Durability of Recycled Polyethylene for Geotextile Purposes

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Abstract

In recent years the incorporation of waste plastic into fibres intended for geotextiles has attracted interest as aesthetic requirements are generally not important. However the addition of waste material to virgin polymers raises questions of durability. In this paper the addition of waste HDPE to PP to produce fibres that were subsequently subjected to accelerated thermal ageing is described. Compared to PP fibres containing virgin HDPE the fibres containing the recycled HDPE had poor durability. However, addition of a recycling stabiliser package (Recyclossorb 550) resulted in much improved durability albeit at the expense of drawability.

1.Introduction

Recycling of polymer waste has received much attention in recent years due to rise in environmental concerns [1]. One problem with polymer wastes is that they generally contain pigments and additives that make their regeneration into other articles aesthetically less appealing. One possible use for recycled plastics is their incorporation in to geotextiles (technical textiles intended for civil engineering and construction industry) since in these applications aesthetic requirements are not usually important. Of the polymers that are used for geotextiles the most common is polypropylene due to its low cost and resistance to acid and alkaline soils, although it is not normally used in conditions where it is likely to experience high mechanical loadings due to its tendency to creep[2].

One problem that is likely to be encountered when incorporating polymer wastes into polypropylene based geotextiles is their effect on long term durability. Polymer waste may have already experienced degradation in the environment and the addition of these wastes into virgin materials may adversely affect long term durability. In a geotextile long term durability is of importance since they may be used in applications that have a life of 100 years or more [3]. Another problem with using polymer wastes is the lack of homogeneity or purity of waste. Although polyolefins are relatively simple to separate from other plastics the separation of the two principal polyolefins, i.e. polypropylene from polyethylene is much harder due largely to their similarity in densities. As these two polymers are immiscible in one another a heterogeneous polymer blend is likely to be formed if incorporated into virgin material. This in turn can lead to weakness in the physical properties of the resulting fibres and hence their inability to function over long periods.

In recent years a number of stabilisers have been developed specifically for improving the performance of recycled plastics. Ciba Speciality Chemicals has developed a number of recycling stabilisers particularly suitable for polyolefins [4]. This study investigates the effect of a particular type of stabiliser, namely Recyclossorb 550 on proportionate blends of HDPE wastes and virgin HDPE into polypropylene.

2. Experimental

Commercially recycled HDPE originating from crates and tray production were supplied by Linpac, Castlefield, West Yorkshire (Polymer A). Virgin HDPE Rigidex 5211 was obtained from BP and used as controlled blend (Polymer B). Some of this polymer was put through a plastic recycling extruder by Munchy Ltd, Wallingford for three times to produce a laboratory waste of known history i.e. with no long-term exposure to UV (Polymer C). Details of the polymers used are given in Table 1.

Table 1. Polymers used and their properties

Properties	**Unit**	**Novellen 1101R (PP)**	**Linpac HDPE (polym. A)**	**Rigidex HDPE5211 (polym. B)**	**Munchy HDPE5211 (polym. C)**
MFI	g/600sec	24	6	11	12
Density	g/cm^3	0.91	0.955	0.951	0.95
Elongation at Break	%	>50	50-150	1000	600
Melting point	°C	163	-	131	131

All three HDPEs were blended with Novellen 1101R polypropylene (PP) in variable percentages of 5,10, 15 and 20% and a Plasticisers Engineering Labline melt extruder stationed at Bolton was used to produce geotextile grade fibres. To study the effect of additional stabiliser on durability, 0.2% Recyclossorb 550 was incorporated in further blends of polymer A and polymer C. A 100% PP fibre was also produced to allow comparative assessments. In all cases the fibres were drawn 6:1 and had a linear density of ~2 tex. Fibres were subjected to tensile testing on a Textechno Statimat M tensometer fitted with a 10N load cell to determine tenacity and elongation-at-break.

To simulate ageing in the environment, samples of each blend were wrapped round tests tubes to prevent undue stresses and aged over a total period of 70 days in a Leader Engineering Oven fitted with circulating fan at a temperature of 100°C. Details of the experimental plan are given in table 2.

Table 2. Ageing regime

Fibre	**7 days**	**14 days**	**28 days**	**35 days**	**42 days**	**56 days**	**70 days**	
100% PP	*	*	*	*	*	*		
5%,10%,15%,20% Linpac recyclate (polym. A)	*	*	*	*	*	*		
5%,10%,15%,20% HDPE (polym. B)	*	*	*	*	*	*		
5%,10%,15% (polym. C)	*	*	*	*	*	*		
5%,10%,15%,20% Linpac recyclate (polym. A + stabilizer)	*	*	*		*	*	*	
5%,10%,15% (polym. C + stabilizer)	*	*	*		*	*	*	

The fibres were also subjected to DSC analysis using a Polymer Laboratories DSC operating at 10°C/min up to 250°C under flowing nitrogen to determine their melting points at all stages during ageing. FTIR analysis using a MATTSON 3000 FTIR Spectrometer was also used to obtain both crystallinity and carbonyl indices of unstabilised and stabilised blends throughout the ageing period [5].

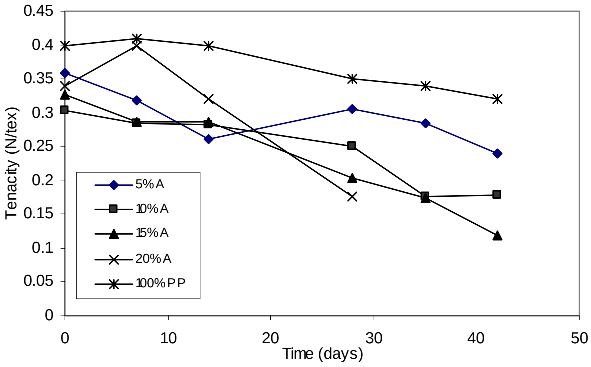
3. Results

It was observed that adding HDPE to PP and extruding generally had a detrimental effect on the drawabilty of the fibres produced; with the general rule that the higher the amount of HDPE included the worse was the drawability. It was also found that the addition of the recycling stabiliser also had an adverse effect on fibre drawabilty.

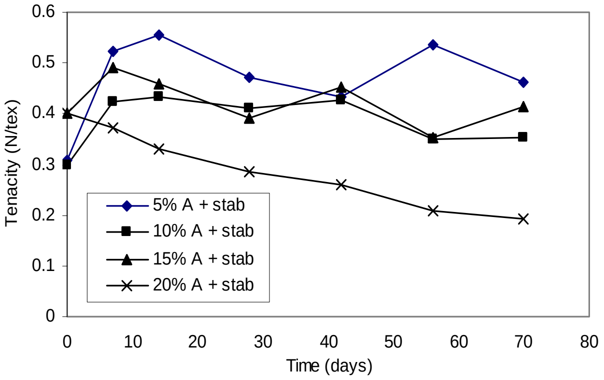
3.1 Tensile tests

Virgin polypropylene fibres aged over 55 days showed a small initial rise in tensile strength up to and including 7 days of ageing before gradually deteriorating and falling with time (Figure 1a). The downward trend is somewhat accelerated with proportional addition of polymer A when examined at all stages of ageing. However, fibres containing 20% polymer A showed, somewhat surprisingly, an initial rise in strength followed with a sharp drop but did not survive beyond 30 days of ageing. Addition of the stabiliser, as shown in Figure 1b, appears to positively influence both the tensile strength during the ageing period and the durability of the fibres allowing them to survive up to 70 days of ageing. This trend is seen to be largely independent of the blend concentration except for the 20% input where tenacity falls steadily from the outset. Effect of blending polymer B or the virgin HDPE on tensile properties with ageing time is shown in Figure 1c. The fall in strength and durability is relatively small compared to the polymer A blend.

1a)

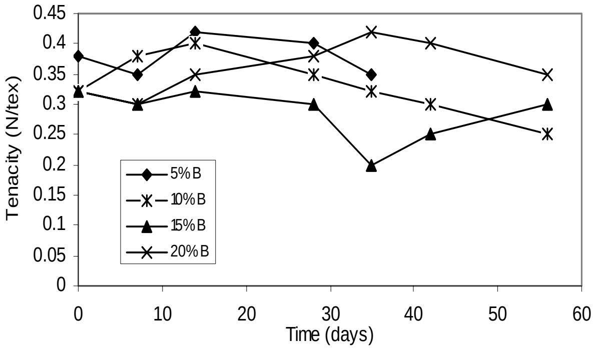


1b)



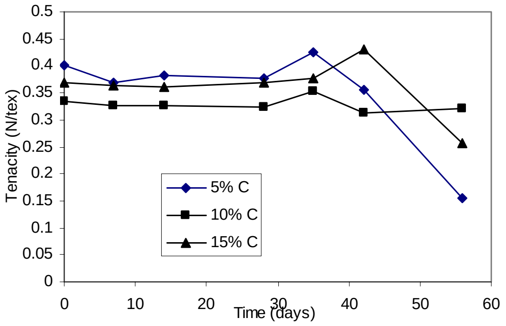
Proportional addition of polymer B or virgin HDPE has relatively less detrimental effect on long-term durability although the trend for most combinations is downward after the initial 28 days of ageing. As shown in Figure 1c these fibres with the exception of 5% blend survive up to and including 56 days. The 20% blend, shows initially a small reduction in strength but appear to improve after the first week of ageing i.e. tenacity rise from ~0.32 N/tex to a maximum of ~0.42 N/tex before falling off.

1c)

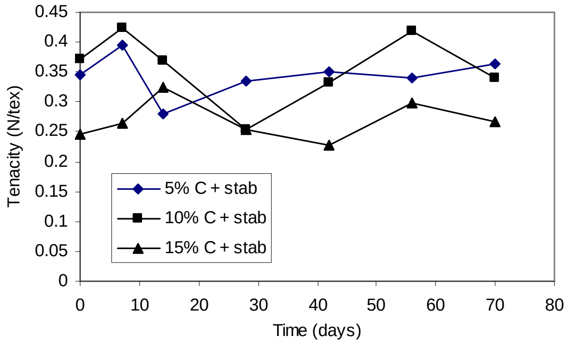


All fibres blends containing polymer C showed consistent and stable tenacities up to and including 45 days of ageing before deteriorating in strength. However, samples containing 20% blend could not be effectively drawn and hence does not appear in the graph (Figure 1d). Addition of the stabiliser allows these fibres to survive beyond 70 days of ageing with little change in their periodic tenacities, although results referring to 10% blend are somewhat erratic (Figure 1e).

1d)



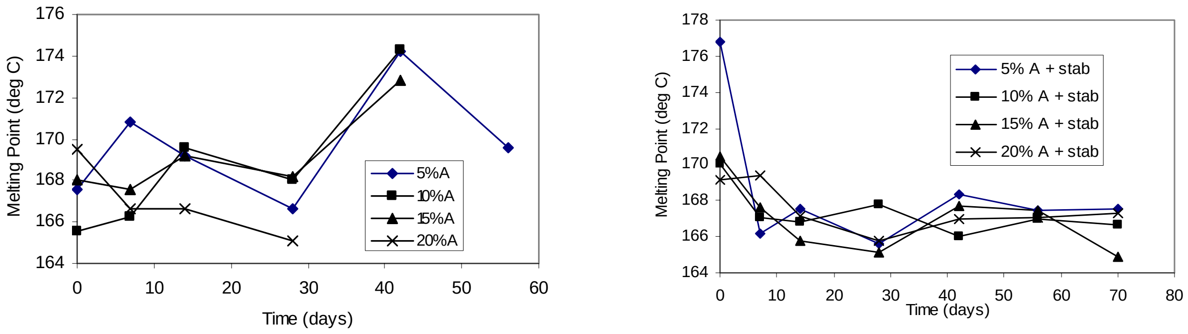
1e)



A large drop in breaking elongation was also noted in the first few days of the ageing period for all fibres tested throughout this work. This is believed to be due to an annealing effect caused by the high oven temperature. However, after this initial period the drop in breaking elongation is much less marked although the fibres containing polymer A, or the commercial waste show the greatest loss in elongation with time. Fibres containing the additional stabiliser show the least loss in elongation.

3.2 DSC and FTIR Spectroscopy measurements

From the DSC measurements the melting peak points of both PP and HDPE components in the blends tend to rise with ageing time. This is particularly pronounced for fibres containing polymer A or the commercial waste. Figure 2a shows the variation in melting peak of PP component of polymer A blend with ageing time. Inclusion of Recyclossorb in the blend appear to have considerably stabilised this variation right up to 70 days of ageing (Figure 2b).


2a)

2b)

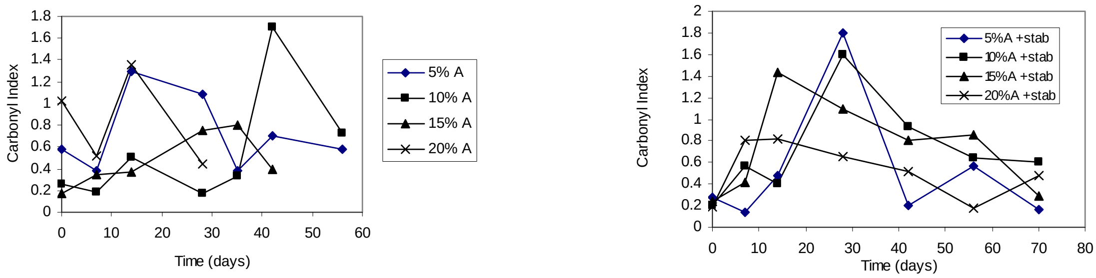
Figure2: Effect of oven ageing on the melting point of polypropylene component a) without stabiliser and b) with stabiliser

Crystallinity measurements for polypropylene are obtained from the ratio of the values of peaks at wave numbers 974cm $^{-1}$ and 998cm $^{-1}$ and carbonyl index are extracted from the ratio of carbonyl peaks in the ~1708cm $^{-1}$ region and that at 974cm $^{-1}$. Measurement of crystallinity using this method has been somewhat erratic and therefore not entirely reliable. Table 3 shows the values obtained for the crystallinity indexes of the Polymer A inclusion fibres without and with additional stabiliser. It may be seen that the values of crystallinity index range from 0.6198 to 0.9262 with no apparent trend, suggesting that any changes in crystallinity are being swamped by experimental error.

Table 3. Effect of oven ageing at 100°C on the Crystallinity Index I_{cry} of the PP fibre containing commercial recyclate (polymer A) without and with additional stabiliser (Recyclossorb 550)

Ageing Time	**0 days**	**7 days**	**14days**	**28days**	**35days**	**42days**	**56days**	**70days**
Fibre	**Crystallinity Index, I_{cry}**							
100% PP	0.7867	0.8133	0.6416	0.8066	0.6423		0.7381	
5% Polm. A	0.6755	0.7244	0.8157	0.75	0.7959	0.9262	0.7295	
10%Polm. A	0.8356	0.7356	0.71	0.7375	0.7698	0.7057	0.7544	
15%Polm. A	0.6198	0.757	0.7287	0.7522	0.7126	0.6939		
20% Polm. A	0.83515	0.7111	0.6741	0.7925				
5% Polm. A +stab	0.7599	0.7319	0.741	0.8427		0.8401	0.7046	0.7183
10%Polm. A+stab	0.7933	0.718	0.7618	0.6548		0.7611	0.768	0.7296
15% Polm. A + stab	0.8049	0.7376	0.6327	0.8282		0.6785	0.8198	0.7482
20% Polm. A + stab	0.8105	0.6712	0.8054	0.6974		0.6894	0.7239	0.7759

The carbonyl measurements however, are more revealing. Figures 3a and 3b show carbonyl index data for fibres containing the polymer A blend without and with the additional stabiliser. Carbonyl products of polymer A blend appear to increase with ageing time for some blend ratios and indeed similar trends are followed when the additional stabliser is included. However, blend containing the additional stabiliser appear to produce less carbonyl products beyond 35 days of ageing for all combination of polymer A blends. This suppression of carbonyl formation is believed to be due to the additional stabiliser which also helps to enhance durability beyond 56 days. Both crystallinity and carbonyl index measurements are prone to some experimental error and therefore these results must be treated with some caution.


3a)

3b)

Figure 3. Effect of oven ageing at 100°C on the Carbonyl Index for fibres containing polymer A or the commercial recyclate a) without stabiliser and b) with stabiliser

4 Discussion

The addition of HDPE to PP was generally found to reduce drawability of the fibres; the greater the proportional blend of HDPE the more difficult the fibres were to draw. It was noted that the fibres containing the commercial recyclate (Polymer A) were the most difficult to draw; this is possibly due to the greater difference in MFI of this polymer and the PP it was blended with compared to the other polyethyenes. In all cases the draw rollers were set at 70°C, however, better drawing may result at higher roller temperatures as reported by Ward *et al* [6] and Greco *et al* [7-9]. Alternatively compatibilisers [10] maybe used to improve drawability although these are not generally used in fibre manufacturing. More recently better drawability has been reported by using paraffin as a compatibiliser for polyolefin blend fibres [11].

Inclusion of the stabiliser, Recyclossorb 550 on the whole also made fibre drawing more difficult, this may be due to the fact that this particular stabiliser was originally intended for bulk or film applications and not fibres.

Generally, addition of HDPE to PP reduced tenacities of the drawn fibres. Nominal tenacities of the blended fibres were within 0.3-0.35 N/tex compared to approximately 0.4N/tex for 100% PP fibre. Also greater the percentage of HDPE blends the lower the recorded tenacities. Addition of HDPE to PP in bulk has been reported by some experimenters to have beneficial effect on mechanical properties of the blend [12] but this was not generally found to be the case for the blended fibres studied in this work.

The ageing of the blended fibres containing polymer A or the commercial recyclate reduced fibre tenacities; the higher inclusions of the HDPE had greater detrimental effect on durability as measured by tensile testing. In contrast however, the fibres containing polymer B or the virgin HDPE and certainly those containing polymer C or HDPE with known history retained their tenacities for longer periods. These findings clearly show that recycling alone where polymer is largely exposed to thermal degradation may have little effect on polymer long-term durability when blended with virgin PP. Other factors such as UV exposure or undue stresses may play a larger role in degradation of these polymers as has been shown with inclusion of polymer A or commercial recyclate with unknown history. As discussed earlier the commercial recyclate originate from crates and trays, typically used for milk bottle carriage where they must generally be subjected to rough handling and exposure to outside conditions for extended periods.

The measurement of melting point by DSC suggests that there was a rise in crystallinity followed by a fall for all three sets of fibres without additional stabiliser. It has been suggested that since degradation occurs in the amorphous regions this may result in the ends of the broken chains being absorbed into the crystalline regions [13] hence the noted increase in crystallinity. The drop in melting points that were observed with some samples could be due to oxidation occurring in the amorphous regions and the surfaces of the crystalline regions towards the end of the ageing period. In the two waste blends (polymer A and C) with the additional stabiliser it was observed that any rise in the melting points was not as pronounced suggesting that changes in the crystallinity in these latter cases are not as great.

The measurements of the crystallinity and Carbonyl indices by FTIR spectrometry appeared to be affected by experimental errors raising questions about the validity of the results obtained. At least some of the measurements of carbonyl index however suggested that the additional stabiliser may have had a ‘mopping up’ effect on some of the degradation products. This may indicate that the species that give rise to the initial formation of C=O groups are prevented from being reformed by the stabiliser and that the C=O groups are themselves being degraded under oven ageing conditions.

5 Conclusions

Addition of HDPE in the following forms i.e. virgin, recyclate with known history and commercial waste with unknown history to polypropylene has proved to have been a fruitful exercise. The study has shown that generally long-term durability suffers with proportionate increase in HDPE blend component. The deterioration in strength appears to be related to the type of waste and its history. The commercial waste with unknown history shows the greatest loss in overall strength whereas the virgin HDPE and that with known history show little difference in tensile properties suggesting that in the latter case that repeated extrusion alone may not have a significant effect on the properties of the blended fibres. Compared to the fibres without additional stabilizer, the filaments with the Recyclossorb 550 were still showing most of their original strength at the end of the 10 week ageing period. This is true for both the commercial waste inclusion and those with known history. This suggests that the use of this stabilizer could be a way of extending the long term life of the fibres in service if problems with extrusion could be overcome.

Tensile testing appears to be the most reliable method of testing for degradation as other methods such as DSC and FTIR appear to be subject to excessive experimental error with respect to possible changes occurring, although they do yield useful information about changes in crystallinity.

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