Properties of Recycled High Density Polyethylene from Milk Bottles

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SYNOPSIS

The effect of virgin, high-density polyethylene (HDPE)/recycled HDPE composition on the physical properties of the blends was investigated. The recycled HDPE was obtained from a postconsumer cycle of milk bottles. It was found that elongation at break was the mechanical property mostly affected by the content of recycled HDPE. Overall, however, the recycled HDPE from milk bottles was found to be a material with useful properties not largely different from those of virgin resin and thus could be used, at an appropriate concentration in virgin HDPE, for different applications.

INTRODUCTION

The accumulation of solid waste has become a major issue in recent years because of the decreasing number and space of landfills. Much attention is being given all over the world to the impact of plastics on the environment and upon their disposal, primarily because of plastic's visibility as films and containers.

In the USA, many states are increasing their reliance upon recycling, especially that of packaging, as an alternative to disposal. Three hundred ten million kg of high density polyethylene (HDPE) was used in 1986 in the USA for the production of milk bottles, making them the largest single type of plastic bottle manufactured (polethylene terephthalate soft drink bottles amounted to 290 million kg).[1] Therefore, in any plastic recycling strategy that hopes to have a significant impact on solid waste, the recovery of HDPE milk bottles plays an important role.

Several approaches to the recovery of thermoplastics have been proposed: (a) incineration, (b) pyrolysis, and (c) recycling. Although more effective than landfilling, incineration and pyrolysis are probably less effective methods of the three proposed in terms of recovering the value of the thermoplastics. Through recycling, on the other hand, these materials can be turned into many different and valuable products, thus recovering much more efficiently the value of the original material. In addition, incineration of several polymers, such as polyvinyl chloride (PVC), polyacrylonitrile (PAN), and polyurethanes (PUR), may pollute the environment with toxic fumes unless very strict and costly measures are taken.

For the recycling of polymers in general, and of HDPE in particular, to be effective, information about the properties of HDPE is required. It is well established[2-3] that recycled polymers have inferior properties compared with their virgin counterparts, and that the magnitude of this effect depends upon the polymer type and upon the number of cycles and conditions that exist during reprocessing. Various investigators have studied both the mechanism of polymer degradation during processing and the effect of degradation on performance.[4-5] There is, however, very little published information dealing with the effect of consumer use cycles on polymer properties, and there is none that we could find pertaining specifically to HDPE milk bottles.

The purpose of this investigation is to evaluate and compare some of the mechanical and physical changes in HDPE from milk bottles before and after a consumer use cycle.

EXPERIMENTAL

Materials

Virgin, high-density polyethylene used in property evaluation was Fortiflex A60-70-119 from Soltex

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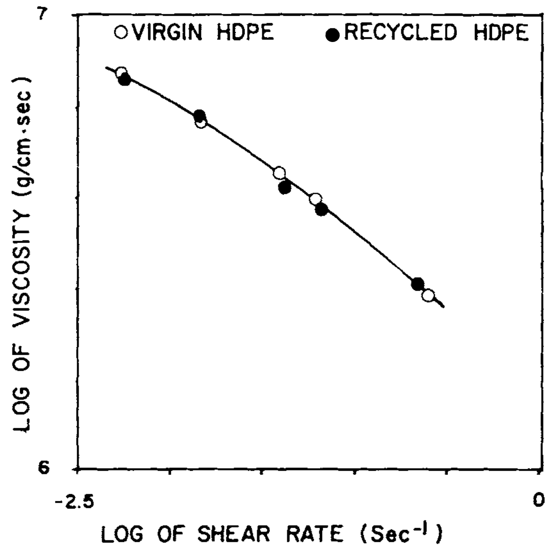

Figure 1 Flow curves of virgin and recycled HDPE.

Polymer Corp. Half-gallon bottles, blown from the same batch of resin but never filled, were obtained from Heatherwood Farms Dairy, Lansing, Michigan. Other bottles from the same batch were filled with milk and distributed through normal channels. The bottles, purchased from a local retailer, were stored in household refrigerators for about two weeks, then emptied and rinsed.

In addition, a variety of one gallon and half-gallon HDPE milk bottles were collected from consumers. Some of these were sorted by brand and others were used as a mixed master batch.

All bottles were rinsed with cold water, dried at room temperature, and flaked (after removal of caps and labels) in a Polymer Machinery Corp., lowline granulator, model 68-913. Contaminants (dirt, etc.) were separated by a cold wash and screening process, and the cleaned flakes were dried at room temperature.

Blends of recycled and virgin HDPE were prepared by mixing (for 30 min) specified percentages of the recycled master batch with virgin polymer in a propeller feed mixer.

Test Methods

Melt Flow Index determination was carried out according to ASTM Method 1238, utilizing a Ray-Ran Model 2A Digital Auto Melt Flow Indexer, at 190°C, with a 2.16 kg dead load.

GPC data were obtained using a Waters GPC instrument. The column was 300 mm long and 7.8 mm ID, filled with waters micro/styragel. The solvent was 1,2,4 trichlorobenzene at 135°C at a flow rate of 1 mL/min.

Determination of tensile properties was carried out at standard conditions[6] on specimens cut from 5 in. × 5 in. × 0.1 in. (12.7 cm × 12.7 cm × 0.25 cm) sheets, compression molded on a Carver Laboratory Press. Molding conditions were 210°C, 25,000 lbs (11,400 kg) for 6 min, followed by cooling to 50°C within 8 min. Specimens were cut into ASTM, dumbbell-shape, type I using a bandsaw and a Tensilkut machine. Samples were conditioned at 24°C, 50% RH for 2 d before testing on an Instron Model No. 114 at 2 in./min (5.1 cm/min) crosshead speed. Abrasive paper was used in the grips to prevent slippage.

Impact strength determination followed ASTM standard 256-81. Molding conditions for the 5 × 5 × 0.125 in. (12.7 × 12.7 × 0.32 cm) sheet were the same as described above. Test samples were cut to 2.5 in. (6.35 cm) long and 0.5 in. (1.27 cm) wide, and were notched with a 0.1 in. (0.25 cm) deep cut on the narrowest side (thickness). Samples were conditioned before testing as described above.

Statistical analyses were carried out using the personal computer program MSTAT with one way analysis of variance at the 95% confidence level.[7]

RESULTS AND DISCUSSION

The MFI values of three different HDPE grades used for blow molding of milk bottles ranged from 0.58 to 0.73 g/10 min. The MFI of the same grade HDPE was 0.73, 0.75, and 0.72 for the virgin polymer, unused, and postconsumer-used bottles respectively. These differences were statistically insignificant. The MFI of the different postconsumer bottle grades ranged from 0.64 to 0.75 g/10 min.

Blends of recycled and virgin resin showed no significant change in MFI for 10%, 20%, 50%, 80%, and 100% postconsumer recycled HDPE. In Figure 1, the flow curves of virgin and recycled HDPE are

Table I Molecular Weight Averages of Virgin and Once-Molded HDPE

Average[a]	Virgin	Bottle
$\bar{M}_n$	31,930	30,040
$\bar{M}_w$	236,100	238,600
$\bar{M}_z$	570,700	570,600
D_n	7.47	7.94

[a] Average of two GPC runs.

Table II Mechanical Properties of Different HDPE Compositions

	Run 1			Run 2		
Material	Modulus (M Pascal)	Tensile Strength (M Pascal)	Elongation (%)	Modulus (M Pascal)	Tensile Strength (M Pascal)	Elongation (%)
Virgin HDPE	596	33.7	69.7	579	33.0	74.0
	(88)	(1.1)	(16.5)	(35)	(0.4)	(17.5)
10% recycled	600	33.3	62.7	611	34.2	62.4
	(47)	(0.4)	(10.1)	(81)	(1.1)	(6.5)
20% recycled	689	33.1	47.2	582	34.0	51.3
	(140)	(0.4)	(8.8)	(94)	(1.5)	(12.5)
50% recycled	672	33.8	48.9	634	34.4	41.4
	(123)	(0.7)	(18.1)	(31)	(1.7)	(19.4)
80% recycled	681	34.2	35.1	703	35.0	34.6
	(164)	(1.2)	(9.2)	(125)	(1.3)	(9.4)
100% recycled	640	34.2	36.9	680	34.6	30.7
	(101)	(1.2)	(18.2)	(68)	(1.7)	(4.7)

Note: The numbers in parentheses represent standard deviation.

shown. It can be seen that no change in the flow properties occurred. The Newtonian melt viscosity, η_o, of HDPE, as well as of many other polymers, depends upon the weight average molecular weight, $\bar{M}_w$, according to:

$$\eta_o = K\bar{M}_w^{3.4-3.5} \quad (1)$$

The non-Newtonian melt viscosity and the shape of the flow curve depends also on the weight average molecular weight, as well as on the molecular weight distribution (MWD).[8] Thus, no significant changes in these parameters could be noted as a result of recycling. It has been reported in the literature[9,10] that during reprocessing, degradation and branching or crosslinking reactions may occur simultaneously, therefore some molecules may degrade while others increase in molecular weight. If such processes occur, $\bar{M}_w$ would increase while $\bar{M}_n$ would decrease, since the former depends more on the large molecules while the latter depends more on the small ones. The number dispersion index D_n ($D_n = \bar{M}_w/\bar{M}_n$), would also increase accordingly. In Table I, the molecular weight averages of the virgin HDPE obtained by GPC are compared to those of the material obtained from a molded bottle (before its use by the consumer). It can be seen that a slight decrease in the number average and an increase in the weight average molecular weights had occurred, resulting in a broadening in the molecular weight distribution. These results suggest that chain scission and crosslinking reactions probably had occurred during one molding process, although to a very low extent.

In Table II, some of the mechanical properties of

Table III IZOD Impact Strength of Mixtures of Virgin and Recycled HDPE

	Mean		SD	
Material	(ft × lb/in.)	(nt × m/cm)	(ft × lb/in.)	(nt × m/cm)
Virgin HDPE	2.522	1.346	0.160	0.085
10% recycled	2.693	1.437	0.261	0.139
16.7% recycled	2.608	1.392	0.156	0.083
50% recycled	2.409	1.286	0.138	0.074
80% recycled	2.231	1.191	0.238	0.127
100% recycled	2.201	1.175	0.144	0.077

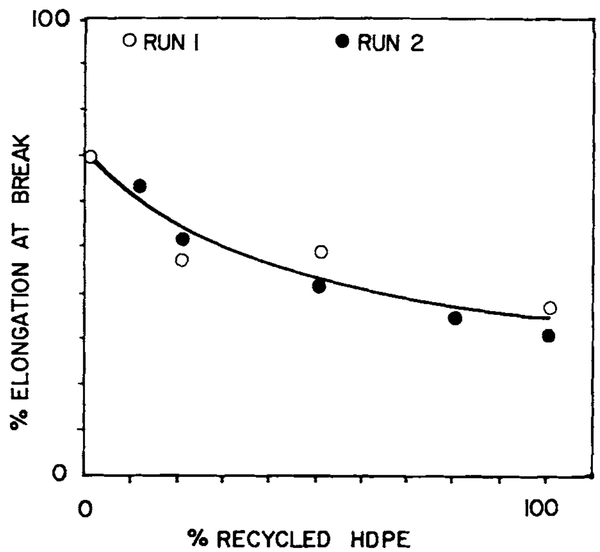

Figure 2 Effect of composition on the elongation at break of virgin/recycled HDPE mixtures.

different compositions of virgin and recycled HDPE are summarized. Whereas no effect of composition on tensile strength is seen, a slight increase in the modulus could be noted. A similar effect of increase in modulus due to degradation of polyethylene (although as a result of weathering and not of reprocessing) was found by Ram et al.[10] In Table III, the Impact Resistance of samples prepared from different compositions is summarized. It can be seen that up to almost 50% of recycled polymer, there was no decrease (actually, there was a slight unexplained increase at low concentrations) in this property. Only at the higher levels of 80% and 100% recycled material was a decrease of 10–15% in impact resistance observed. These results are in agreement with those of other investigations[9–10] showing a stiffening, coupled with embrittlement, of the polymer as a result of degradation. In some cases this change was a result of, or accompanied by, a slight change in the morphology of the polymer. Differential Scanning Calorimetry (DSC) tests run on our virgin, recycled unused and recycled-used HDPE, showed no significant differences in melting range or enthalpy. DSC however is probably not sensitive enough to detect small changes in morphology. In Figure 2, the effect of composition on the elongation at break is shown. It can be seen that the higher the percentage of the recycled HDPE, the lower the elongation at break. Miltz and Narkis[10] and Ram et al.[9] have found that elongation at break was the mechanical property most affected by degradation. The results of the present study are therefore similar to their findings.

Overall, the recycled HDPE from milk bottles is a material with useful properties not largely different from those of the virgin resin. As such, it is a promising raw material for a variety of applications. As recycling grows, more of this material will be available at prices significantly less than those of virgin resin, and also less than offgrade resins. Manufacturers can do their share toward alleviating the nation's solid waste crisis by utilizing this recycled material at an appropriate percentage, wherever it is appropriate and available.

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