	11.3	19.73	18.81
183	53.2	0	21.3	10.7	10.7	13.49	7.82

[a] Stabilizers not taken into account.

seen in Table III. T_{g_1}, the most significant transition temperature, seems to be somewhat increased in presence of coconut fiber, E' indicating modulus of elasticity at different temperatures is almost the same for coconut-fiber-filled and unfilled plasticized PVC.

Coconut Fiber in Hard PVC

Hard PVC types modified with some acrylic copolymers have an important share of market of commodity plastics. A contemporary formula of such compound contains an acrylic copolymer as impact modifier, e.g., Paraloid K 120 N, a polyethylene wax as processing aid, e.g., PE 520 as well as standard conventional stabilizers and costabilizers. Such a formula has been modified with a reactive UP resin and chopped coconut fiber, as is seen on

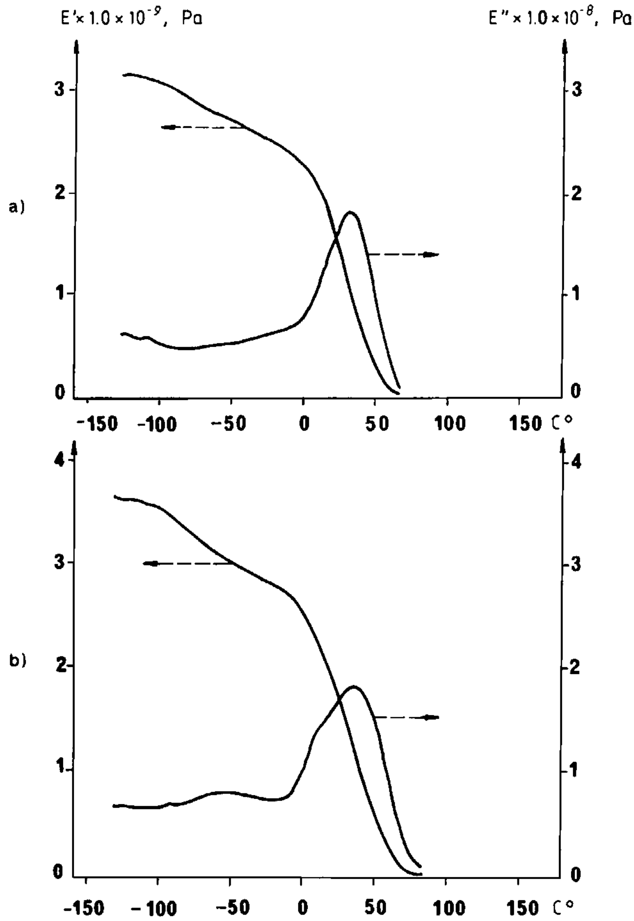

Fig. 4. Dynamical mechanical analysis of: (a) plasticized PVC; (b) composite of plasticized PVC (100) + irradiated coconut fiber (40); E' = real part of the complex modulus of elasticity; E'' = imaginary part of the complex modulus of elasticity.

TABLE III
Dynamical Mechanical Analysis of Coconut-Fiber-Filled Plasticized PVC[a]

(g/100 g PVC) No.	Fiber content Untreated	Irrad.	T_{g1} (°C)	E_1'	T_{g2} (°C)	E_2'	E_{-20}'	E_0'	E_{24}'
186	0	0	31	1.18	5	2.31	2.97	2.49	1.55
151	10	0	38	1.11	−24	2.90	2.86	2.61	1.85
152	20	0	42	1.41	−18	3.05	3.08	2.79	2.18
153	30	0	50	1.20	10	2.63	3.12	2.87	2.26
154	40	0	38	1.11	−19	2.33	2.33	2.19	1.66
155	50	0	46	1.31	−29	3.00	2.92	2.72	2.16
187	0	0	32	1.06	—	—	2.49	2.27	1.48
180	0	10	38	1.15	10	1.20	2.73	2.44	1.76
181	0	20	28	1.14	—	—	2.62	2.30	1.34
182	0	30	41	1.15	−23	2.76	2.74	2.54	1.90
183	0	40	35	2.24	—	—	2.78	2.52	1.77

[a] T_{g1}, T_{g2} = glass transition temperatures; E_1', E_2' = real part of the complex modulus of elasticity at temperatures of T_{g1} and T_{g2}, respectively, in 10^9 Pa units; E_{-20}', E_0', E_{24}' = the same, at temperatures −20, 0, and 24°C.

TABLE IV
Torque Rheometry of Coconut-Fiber-Filled Hard PVC (at 190°C)

	Composition[a] (g)								Torque (N m)	
No.	PVC S 5058	Coconut fiber	Irrad. coconut fiber	Paraloid K 120 N	PE 520 wax	Ca-stearate	Polikon R	TBPB	M_{max}	M_6
310	28.50	0	0	0.86	0.14	0.14	0	0	19.0	15.9
311	25.60	2.56	0	0.77	0.13	0.13	0.51	0.02	18.5	16.9
312	23.18	4.64	0	0.70	0.12	0.12	0.93	0.05	18.2	17.7
313	21.20	6.40	0	0.64	0.11	0.11	1.27	0.06	21.5	20.0
314	19.53	7.80	0	0.59	0.10	0.10	1.56	0.08	20.4	19.0
315	18.11	9.10	0	0.54	0.09	0.09	1.81	0.09	22.4	20.2
316	16.87	10.12	0	0.51	0.08	0.08	2.02	0.09	22.0	17.5
331	25.60	0	0	0.77	0.13	0.13	0.51	0.02	17.4	12.3
332	23.18	0	2.56	0.70	0.12	0.12	0.93	0.05	15.5	11.6
333	21.20	0	4.64	0.64	0.11	0.11	1.27	0.06	18.0	13.4
334	19.53	0	6.40	0.59	0.10	0.10	1.56	0.08	23.2	13.5
335	18.11	0	7.80	0.54	0.09	0.09	1.81	0.09	25.6	15.5
336	16.87	0	9.10	0.51	0.08	0.08	2.02	0.10	28.0	15.6

[a]Stabilizers (BCD, Irganox 1010) not taken into account.

Table IV. This time all the filled samples received some peroxy initiator. As is shown in Table IV, the quasiequilibrium torque (M_6) is again lower in the case of preirradiated coconut fiber as compared to the same composition with untreated fiber. It means an easier plastic transformation process—e.g., extrusion, of treated composites filled up to 30% natural hard fiber.

The mechanical properties of these hard PVC composites are shown on Table V. The up-to-date formula of an impact-resistant PVC represents quite

TABLE V
Mechanical Properties of Coconut-Fiber-Filled Hard PVC

	Composition[a]					Properties	
No.	PVC S-5058	Coconut fiber	Irrad. coconut fiber	Paraloid K 120 N	Polikon R	Tensile strength (N/mm^2)	Impact-tensile strength (kJ/m^2)
190	95.0	0	0	2.9	0	53.13	34.51
191	85.2	8.5	0	2.6	1.7	48.36	14.36
192	77.2	15.4	0	2.3	3.1	41.77	8.48
193	70.6	21.2	0	2.1	4.2	41.28	6.39
194	65.1	26.0	0	2.0	5.2	34.54	4.76
195	60.3	30.2	0	1.8	6.0	28.46	4.19
207	89.7	0	0	2.7	5.4	57.74	30.88
201	85.3	0	8.5	2.6	1.7	48.68	5.80
202	77.3	0	15.5	2.3	3.1	33.47	3.03
203	70.7	0	21.2	2.1	4.2	32.89	3.74
204	65.3	0	25.1	2.0	5.2	21.44	3.76
205	60.5	0	30.3	1.8	6.1	18.48	3.25
206	56.4	0	33.8	1.7	6.8	33.05	4.09

[a]Mass %; stabilizers and PE-wax not taken into account.

TABLE VI
Torque Rheometry of Coconut-Fiber-Filled PP (at 200°C)

	Mass composition (g)					Torque (N m)	
No.	Tipplen H-601	Coconut fiber	Irrad. coconut fiber	Polikon R	TBPB	M_{max}	M_6
301	30.0	0	0	0	0	21.0	4.3
302	27.0	3.0	0	0	0	18.0	5.1
303	24.0	6.0	0	0	0	18.0	9.1
304	21.0	9.0	0	0	0	16.0	9.3
305	18.0	12.0	0	0	0	17.0	9.4
306	15.0	15.0	0	0	0	19.0	13.0
341	23.9	0	3.0	3.0	0.15	22.2	9.0
342	20.9	0	6.0	3.0	0.15	21.8	10.2
343	17.9	0	9.0	3.0	0.15	20.0	7.2
344	14.9	0	12.0	3.0	0.15	24.3	8.2

high mechanical properties, which cannot be improved by simple addition of chopped coconut fiber. Especially the impact resistance is deteriorated by adding 30% coconut fiber and more. However, 3–5 kJ/m^2 of impact strength still represents a useful range of resistance, enough for many applications in the building industry like PVC tiles or exterior cladding for houses. The high value of tensile strength diminishes moderately by adding coconut fiber: even with 20–30% of fiber tensile strength of 20–30 N/mm^2 can be produced. Dynamical mechanical analysis of filled hard PVC samples does not show any significant change in the most important characteristics as compared to the unfilled hard PVC.

TABLE VII
Dynamical Mechanical Analysis and Mechanical Properties of Coconut-Fiber-Filled PP[a]

	Fiber content (%)		DMA data						Tensile strength	Tensile-impact strength
No.	Untreated	Irrad.	T_{g1} (°C)	E'_1	T_{g2} (°C)	E'_2	E'_{-20}	E'_{24}	(N/mm^2)	(kJ/m)
101	0	0	19	2.34	−21	3.38	3.37	2.06	36.3	46.49
102	10	0	17	2.31	−21	3.19	3.17	1.99	26.7	9.56
103	20	0	11	2.40	−23	3.10	3.06	1.88	21.8	6.89
104	30	0	17	2.12	−31	2.90	2.80	1.88	22.0	4.63
105	40	0	17	2.30	−23	2.89	2.87	2.07	24.0	5.91
106	50	0	17	2.43	−37	3.23	3.06	2.19	15.9	4.81
107	0	10	19	2.34	−27	3.22	3.13	2.04	26.8	9.17
108	0	20	19	2.30	−30	3.29	3.17	2.06	24.4	5.67
109	0	30	18	2.49	−29	3.41	3.31	2.27	26.5	5.72
110	0	40	21	2.31	−21	3.00	2.99	2.19	24.9	4.69

[a] T_{g1}, T_{g2} = glass transition temperatures; E'_1, E'_2 = real part of the complex modulus of elasticity at temperatures of T_{g1} and T_{g2}, resp., in 10^9 Pa units; E'_{-20}, E'_{24}: the same, at temperatures −20° and 24°C.

Coconut Fiber in Polypropylene

Torque rheometry of polypropylene filled with chopped coconut fiber is shown on Table VI. The maximum as well as equilibrium torque (M_{max} and M_6) are all on an acceptable level up to 40% filler content. This means again an easy workability on calander, extruder or even on injection molding machine. Table VII shows that mechanical properties are not improved: The particularly high impact strength of PP is lowered very much in all the cases. Tensile strength remains in a useful range up to 40% filler content. Preirradiation of coconut fiber does not influence very much all these phenomena. The same has been found for dynamical-mechanical properties: E'_{-20} and E'_{24}, indicating modulus of elasticity at $-20°C$ and at room temperature, remain practically the same, as all the other DMA data.

CONCLUSIONS

Short-cut coconut hair as a typical hard cellulosic fiber has a good chance to be applied in composites with commodity thermoplastics such as plasticized PVC, hard PVC, and PP. Workability by thermoplastic processing such as extrusion, calandering, etc., is not deteriorated if hard fiber content of these composites does not exceed 40% (by mass), and suitable processing aids are used. As a reactive additive, a small amount of unsaturated polyester resin has been added together with a moderate-dose radiation treatment of fibers. Our concept was to apply radiation treatment as an initiation method of graft copolymerization to be carried out during the thermoplastic processing such as extrusion and injection molding, i.e., in the processing machine itself. A similar advantageous processing possibility has been elaborated by us for composites of PP and wood fiber.[9] A series of different composite materials have been described recently[10] using radiation treatment of the fibrous natural component such as wood saw dust, bamboo, bagasse of sugar cane, etc.

Up-to-date formulations of the commodity plastics of today without fiber component, but with advanced additives, e.g., impact modifiers, stabilizers, processing aids, are showing excellent mechanical properties, which can hardly be improved by simple adding of hard cellulosic fibers. In the present work it has been found that even preirradiation-created active initiating centers on coconut fiber and simultaneous application of reactive additive (UP resin) cannot result in better bounding forces between natural fiber and thermoplastic matrix. Tensile and impact strength as well as dynamical mechanical analysis data of such fiber composites have not been found superior to that of the starting thermoplastics. Considering, however, coconut fiber as a cheap filler, composites with acceptable tensile and impact strength and other important properties could be produced with coconut fiber content as high as 30%.

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References

1. M. A. Semsarzadeh, *Plast. Eng.*, **41**(10), 47 (1985).
2. R. A. Padilla and S. A. Sanchez, *J. Appl. Polym. Sci.*, **29**, 2405 (1984).

3. R. N. Mukherjea, S. K. Pal, and S. K. Sanyal, *J. Appl. Polym. Sci.*, **28**, 3029 (1983).
4. P. Ghosh, A. R. Bandyopadhyay, and S. Das, *J. Macromol. Sci.-Chem.*, **A19**, 1165 (1983).
5. P. Ghosh and S. Kumar Paul, *J. Macromol. Sci.-Chem.*, **A20**, 169 (1983).
6. R. Mohan and S. Kishore, *J. Mater. Sci. Lett.* **2**(3), 99 (1983).
7. O. Owolabi and T. Czvikovszky, 5th TIHANY Symp. on Rad. Chem., Akademiai Kiad. Budapest, 1983. Vol. 2, pp. 799–801.
8. O. Owolabi, T. Czvikovszky, and I. Kovács, *J. Appl. Polym. Sci.*, **30**, 1827–1836 (1985).
9. T. Czvikovszky, Kunststoffe Ger. Plast., **73**(11), 710 (1983).
10. U. P. Wang, *J. Ind. Irradiation Techn.*, **1**(1), 1 (1983).

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