Properties of High-Density Polyethylene Filled with Waste Crosslinked Foam

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ABSTRACT: Crosslinked polyethylene foam is widely used in packaging and as an insulation material. Finely ground waste of such crosslinked foam mesh size 7 or particle size less than 2815 μm is used as a filler in high-density polyethylene (HDPE) of two different grades (7.5 and 21 MFI). Mechanical, thermal, and morphological properties of filled composites is studied experimentally. Waste foam powder concentration was varied up to 40% by weight basis. Impact strength of base HDPE increased by a factor of six. The overall changes in mechanical properties are similar to the crosslinking effect. It is believed that waste foam particles act as a point of entanglement with different chains of polyethylene. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 91: 110–114, 2004

Key words: crosslinking; fillers; waste foam; polyethylene (PE); composites

INTRODUCTION

Polyethylene (PE) is a commodity thermoplastic material used in a variety of applications because of its relative ease of processability, low cost, and easy availability. The properties of polyethylene also depend on the level of branching. Branched PE and copolymers of ethylene such as poly(ethylene-*co*-vinyl acetate) (EVA) are widely used in crosslinked products. Given that branching is known to promote crosslinking, it is relatively easier to crosslink branched low-density PE (LDPE) than high-density PE (HDPE).[1–3]

The crosslinking of PE reduces the chain mobility and also influences a number of other properties. Crosslinked PE forms a dense network of high molecular weight, which improves impact strength, environmental stress crack resistance (ESCR), creep, and abrasion resistance without influencing tensile strength and density to any appreciable extent. However, it increases the viscosity and decreases elongation at break.[4–10]

Crosslinking of PE, poly(propylene) (PP), or copolymer of ethylene and PP by different techniques was previously reported in the literature.[11–13] Industrial crosslinking is practiced primarily by using peroxides or by a grafted silane process crosslinked by water, although electron beam radiation and UV crosslinking have also been reported.[14,15]

Crosslinked PE becomes thermoset in nature and thus it is not possible to process waste crosslinked PE like thermoplastic material. Disposal of crosslinked waste material is a compelling environmental issue. Using waste crosslinked PE as a filler in polyolefins could be advantageous, given that sufficient compatibility between filler and matrix would exist.[8,9] There is no reported work on the use of waste crosslinked polyolefins as a filler in polyolefins. Because linear high-density polyethylene (LHDPE) is comparatively difficult to crosslink, it will be worthwhile to study the effect of waste crosslinked polyolefinic material on the properties of virgin HDPE.

EXPERIMENTAL

Commercial grades of HDPE were used. Two different grades of melt flow index (MFI), 7.5 and 21, were procured from Reliance Industries Ltd. (Mumbai, India). Waste crosslinked polyethylene foam prepared from a mixture of linear low-density polyethylene (LLDPE) and LDPE (gel content of waste foam was 75–80%) was procured from a local market, and it was pulverized into fine powder of mesh size 7 (ASTM). The density of waste filler was about 800 kg/m^3, which ensured that all the foam was crushed and there was no foaming gas or foamed cells left in the filler. A wetting agent (FINALUX G-3, an oleochemical derivative of complex ester blend) was supplied by Fine Organics Ltd. (Mumbai, India).

TABLE I
Mechanical Properties of Filled HDPE (7.5 MFI)

| Material | Tensile property | | Flexural property | | Impact strength (J/m) |
	Elongation at break (%)	Tensile strength (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	
HDPE	1801	28	16	594	63
HDPE + 10% filler	85	30	15	644	79
HDPE + 20% filler	73	30	16	649	94
HDPE + 30% filler	67	40	12	420	330
HDPE + 40% filler	68	34	07	233	425

Compounding

For all compositions, appropriate quantities of HDPE (7.5 and 21 MFI), wetting agent (0.2%), and powdered waste crosslinked polyethylene were tumble mixed for 2 h. The mixture was melt blended using a corotating twin-screw extruder (Model MP 19 PC, APV Baker, UK) having an L/D ratio of 25 : 1. The screw speed was maintained at 80 rpm and the temperature profile for compounding was 160, 175, 210, and 220°C for four zones and 230°C for the die zone. The extrudate was water cooled and pelletized. The extruded pellets were dried and molded into standard ASTM specimens at a temperature profile of 190, 210, and 230°C for the nozzle, by using a microprocessor-based injection-molding machine (Boolani Machineries Ltd., Mumbai, India). The concentration of waste foam was varied up to 40% on weight basis.

Testing

Mechanical properties

Tensile strength, elongation at break, flexural strength, and flexural modulus were measured at ambient conditions using a universal tensile testing machine (LR-50K, Lloyds Instrument, UK), according to ASTM procedures. The crosshead speed was 50 mm/min and 2.8 mm/min for tensile tests and flexural tests, respectively. The notch for impact test was made using a motorized notch-cutting machine (Polytest model 1, Ray Ran UK). Notched Izod impact strengths were measured at ambient conditions according to the ASTM D 256 method using an impact tester (Avery Denison UK) with a striking velocity of 3.46 m/s, employing a 5J striker.

Thermal properties

Thermal analysis was carried out by use of a differential scanning calorimeter (DSC; Perkin–Elmer DSC-7, Perkin Elmer Cetus Instruments, Norwalk, CT). Two consecutive scans were obtained to minimize the influence of possible residual stress in the material resulting from any specific thermal history. Samples of about 8 mg were used. A scanning rate of 10°C/min was employed for both heating and cooling cycles, under a nitrogen purge of 30 mL/min.

Vicat softening point

Vicat softening point of all the samples was measured by a Vicat softening point instrument (Davenport, UK). The specimen used for this test was a rectangular bar of dimensions (in mm) 125 × 13 × 6.5 (length × breadth × thickness). The specimen was dipped in a silicon oil bath, which was heated at the rate of 2°C/min. The oil was continuously stirred and circulated to maintain uniform temperature. A load of 1 kg was applied over the given sample through a pin (1 mm^2) placed on the specimen. The test recorded the temperature at which the pin penetrated to the depth of 1 mm. The entire operation was computer controlled. Each test was duplicated.

TABLE II
Mechanical Properties of Filled HDPE (21 MFI)

| Material | Tensile property | | Flexural property | | Impact strength (J/m) |
	Elongation at break (%)	Tensile strength (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	
HDPE	1750	23	15	689	32
HDPE + 10% filler	387	24	15	608	43
HDPE + 20% filler	83	23	13	465	47
HDPE + 30% filler	67	27	12	335	79
HDPE + 40% filler	63	22	11	328	205

TABLE III
MFI and Vicat Softening Point for Filled HDPE Systems

Composition	MFI (g/10 min)		Vicat softening point (°C)	
	HDPE (I)[a]	HDPE (II)[b]	HDPE (I)[a]	HDPE (II)[b]
Virgin	7.5	21	134	133
HDPE + 10% filler	3.6	11.5	131	130
HDPE + 20% filler	1.3	6.1	127	126
HDPE + 30% filler	0.7	3.2	121	120
HDPE + 40% filler	0.1	1.6	116	114

[a] HDPE (I), 7.5 MFI.
[b] HDPE (II), 21 MFI.

Melt flow index determination

The melt flow index (MFI) was determined using an MFI instrument (Davenport, UK). The MFI, expressed as g/10 min, was determined at 190°C and the load was 2.16 kg.

Scanning electron microscopy

The morphological characteristics were examined by SEM (Cameca, France). The specimens (thin pieces) were etched with xylene for 10–12 h and coated with gold. Xylene dissolved the matrix HDPE material at the surface.

RESULTS AND DISCUSSION

Mechanical properties

Tables I and II show the mechanical properties of blends prepared from HDPE (7.5 and 21 MFI) and waste foam. It is very clear that incorporation of waste foam does not affect the tensile strength of base HDPE to any appreciable extent. As a matter of fact it increases slightly for compositions containing waste filler up to 30%. The flexural strength appears to be unaffected to a filler concentration of 20%. At higher concentrations, it decreases slightly. Similarly, flexural modulus does not change appreciably to a filler con-
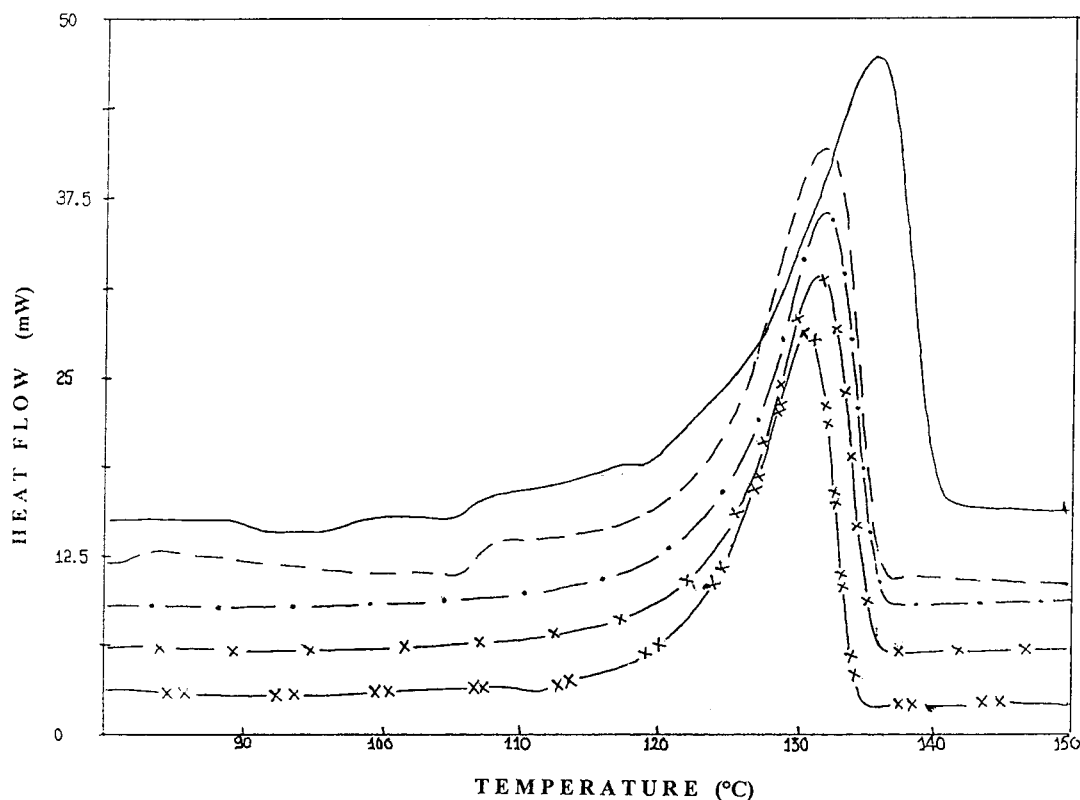

Figure 1 DSC thermograms of filled HDPE system (heating cycle): virgin (—); 10% filler (– – –); 20% filler (—·—); 30% filler (—×—); 40% filler (—××—).
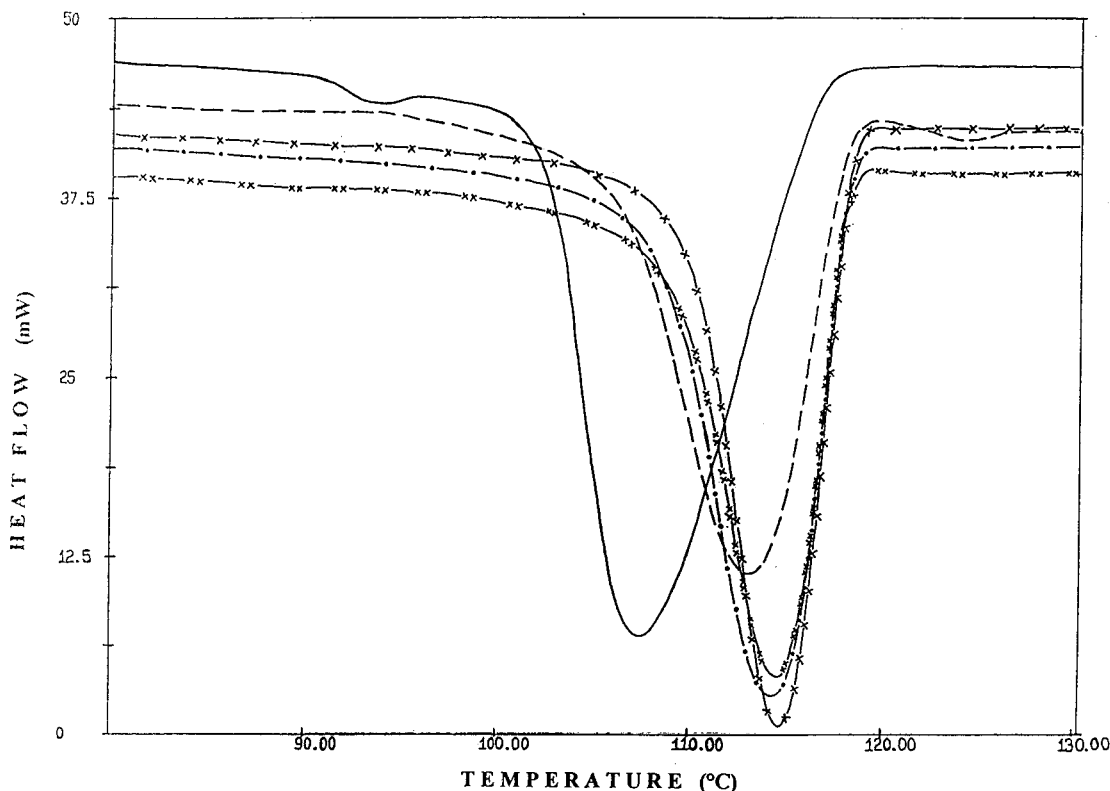
Figure 2 DSC thermograms of filled HDPE system (cooling cycle): virgin (—); 10% filler (– – –); 20% filler (—·—); 30% filler (—×—); 40% filler (—××—).

centration of 20% for HDPE of 7.5 MFI; at higher loading, however, it decreases somewhat. Flexural modulus of HDPE of 21 MFI is higher than that for 7.5 MFI. The flexural modulus of this variety of HDPE (21 MFI) decreases steadily with increase in the filler concentration. There is a significant decrease in the elongation at break for all compositions containing waste foam. The decrease is very sharp for HDPE of 7.5 MFI compared to that for HDPE of 21 MFI.

It is very clearly seen that the significant increase in impact strength of base resin is the result of blending with waste ground foam. The increase is by a factor of six for filler concentration of 40%. Inoue and coworkers[1,2] studied the selective dynamic crosslinking of PP–EPDM blends and reported that the resulting blends had higher impact strength compared to that of PP. Similarly, Loor et al.[3] studied properties of a blend of PP with ethylene vinyl acetate (EVA) and ethylene methyl acrylate (EMA) where the dispersed phase (EVA/EMA) was crosslinked *in situ*. They observed an increase in the impact properties of PP.

Table III shows variation of MFI and Vicat softening point for all compositions. It is very clearly seen that MFI decreases substantially when the percentage of filler is increased. Similarly, there is a slight decrease in Vicat softening point with increase in concentration of filler. The Vicat softening points for virgin HDPE of

two different grades as well as their blends with waste foam were not dependent on MFI.

Comparison of these properties suggests that the effect of incorporating crosslinked filler is very similar to crosslinking of the matrix. One of the reasons for such a significant increase in impact strength could be that crosslinked foam particles form a "network" by providing entanglement points between different chains of the base polymer. However, these contacts are physical in nature, unlike the chemical nature for crosslinked PE. The presence of such entanglements can influence the crystallization process; therefore DSC data were analyzed.

Differential scanning calorimetry

Figures 1 and 2 show the DSC thermograms for the heating and cooling cycles for all compositions. Table IV lists the details of these thermograms. The onset temperature for melting process (heating cycle) seems to be relatively unaffected for all compositions. However, the peak melting temperature is reduced by incorporation of waste foam particles. The enthalpy (J/g) values also seem to be reduced with increasing amount of filler. Similarly, the cooling scans show that the temperature for onset of crystallization remains relatively unaffected, whereas the peak crystallization

TABLE IV
Thermal Properties of Filled HDPE Systems

Composition HDPE/filler	Heating cycle			Cooling cycle		
	Onset (°C)	Peak (°C)	ΔH_m (J/g)	Onset (°C)	Peak (°C)	ΔH_c (J/g)
100/0	125	135	195	116	107	−217
90/10	124	131	165	117	113	−157
80/20	124	131	136	118	114	−160
70/30	124	131	147	118	114	−161
60/40	123	130	123	118	114	−138

temperature was higher when waste foam was incorporated. Also there is a decrease in the enthalpy of crystallization.

The reduction in Vicat softening point (as shown in Table III) and the DSC curves strongly suggest that incorporation of waste foam particles reduces the crystallinity of HDPE. It may even affect crystallite size and distribution. The decrease in crystallinity with an increase in the level of crosslinking of PE was previously reported in the literature.[16,17]

Scanning electron micrography

Figure 3 shows an SEM micrograph for the HDPE : foam (80 : 20) system. It is evident that waste foam particles seem to give rise to a networklike structure. Such a "network" can perhaps be attributable to entanglement of filler particles with matrix polymer chains.
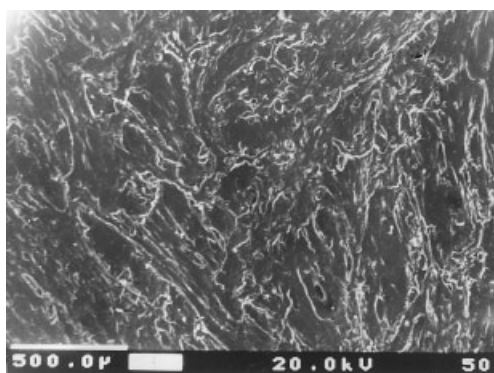

Figure 3 SEM micrograph of HDPE/filler (80 : 20) system.

CONCLUSIONS

Waste crosslinked polyethylene foam is compatible with HDPE. Addition of waste foam as a filler to virgin PE influenced the properties of base HDPE in a manner similar to crosslinking of PE, causing a significant increase in impact strength and reducing the percentage elongation at break without significantly affecting the tensile strength, flexural strength, and modulus. The waste filler particles may form a network through entanglements.

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