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Degradation of recycled polyethylene film materials due to contamination encountered in the products' life cycle

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Abstract: The recycling of polyethylene (PE) film is a straightforward process, provided the material that is being recycled has come from a clean source and is consistent in composition. Most waste film recovered for recycling has been subjected to a series of 'life cycle factors', e.g. comingling with wastes such as dirt, packaging tapes, and other plastic film products. Such contamination can degrade the material's physical properties and this article presents quantitative data on the effects of contaminants on PE films. Results suggest that dirt contamination can adversely affect the elasticity and melt viscosity of the polymer, but the contaminant does not interact directly with it and that polypropylene-based packaging tape is more suited to mechanical recycling than polyvinyl chloride or cellulose-based tapes.

Keywords: plastic film, recycling, contamination, materials testing

1 INTRODUCTION

Plastic film products have been in use as packaging for around 60 years [1–4], but their recycling is a fairly new practice, which is rarely performed on material arising from municipal waste sources. Plastic film is a versatile modern packaging material with a number of applications. There are six main varieties of plastic film, each from a different polymer, whose name and principal application is listed in Table 1. Also shown are the recycling symbol numbers, used to identify the constituent polymer of the product for recycling.

The UK currently produces around 250 million tonnes of controlled (i.e. regulated by the Environment Agency) waste per year [5]. Of this, approximately 30 million tonnes is municipal solid waste and 80 million tonnes is commercial and industrial waste.

In terms of the amounts of plastic film waste produced, current estimations [6] put the amount of plastic film in the UK municipal waste stream at 1.2 million tonnes per annum. Environment agency figures estimate that plastic film in the commercial and industrial sectors are of the order of 1 million tonnes per annum [7]. Therefore, in total, the UK

produces an average of around 2.5 million tonnes of plastic film waste per annum. Of this total, polyethylene (PE) products (high density polyethylene (HDPE), low density polyethylene (LDPE), and linear low density polyethylene (LLDPE)) make up the majority of film consumed.

Waste management practices in the UK are heavily reliant on landfill, which is a diminishing resource because of environmental issues and tightening legislation. Alternative technologies to deal with solid waste (including recycling) have been given increased attention in recent years because of this impending landfill shortage and increased disposal costs. The recycling rate of waste plastic film is currently very poor, with only 10 per cent being recycled into new products [8].

This article highlights the effect of mixing a range of contaminants with PE packaging materials over a range of concentration levels. The type of contamination added was indicative of foreign material that will become mixed with the PE during a typical life cycle as a consumer product. A series of mechanical and material analysis tests were used to quantify the effect of a number of heat recycling processes on the film material.

2 RECYCLING PROCESSES

2.1 Recycling applications and products made

Waste plastic film for recycling is acquired by recyclers from a number of sources. The simplest

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Table 1 Common applications of packaging films

Name	Application	Recycling symbol
High density polyethylene	Packaging film and grocery bags	2
Polyvinyl chloride	Low oxygen permeability packaging such as frozen food bags	3
Low density polyethylene	Food packaging, damp-proof sheeting, agricultural films, and grocery bags	4
Linear low density polyethylene	Stretch wrap and cling film	4
Polypropylene	Woven tarpaulin sheets. Also used as shrink wrap such as film coverings	5
Nylon	Food contact applications where strength is needed such as soup packets and sachets	7 (other)

to process and with least contamination is from post-production sources where off-cuts or batches that have failed quality control are sent directly to the recycler. At the other end of the spectrum is material from a mixed source obtained from kerbside recycling schemes, where composition can be spurious and contamination is higher.

In terms of PE film, the recycling process requires equipment that agglomerates the bulky film into manageable chunks before it is fed into an extruder for melt processing. This process is often referred to as the continuous agglomeration or 'KAG' process. Once in the extruder, the molten material is de-gassed and filtered to remove contaminants before being extruded and pelletized.

Removal of contamination in the melt phase is only possible if the amount of contamination is relatively small, otherwise filters become blocked resulting in poor production rates and equipment damage. In the case of plastic film, small amounts of contamination can result in discontinuities, such as air bubbles or breaks in the recycled sheet, which is not acceptable when producing a high quality film product.

2.2 Factors affecting recycling

The problem of whether plastic film can be recycled effectively is dependent on the condition of the material arising from its 'life cycle' prior to being recycled. This article presents research that quantifies the effects of contamination on film grade PE. Understanding such effects is crucial in persuading film recyclers to use material that has come from a feedstock that contains small amounts of contamination.

Much of the activities in modern plastic film recycling are based on recovery of scrap or off-cut

material at manufacturing sites. This is undertaken for economics, such that value can be gleaned from manufacturing scrap, rather than wasting what is essentially usable material. The recycling market for back-of-store packaging film, material that has not yet passed into the consumer domain is steadily growing, but film recycling from postconsumer sources is virtually non-existent. This is mainly attributable to two factors.

1. The inability to quantify the effect of dirt and packaging tape contamination on plastic film products, such that the mechanical properties of the material feedstock can be estimated prior to the expense of recycling.
2. The lack of a system to reliably audit the history of a material feedstock in order to trace the origins of a particular material.

As with the recovery of any waste material, the net economics of the whole process, from initial consumer disposal through the recovery and recycling phases to remarketing as a new consumer product, must be favourable or the recovery operation will not function economically. Removal of contamination has been cited [9] as causing additional costs in the recycling process, which can lead to an economically infeasible process.

Previous studies have shown that recycled plastic is often of inferior quality to that of virgin material [10]. Studies associated with actual material life cycles and associated contamination have shown that gluing makes mechanical recycling less favourable owing to troublesome dismantling and the high degree of contamination from the glue [11]. Dirt and organic food wastes can also cause problems when recycling, which should be quantified [12].

3 EXPERIMENTAL METHODOLOGY

The experiments covered herein involve the mixing of two contaminants, i.e. dirt and packaging tapes with PE film materials. In the case of the simulated dirt, a pure natural talcum powder was used. For the mixing of packaging tapes with the PE film, three varieties of tape were used: polyvinyl chloride (PVC), polypropylene (PP), and natural cellulose based tape. These are typical varieties of tape to be used in conjunction with PE films.

The input materials used for the experiment were as follows.

1. Material A – a fresh pelletized polymer purchased from an industrial source. The material had a narrow specification for use in packaging applications.

2. Material B – a broad specification LDPE supplied by an industrial extrusion company.
3. Material C – a modern reprocessed film product produced from a mixed waste PE source that was sourced from a recycling company [9], composition mostly LDPE and HDPE. The material was washed and melt-filtered to remove contaminants.

3.1 Addition of contamination

The concentration of the contaminant was varied from 0 up to 15 per cent and the material was tested for each level of contamination. This is outlined schematically in Fig. 1. Contamination was introduced to the polymer before production of the test specimens and agitated to ensure an even distribution. Mixing was performed in a dry environment to avoid cohesion.

The equipment used in the manufacture of test specimens was a Fox and Offord 'Polylab' Universal moulding machine. The machine was capable of heating the polymer charge to 300 °C and delivering moulding pressures of up to 17 MPa. According to the user guidelines, the moulding conditions were set to 180 °C and 10 MPa moulding pressure, as these were the advised conditions for PE-based material.

3.2 Tensile testing

The test pieces were evaluated for tensile properties using a Testometric M500-50 series tensile testing machine. The samples were pulled apart at a speed of 500 mm/min [13]. The Testometric machine was capable of displaying load/extension or stress/strain curves for any specimen, but these were not used directly because extension of plastic specimens occurs over the whole sample, rather than just the tested portion of the sample. Extension was therefore measured independently from the machine's output.

3.3 Measurement of melt flow index (MFI)

A device capable of measuring the melt viscosity, or MFI, of polymers was employed to conform to testing standards [14]. Details for the testing of

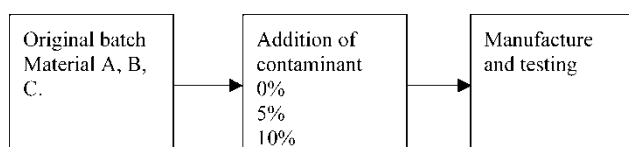


Fig. 1 Schematic of the process by which contamination was added to the polymer and tested

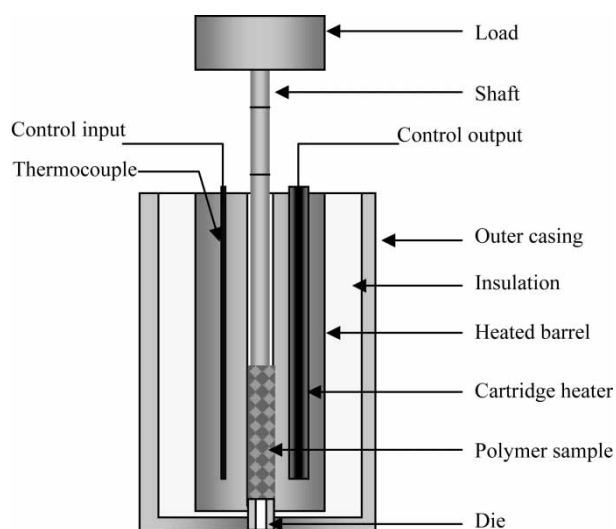


Fig. 2 Schematic of the MFI apparatus

specimens are also shown in this standard. Conditions for the testing of PE samples were nominally 190 °C under a load of 2.16 kg. Figure 2 shows a schematic of the apparatus. The polymer sample was loaded into the heated barrel and a loaded shaft was placed on top to force the polymer out of the die. The mass flowrate of the polymer moving through the die was then used as a measure of its melt viscosity. Temperature control was achieved via a microprocessor controller to within ± 1 °C.

3.4 Measurement of crystallinity via X-ray diffraction (XRD)

For this experiment, XRD analysis was carried out using a Philips PW 1710 automated powder diffractometer. This apparatus used copper $K\alpha$ radiation, set at 35 kV and 40 mA. The software used to operate the machine was PW1877APD version 3.6, which was run on a PC for simultaneous traverse control, data collection, and storage.

The apparatus was set to traverse and gather diffraction data from 3° to 60°. An initial scan of wider diffraction angles with a variety of PE samples showed that there were no useful data outside of this interval and thus data above 60° or below 3° were not collected to save traverse times and data file size. Each scan was taken in 356 discrete steps with eight measurements being taken per step, which were averaged for consistency. The results were then exported from the logger PC and loaded into a spreadsheet package. Initially, these results were too similar to discern quantitative changes in crystallinity and it was decided to calculate the crystallinity based on a recognized deconvolution method [15].

3.5 Measurement of molecular weight and polydispersity index (PI) via gel permeation chromatography (GPC)

The hardware used was a polymer laboratory GPC220 instrument fitted with a Viscotek differential pressure (viscosity) detector, located at Rapra Technology Ltd, Shropshire. The polymer was first made into a gel by being dissolved in trichlorobenzene at 190 °C to a concentration of approximately 0.1 per cent by weight and immediately inserted into sample injection vials. The samples were then injected into the GPC columns, where they were processed at 160 °C. The gel-phase sample was then allowed to pass through the filter columns at the rate of 1 ml/min. The columns were two 300 × 7.5 mm tubes containing 10 µm Plgel filter beads. After passing through the columns, concentrations of the gel were measured at the detector and logged into a computer. This was then used to build up a chromatograph of the polymer sample. The results were returned in electronic format and loaded into a spreadsheet package.

4 RESULTS AND DISCUSSION – TALCUM ADDITION

To investigate the effect of simulated dirt on the PE film products, the following results are shown with natural talcum as contaminant.

4.1 Tensile testing results

Figure 3 shows the percentage change in ultimate tensile strength (UTS) with increasing concentrations of talcum. The results show different

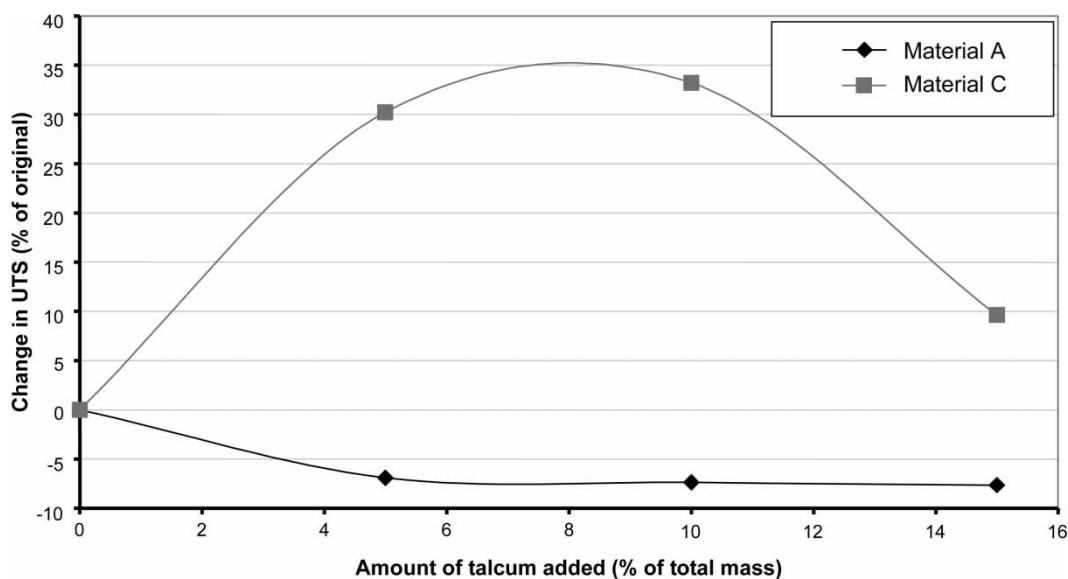


Fig. 3 Ultimate tensile strength of two polymers versus percentage of talcum by mass

responses from the two materials: material A reduced in UTS, whereas material C increased.

It can be seen that the addition of talcum-like contamination seems to have little significant impact on the UTS at values 15 per cent, which would be a realistic maximum in a practical sorting and cleaning operation. The concentration of talcum seems to show a slight decrease in the UTS of material A up to <15 per cent contamination. Material C initially increases in UTS, but at 15 per cent contamination, this increase is only 10 per cent greater than the original UTS.

Figure 4 shows the variation of percentage elongation of the PE at failure with increasing amounts of talcum.

Unlike the UTS relationship, there is a fairly substantial drop in the elongation at failure of the range tested. The talcum has stiffened the material, as seen in industrial practice where chalk is used as a stiffener (B. Keeling, personal, communication). The data point at 5 per cent contamination for material A is higher than expected, possibly due to inconsistent mixing, indicated by a larger amount of scatter observed at this value. The overall change is of the order of a 55 per cent reduction after 15 per cent talcum contamination. For material C, the overall change was a 60 per cent reduction after 15 per cent contamination, but the data appear to show that the change in maximum elongation is more pronounced at first and levels-out above 10 per cent contamination.

4.2 MFI measurement

Figure 5 shows the relationship between MFI and talcum contamination over the 0–15 per cent

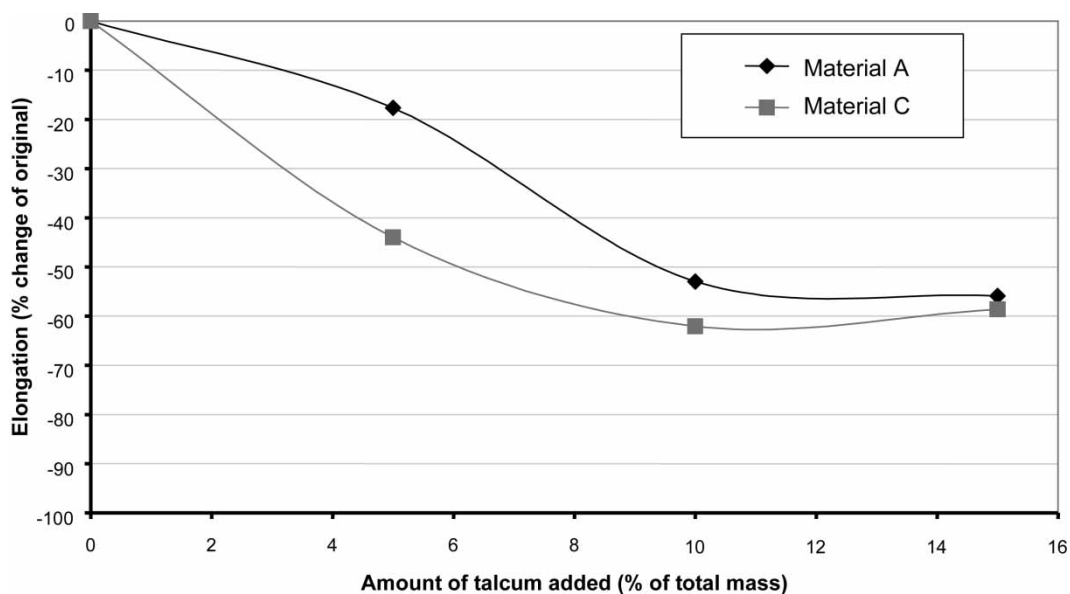


Fig. 4 Percentage elongation of two polymers versus percentage of talcum by mass

region of talcum added to the two mixtures. The change in MFI follows a similar trend for both materials, although there appears to be an anomaly at 5 per cent contamination with the material A. As the value was similar to the original value at 0 per cent contamination, it could be due to either a sample with a much lower talcum concentration than expected or the contamination having no effect until a higher threshold value was reached.

Despite the difference of 0.5 g/10 min in MFI between the two PE materials examined (materials A and C), the decrease in MFI was around 6 per cent per 1 per cent talcum addition in both cases. It is logical to assume that these changes would be due to a fairly simple relationship because the

talcum is acting to increase the viscosity of the material. Thus, the amount of talcum added should have a direct effect on the change in MFI. The index is a far more important property during manufacture, thus dirt contamination is more detrimental during the processing stage than its effect on the end product.

4.3 XRD measurements

Figure 6 shows the XRD results for the talcum addition experiments as a plot of contamination versus crystallinity. Sharp peaks on the XRD spectra were identified as a talcum compound with the

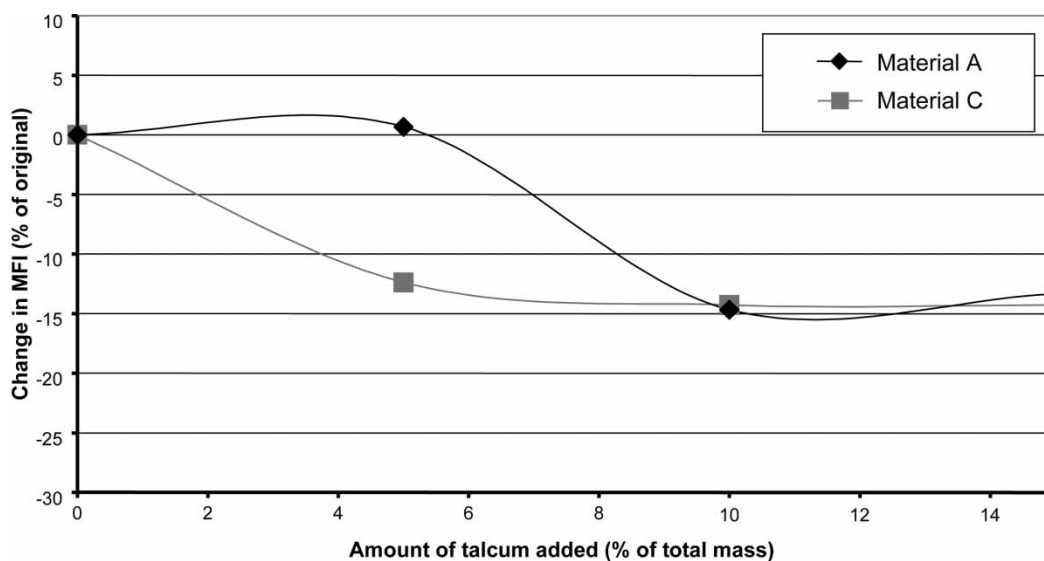


Fig. 5 MFI of two polymers versus percentage of talcum by mass

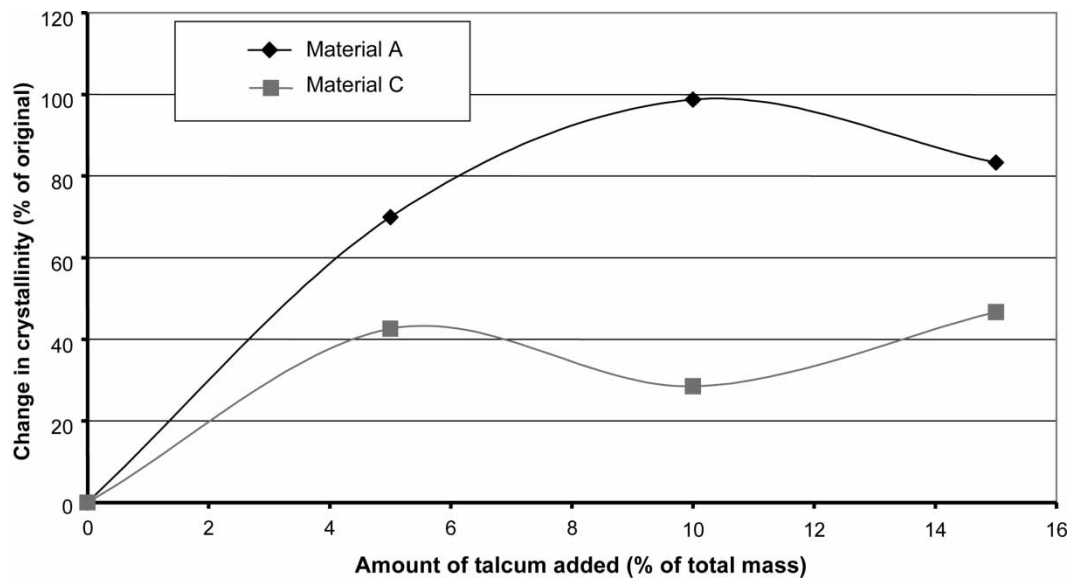


Fig. 6 Crystallinities of two polymers showing different talcum concentrations

chemical formula $Mg_3Si_2O_{10}(OH)_2$ using the crystal salt database contained within the XRD software.

The data show that there is an initial increase in crystallinity for both polymers below 10 per cent contamination. It is also interesting to note that the values of crystal plane spacing in the polymer spectra did not change with increasing amounts of talcum. This implies that the talcum did not affect the way in which the crystal lamella of the polymers were aligned. It is, therefore, logical to assume that on a crystalline level, the talcum has virtually no effect of the polymer chains and their alignment, but it merely 'sits around' them rather than interacting directly with them.

4.4 Addition of packaging tape

To assess the impact of adding increasing amounts of packaging tapes, this series of experiments used three varieties of tape: parcel packaging (PP) tape, cellulose-based tape, and PVC tape. The three were chosen as they are examples of commonly used varieties.

4.5 Tensile testing

Figure 7 shows the effect of adding the tapes to PE film material, note that there were two types of PE film used, namely materials B and C.

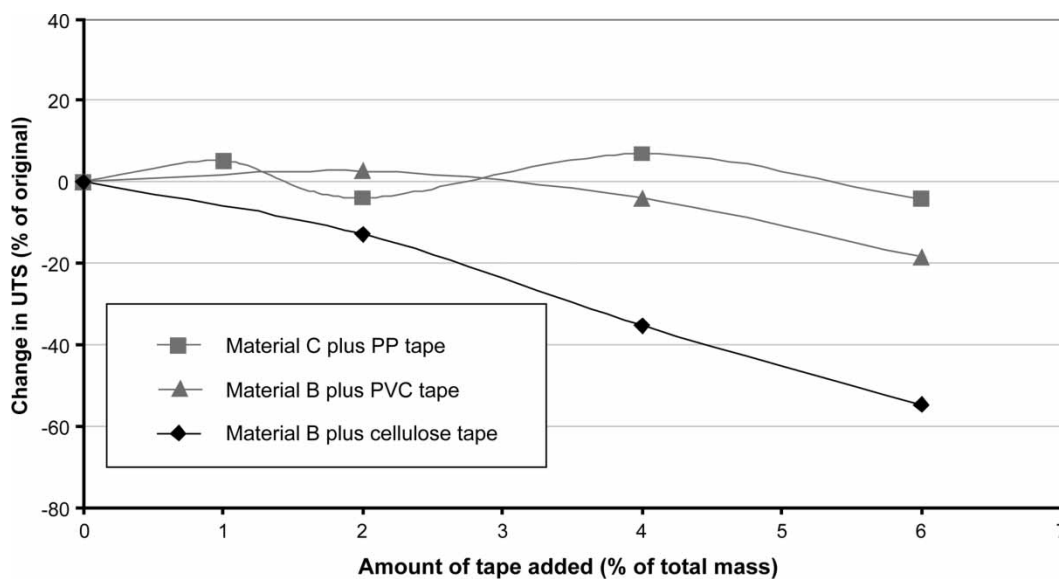


Fig. 7 UTS versus amount of tape added to the PE

The data show that the PP tape had the smallest effect on material C. The PVC tape caused a drop in UTS of approximately 3 per cent per 1 per cent of tape added. The cellulose-based tape had the most detrimental effect of a reduction of about 9 per cent per 1 per cent of tape added.

Figure 8 shows the effect the tape has on the plastic film in terms of the change in maximum elongation. In all three cases, the tape causes an overall reduction in the value.

The smallest effect came from the PVC tape, a reduction of only 5 per cent in maximum elongation over the contamination range, but this material initially showed an increase in maximum elongation at 2 and 4 per cent contamination. The PP tape caused the maximum elongation of material C to reduce by 37 per cent at 6 per cent contamination. Again the cellulose tape had the most detrimental effect, causing the maximum elongation to reduce by 53 per cent over the range of the test. This shows that the tapes were affecting mechanical properties differently, depending on the type of tape used and that the cellulose tape contamination has the largest (and most weakening) effect on the tensile properties examined.

4.6 MFI results

Figure 9 shows the change in MFI as tapes are added to the PE materials. Again, it is clear that the cellulose tape had the opposite effect than that of the PP and PVC tapes.

Over the 6 per cent contamination range, the PP and PVC tapes had the same effect, i.e. to increase

the original MFI value by 12 per cent. There was more scatter in the material C/PP tape data, probably due to inconsistent mixing. Qualitatively, it can be seen that the effects of the PP and PVC tapes are similar, despite using different PE materials for the study. The effect of the cellulose-based tape was the largest and shows a reduction in MFI, i.e. an increase in melt viscosity, which would make manufacturing the contaminated material more difficult. This relationship showed a decrease of 22 per cent from its original value over the 6 per cent contamination range.

4.7 XRD measurements

Figure 10 shows the effect of addition of PP tape to material C. There was perhaps a small increase in crystallinity of approximately 5 per cent over the 5 per cent range of contamination, but this is not seen as significant.

4.8 GPC results

Figure 11 shows the relationship between the molecular weight and the PI of material A with increasing amounts of PP packaging tape.

Overall, it appears that there is an 8 per cent increase in molecular weight and a 7 per cent decrease in PI. This would be consistent with the PP tape increasing the overall molecular weight of the sample mixture. It is interesting to note that most of these changes occur within 2 per cent tape addition, thereafter the tape having little effect. If the molecular weight and PI are both independent

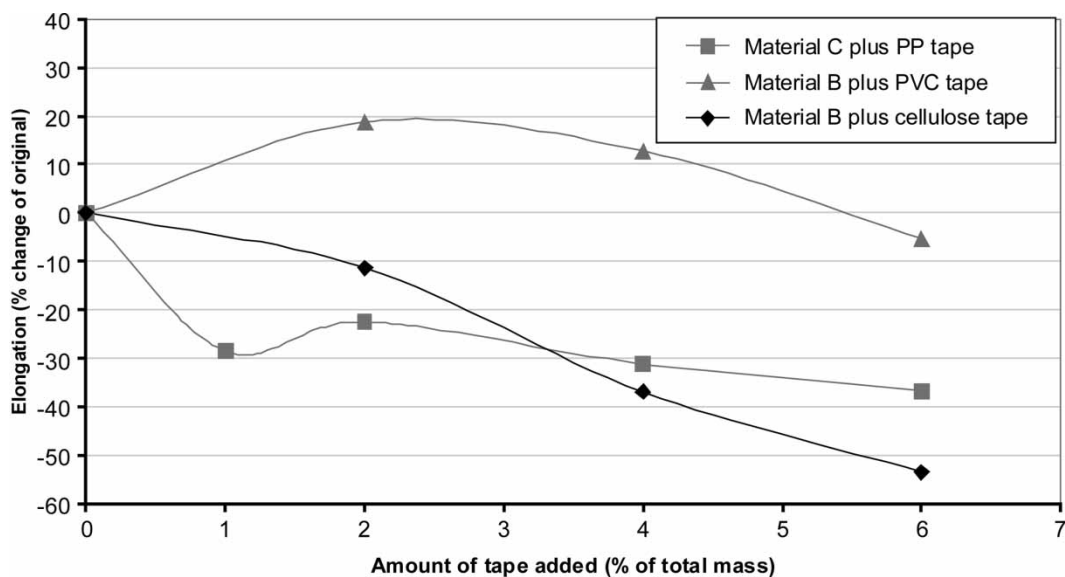


Fig. 8 Maximum elongation versus amount of tape added to the PE

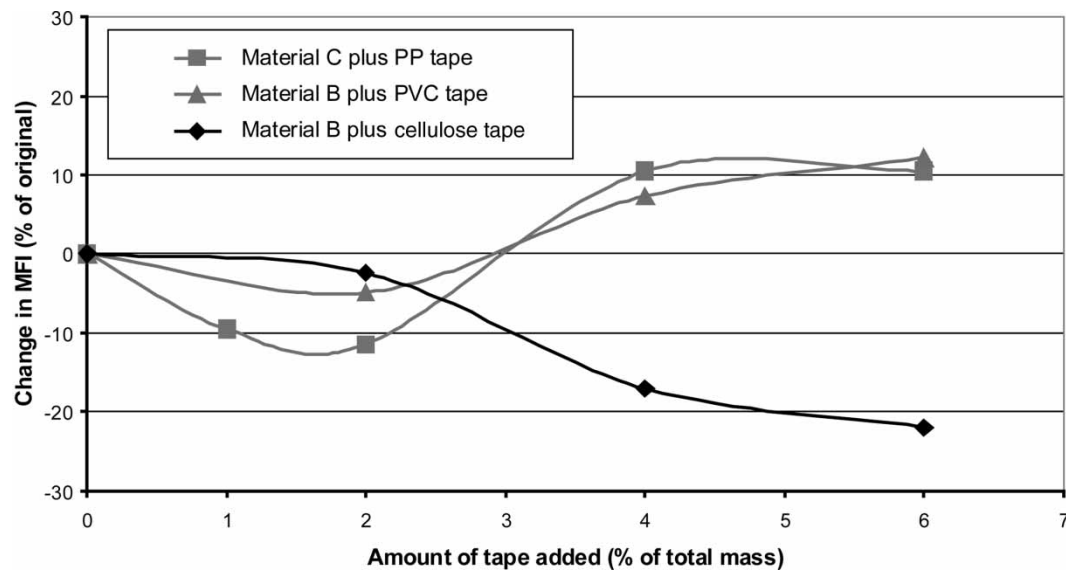


Fig. 9 MFI versus amount of tape added to the PE

of the amount of tape added (up to 6 per cent tape by mass), then it appears that the tape has virtually no effect on the PE polymer chains at a molecular level.

5 INTERPRETATION AND DISCUSSION OF RESULTS

Increasing the amount of talcum made the materials stiffer and inherently more viscous, and thus harder to process. The biggest problem in terms of contamination is its effect on the MFI rather than on the mechanical properties. The crystallinity of the

polymer itself does not change because the talcum does not interact directly with it.

The effect to which a tape changes the tensile and melt-flow properties is very much dependent on the kind of tape that has been added. Cellulose-based tapes have the most detrimental effect on the material properties of those tapes tested. Packaging tapes have little or no effect under the concentrations tested. Tapes made from cellulose tend to be stiffer and more viscous and thus had the biggest effect on the films tested. On a molecular level, packaging tape has little effect on the polymers within the PE structure.

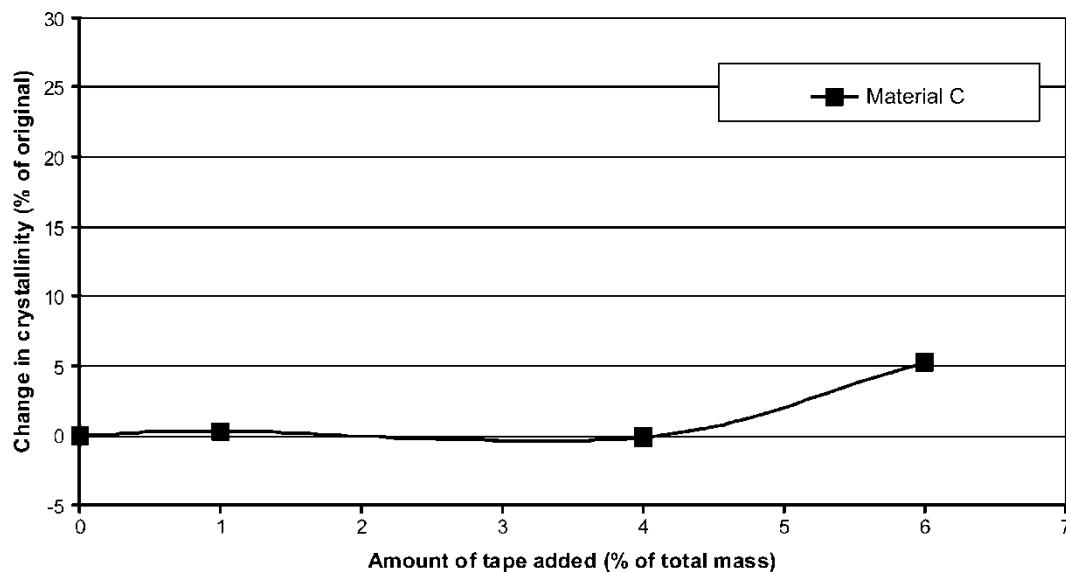


Fig. 10 Change in crystallinity with amount of packaging tape

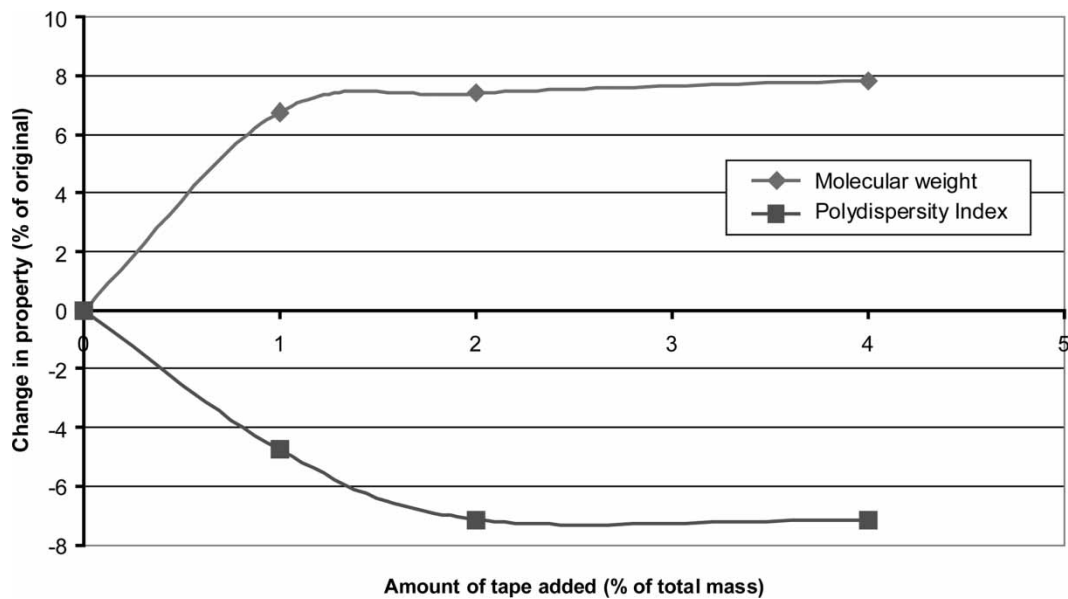


Fig. 11 Molecular weight and PI versus amount of packaging tape added to material A by mass

Any talcum or tape contamination effects are far more likely to be a problem on thin films rather than on thicker sections. An unevenly mixed section of recycled PE would suffer substantial processing problems if there were a large amount (above 6 per cent) of cellulose tape or PVC tape present.

It is also worth mentioning the safety aspects of PVC contamination in PE recycling systems, which can potentially lead to the liberation of toxic chlorine-based gases, which is the reason why PVC contamination is so unpopular in the industry (D. Mercer, personal communication).

In the light of the results presented in this article, plastic film recycling practices could benefit from quantitatively understanding the effects of life cycle contamination on the properties of recycled film. Industrial practice is still geared towards obtaining the cleanest and cheapest possible feedstock, rather than undertaking the higher-risk option of cleaning contaminated ones. The results of this study prove that it can be difficult to estimate how a contaminated feedstock will perform in a recycling facility. The results also prove that these effects can be quantified and potentially modelled, based on a set of input criteria, such as amount and type of contamination. Improving the net quality of a recycled feedstock by blending it with a virgin material of known properties is another option open to film recyclers.

The results have shown that the PE material itself is not degraded by the action of mixing it with contamination, but the resultant mixture is affected. This suggests that cleaning and melt-filtering the feedstock will restore its original material properties. Technology is available to perform these functions,

which can be tailored to meet the waste stream under consideration.

Investment in washing and melt-filtering technology is therefore required from recycling businesses. Previous studies undertaken by the authors have shown that cleaning PE film for recycling applications is feasible, but highly sensitive to economics, [9, 16]. Cleaning is an expensive operation and can only be made economically feasible if the recycler produces a high-quality output that will fetch a good price.

6 CONCLUSIONS

The change in physical properties of PE film when mixed with dirt contamination is such that the material becomes stiffer and more difficult to process in the melt state. The severity of this change is dependent on the type of PE film used and the amount of contamination it is mixed with. The results within this article have shown that MFIs can fall by as much as 17 per cent with 10 per cent dirt contamination. XRD and GPC testing showed that although the talcum altered the mechanical properties of the film product, it did not interact with the polymer chains themselves.

The effect of mixing PE film for recycling with packaging tapes is dependent on the variety of tape used. Cellulose tapes have the most detrimental effect in terms of reduced strength, maximum elongation, and MFI. PP tapes have the smallest effect on the PE film, because the tape is similar in mechanical properties to the PE itself; little evidence

was found to suggest that the adhesive from the tape was interacting with the PE film. It would therefore be logical to suggest that any dirt contamination be well mixed into recycling feeds, rather than existing as highly concentrated 'clumps' and that the type of packaging tape used should be PP-based wherever possible.

In terms of possible effects on the waste management industry, this research has shown that dirt-like contamination will make recycled films more difficult to process due to increased melt viscosity. The effect of tapes will be unpredictable because the variety of tape in the feedstock is entirely dependent on the product life cycle. Heavily contaminated films are therefore more likely to be utilized in products where production consistency is not of prime importance. Examples of this include the manufacture of thicker profiles and plastic lumber sections. These applications can limit further recycling, but are more environmentally beneficial than directly landfilling plastic film waste.

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