Impact of Degradable and Oxo-fragmentable Plastic Carrier Bags on mechanical recycling

TESTING

12/11/2012

Foreword

This independent testing has been performed on behalf of EuPC (European Plastics Converters), located in Brussels. EuPC would like to thank all companies that provided carrier bags (free of charge) which were placed on the EU market in 2012-2013, in order to perform these tests and publish the results of the investigation.

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Introduction

In the last few years recycling has become more and more important – both for society and industry. As a result, the recycling mass flow of plastics has modified significantly and degradable plastics may already be present as components of the recycling mass flow of plastics.

EuPC raised the question of what effects the addition of degradable plastic bags has on the recycling-cycle. To answer this question EuPC started extensive industrial test series, starting with the recycling process and the production of films through to mechanical tests. Therefore, four mixtures of non-biobased degradable plastic bags and one virgin-LDPE recycling material were tested over the course of 6 months and over 3.700 measurements were made and a total of 9.45 tonnes of plastics have been processed. An overview about the material composition and the following processes is shown in Fig. 1.

About TCKT - Transfer Center for Polymer Technology (TCKT)

Austria has a small but healthy polymer industry, generating approximately €13bn in annual sales. Upper Austria, one of the nine provinces, achieves about €7.6bn with more than 220 companies in involved in the polymer industry starting with raw material suppliers, manufacturers of plastic processing machines, tool makers, plastic converters and manufacturers of recycling machines like EREMA. Some of the companies are world market leaders in their niche. The structure of the Austrian and Upper Austrian polymer industry is distinguished by small and medium enterprises (SME). However, the research expenditure of the Upper Austrian companies involved in polymer industry is around 4% of their sale. Within this scenery **the Transfer Center for Polymer Technology (TCKT)** was founded in 2001 as an application oriented R&D institute, whose primary purpose was to assist Upper Austrian SME's with technical problems related to plastics. In the first years up to 2008 the TCKT was a department of the Upper Austrian Research GmbH, which is the leading organization for non-university research of the federal province of Upper Austria. In 2008 the TCKT became a separate legal entity with the main owner being the Upper Austrian Research GmbH. The Johannes Kepler University Linz and the University of Applied Sciences Upper Austria also joined as minority shareholders in 2008. The focus is still to support the SME base in Upper Austria but also SME's and bigger companies in Europe and throughout the world. As mentioned before, the companies in Upper Austria represent the whole value chain starting with the raw material across the wide range of polymer processing to the point of recycling of plastic waste. Along this chain of value the TCKT sets its key areas as material development, material testing, computer aided engineering and process technology. In the area of material development the TCKT has its focus on novel fillers, reinforcements by technical or natural fibers, as well as additives for property modification of polymers.

To verify the results in material development a well-equipped test laboratory was established. In 2004 this test laboratory became an accredited laboratory in which the request from our customers in regards to mechanical, thermal, rheological or physical testing of polymer materials can be fulfilled. In the area of computer aided engineering the scope lies with two different topics. One is the simulation of injection molding processes, and in correlation with the test laboratory the generation of material data for simulation. The other topic is the simulation of strength and stiffness on loaded components made out of polymers. An outlook in this area is the combination of generated material data in process simulation with the input of structural simulation software.

The process technology is represented by investigations on the injection moulding process, the extrusion process and on various processes of composite technologies like the resin transfer moulding (RTM), the vacuum assisted resin infusion (VARI) and the autoclave technology. The first two are dealing with thermoplastic materials and the developments related to the processing of unreinforced and reinforced materials as well as the processing of foamed thermoplastics.
The TCKT is organized by a matrix where the main fields of research overlap the four key areas. The main fields of research at TCKT are:

- Natural fiber reinforced polymers and biopolymers ;
- Material development with additives and fillers in thermoplastics;
- Composite technologies and lightweight structures both in thermosets and thermoplastic materials;
- Interface in composite materials;
- Recycling of polymers, especially the behaviour of multi material compounds.

Methodology

The testing methodology has been based on the European PET Bottle Platform (EPBP) procedure. For these tests the protocol recognised testing levels of 2, 5, 10 and 50% of the tested materials in the recyclates mix. This range has been selected in order to analyse the allowable concentration at low levels and the accumulation effects due to regional market differences across Europe.

Process description:

1. collection of all kind of plastic pre-consumer carrier bags placed on the EU market;
2. preparing mixtures for recycling simulating for end-of-Life scenarios DEG 1, DEG 2, DEG 3, DEG 4;
3. recycling of carrier bags;
4. blown film extrusion;
5. Mechanical testing.

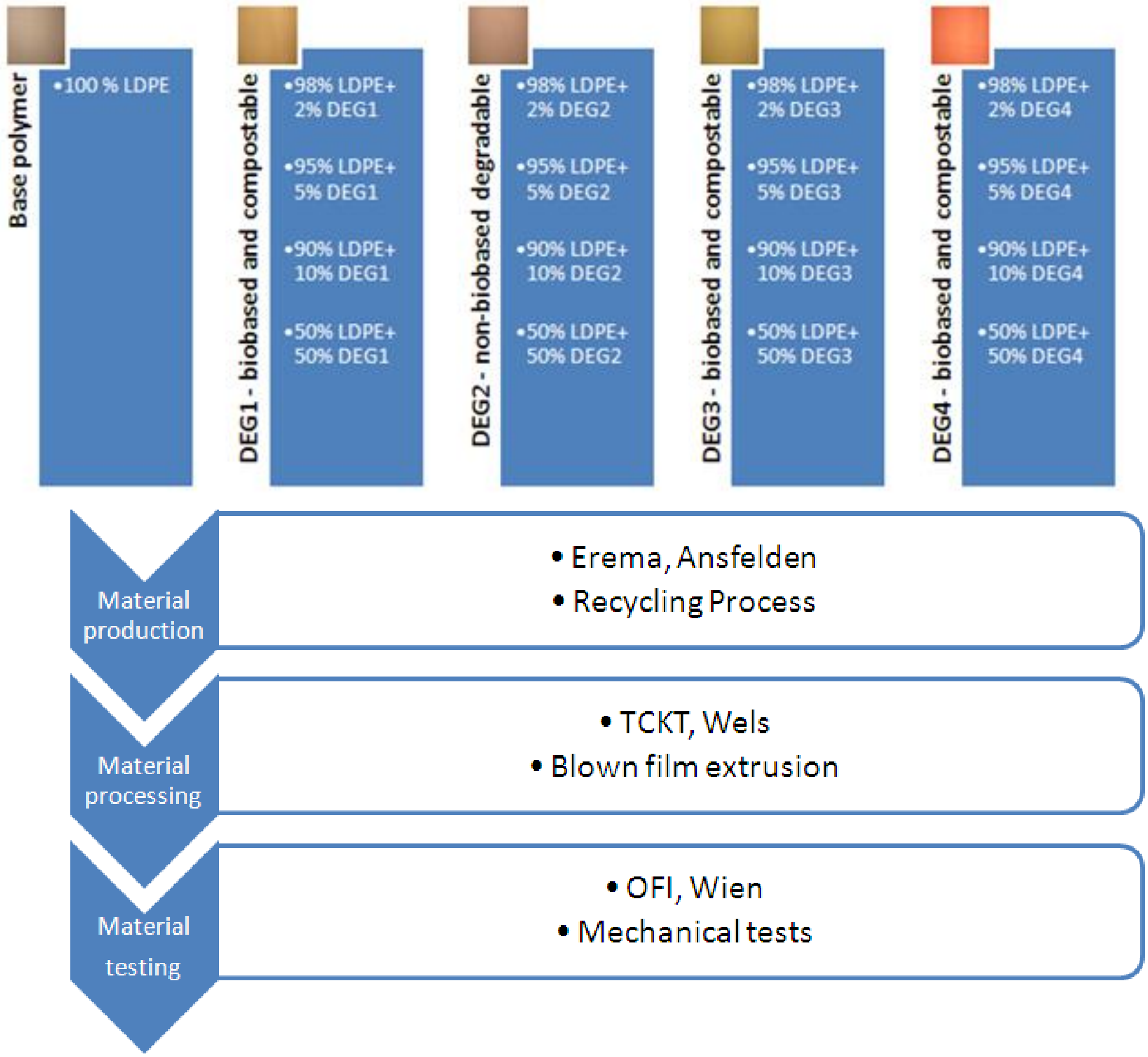

Fig. 1: Overall view materials and processing-steps

Recycling process

The first step of this project consists of the pellet production at EREMA Recycling GmbH. This starts with the production of 100 % LDPE in-house waste (clean and dry- lightly printed) pellets on the Erema 1108TVEplus (Fig. 2). This extrusion machine includes a cutter compactor in which the material is cut, mixed, heated, dried, pre-compressed and buffered. The tangentially connected extruder is filled continuously. In the extruder screw the material is plasticised and degassed in reverse. At the end of the plasticizing zone the melt is directed out of the extruder, cleaned in the filter and returned to the extruder again. The final homogenization of the melt takes place after the melt filter. In the subsequent degassing zone the filtered and homogenized material is degassed. The melt is conveyed by the following metering zone to the water ring pelletizer.

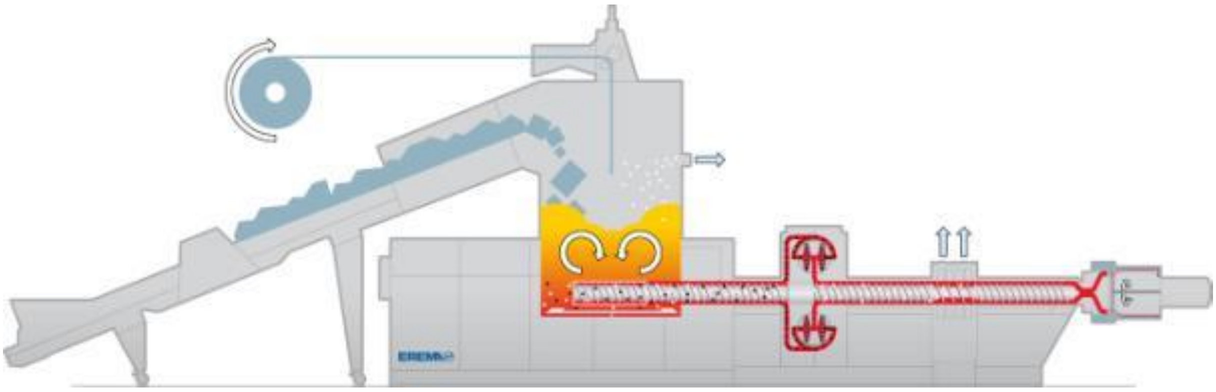

Fig. 2: EREMA 1108TVEplus (extruder screw d: 80 mm, cutter compactor d 1100 mm)

After a constant and stable recycling process was achieved, the different degradable polymers (DEG 1, 2, 3 and 4) were added in varied percentage (2, 5, 10 and 50 %). The processing parameters of all materials were fluctuating, also during the processing on the virgin polymer, as the graphs in Fig. 3 and Fig. 4 show. These inconstancies for the benchmark indicate inhomogeneties which are more or less typical for recycling materials. But for instance the DEG1 material shows a much more important variation in the melt pressure.

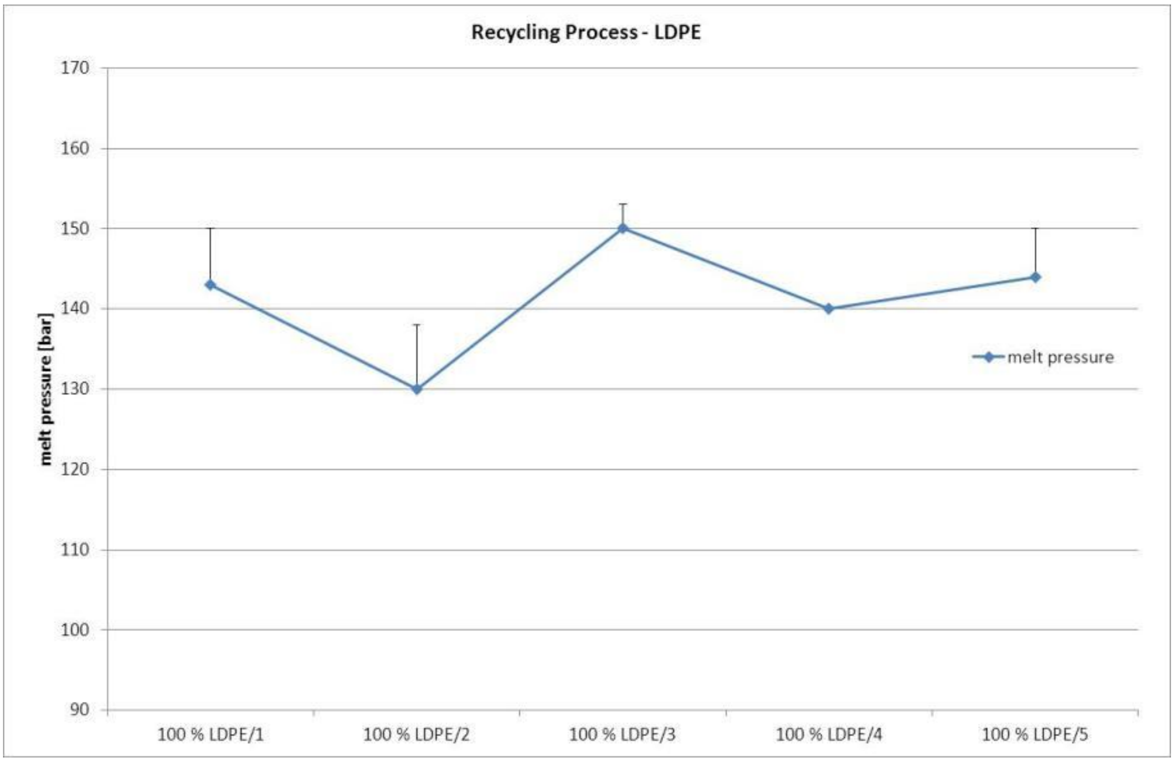

Fig. 3: Recycling process – melt pressure LDPE

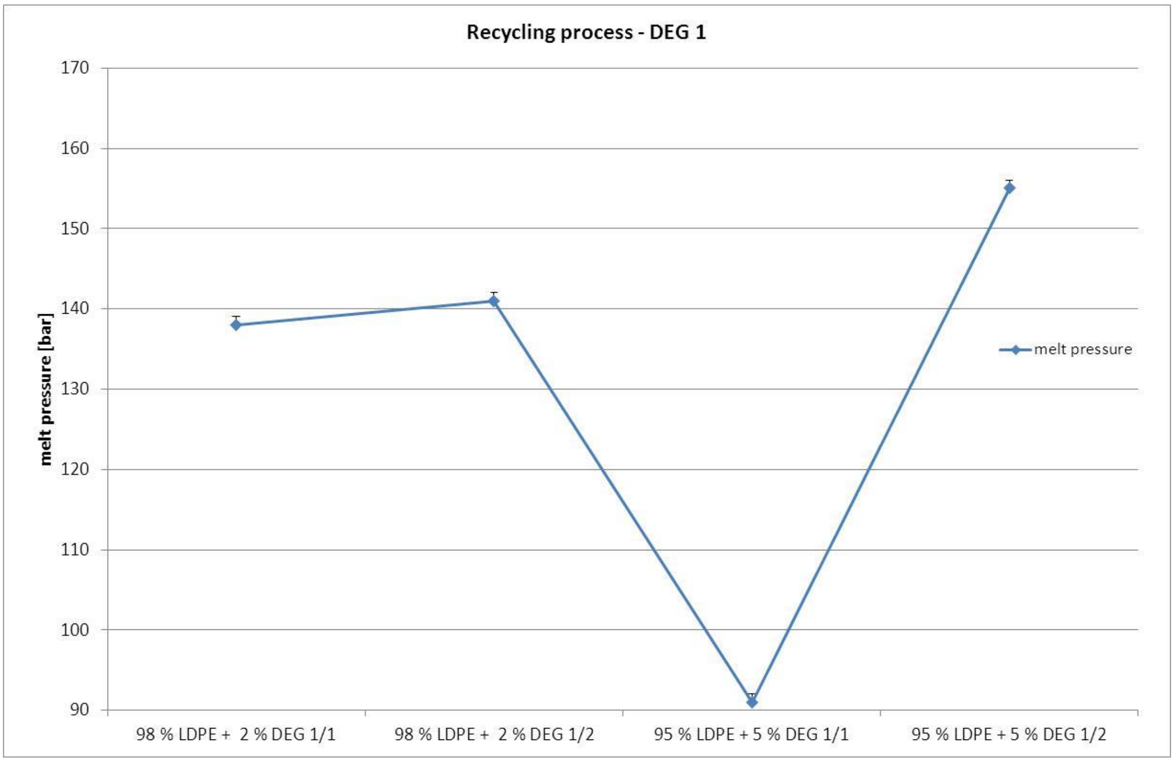

Fig. 4: Recycling process – melt pressure DEG 1

Blown film extrusion

The produced pellets laid the foundation for the next step, the blow-extrusion process. In order to evaluate the production stability during the extrusion process, as well as the visual appearance of the materials and also to get the specimens which are required for the mechanical tests, blown films were extruded from all mixtures.

The production of the blown film was carried out on the blown film extrusion line Hosokawa Alpine HS 35 T (Fig. 5) at the Transfercenter für Kunststofftechnik (TCKT).

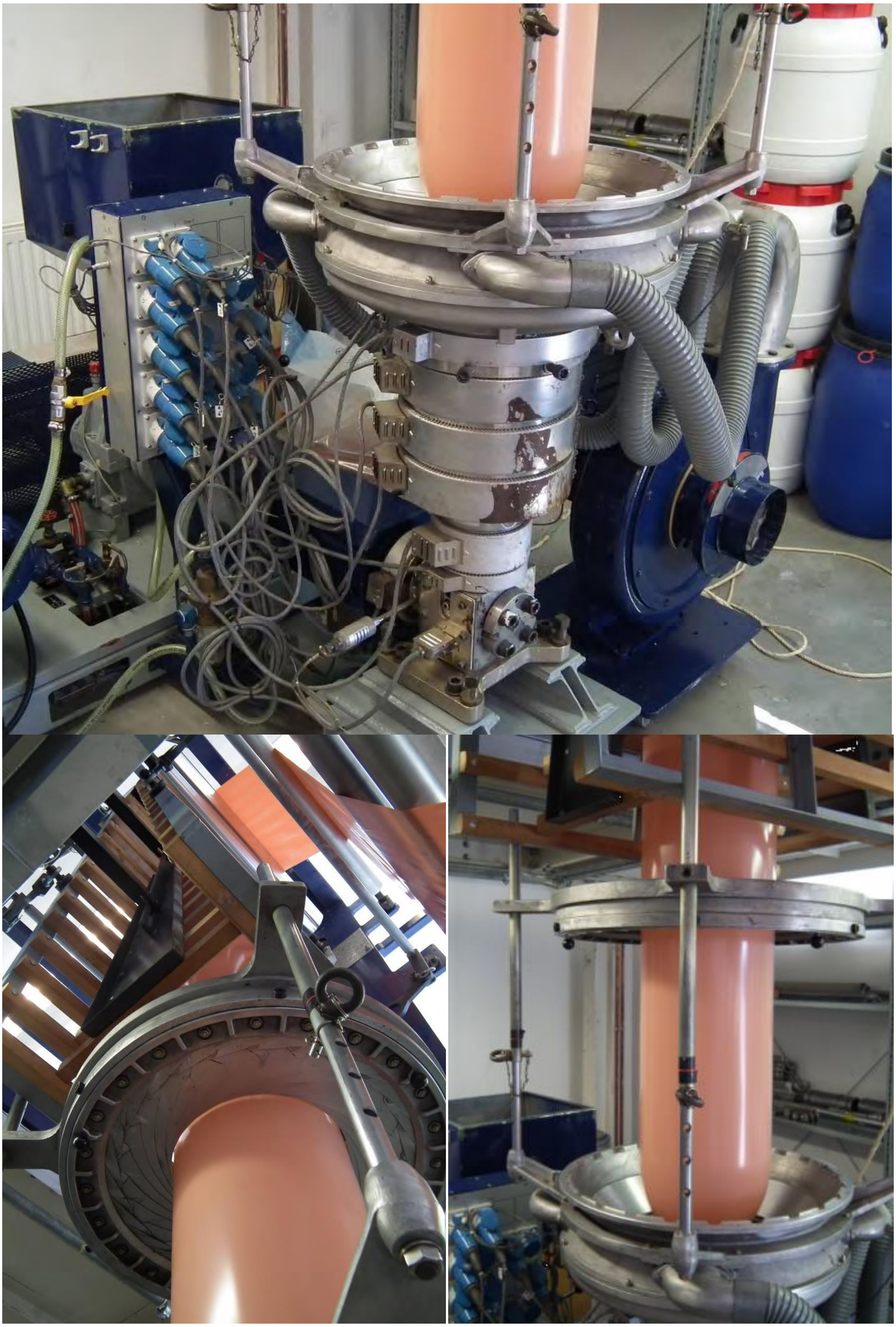

Fig. 5: Blown film extrusion

All materials with 2 % of degradable polymers could be processed without any problems. The materials with 5 % of degradable polymers showed sporadic inhomogeneities which lead to an unsteady bubble and melt pressure varied during the extrusion process. The extrusion process with 10 % DEG runs only for one material without problems, all other materials lead to unsteady bubbles up to the point of collapse. Two of the 50 % mixtures ran stable, the third mixture was creating holes in the film which finally lead to the collapses of the bubble (DEG1).

The following charts (Figure 6 - Figure 9) show the influence of the material quality and the material composition on the process-parameter melt pressure and motor load. The rerun of the 5 % mixture in Fig. 2 & 3 shows that inhomogeneities have an impact on these parameters. In general such inhomogeneities often occur in recycling-materials, leading to irregular film thickness, changes of process parameters and in some cases to the bubble collapse.

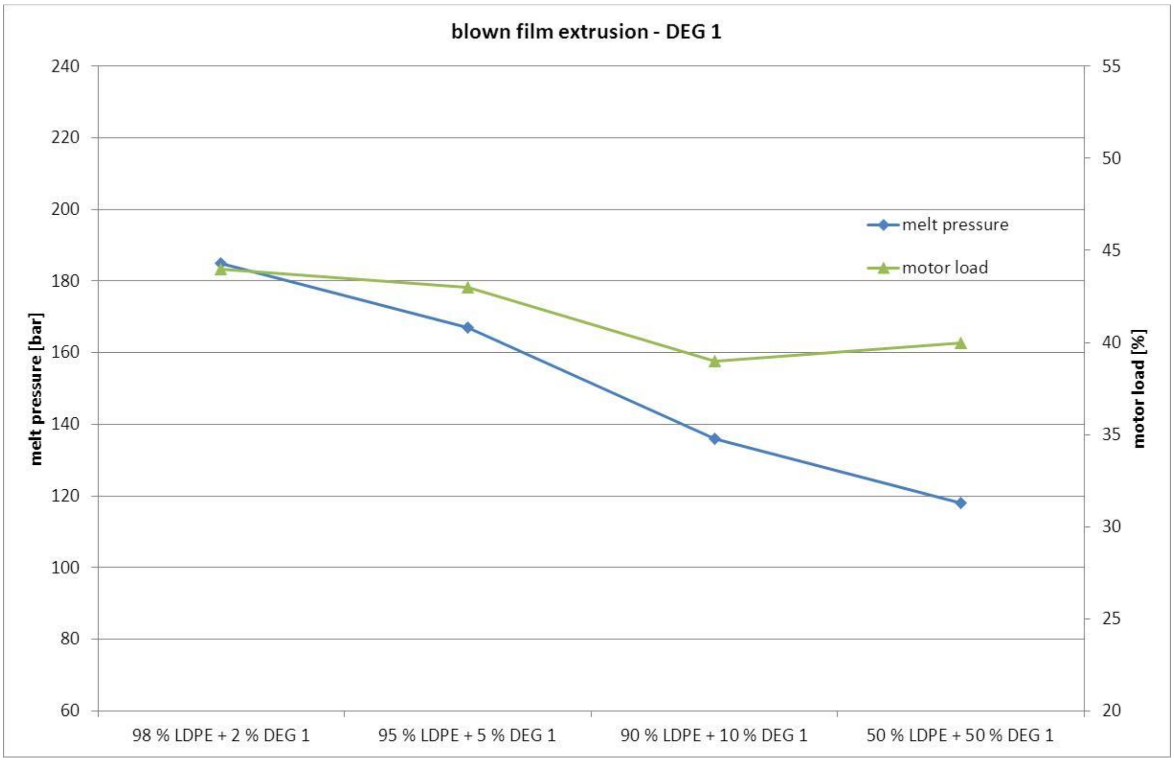

Fig. 6: Process-Parameters – DEG 1

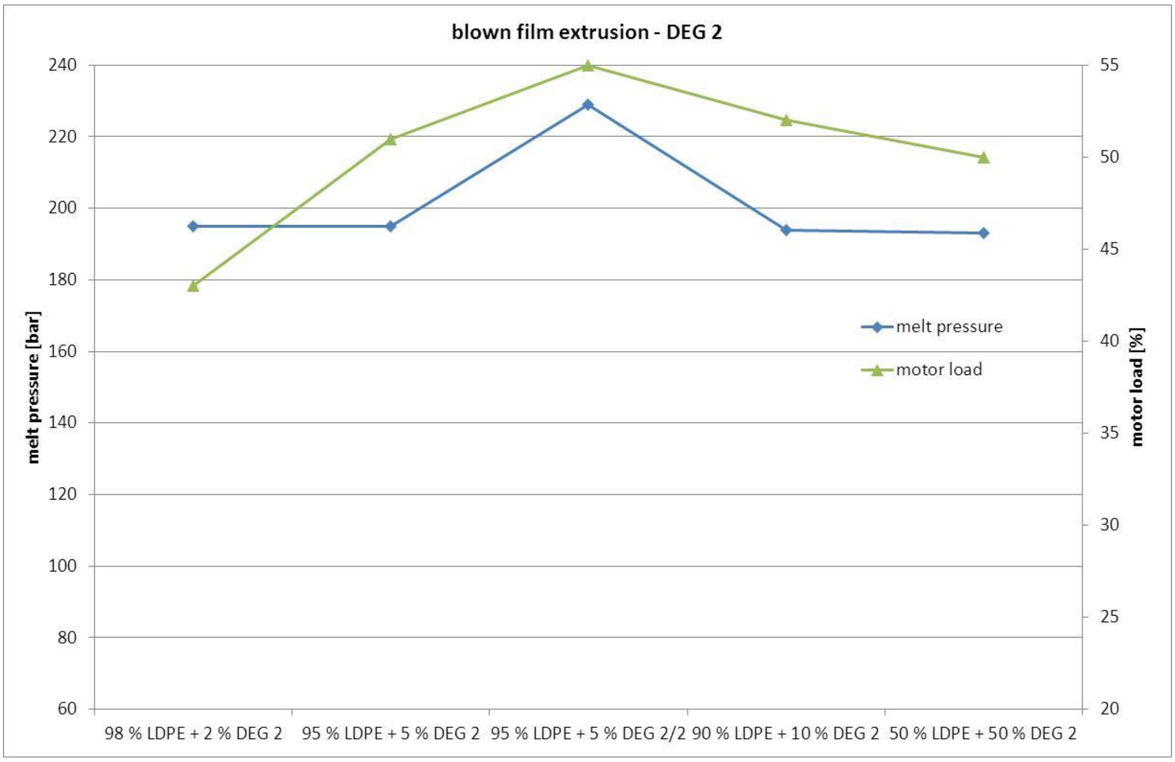

Fig. 7: Process-Parameters – DEG 2

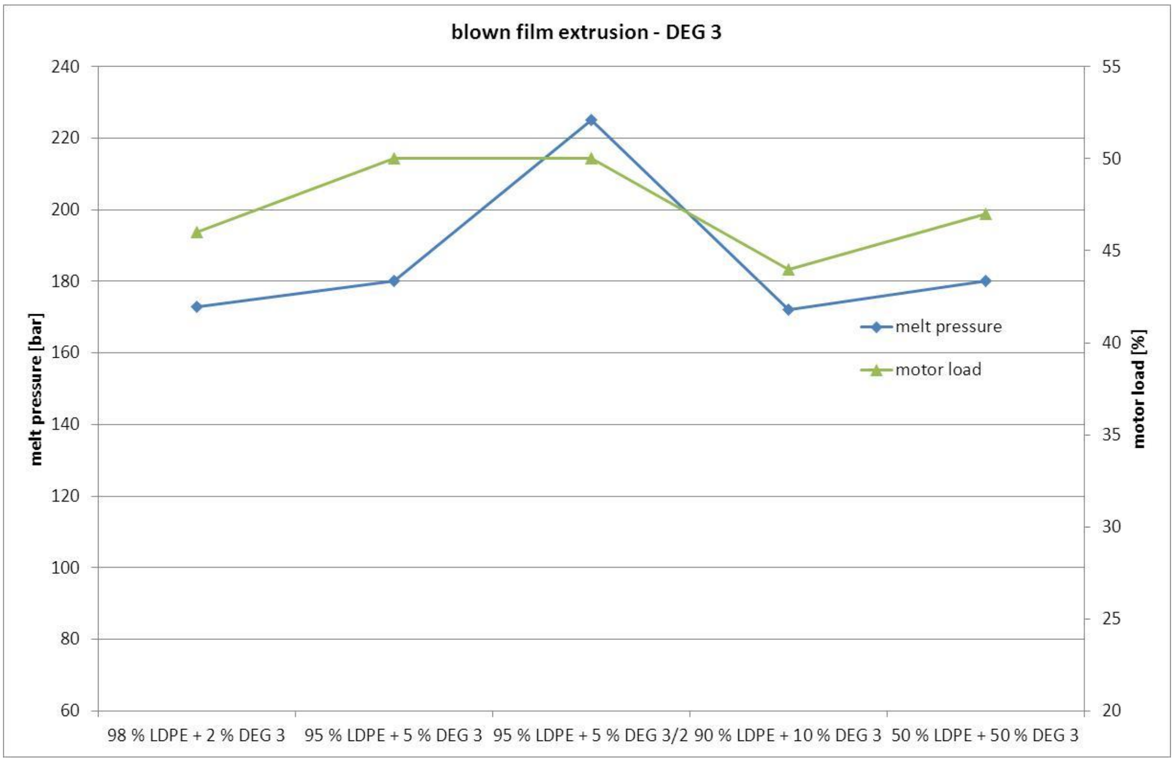

Fig. 8: Process-Parameters DEG 3

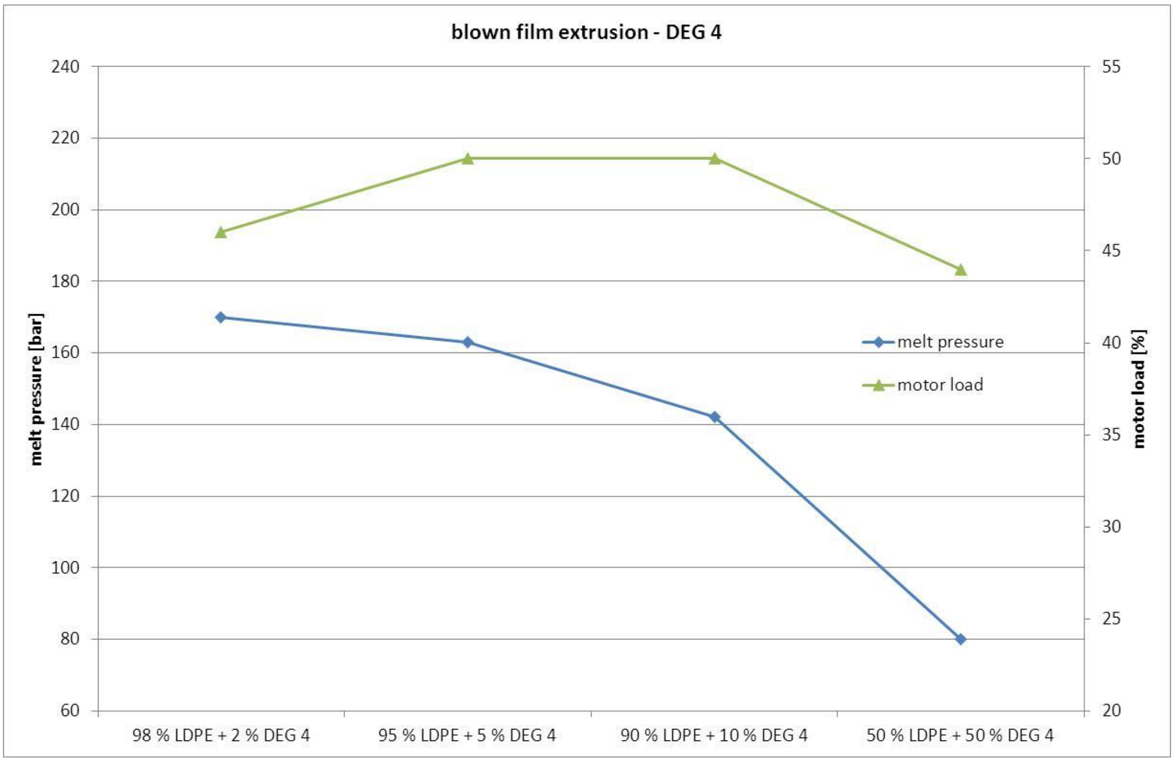

Fig. 9: Process-Parameters DEG 4

The next task was the visual evaluation of the added materials in the blown film. Therefore pictures were made from every blown film, to document the changes of the surface. Mixtures with 2 % DEG express some textures which look like fish eyes. The surface of the blown film with 5 % DEG bear resemblance to sand paper and the 10 and 50 % mixtures exhibit strong fish-scaling look (Figure 10 – Figure 13). In general the colours of the blown films change according to the amount of DEG material, especially with DEG 1 and 4.

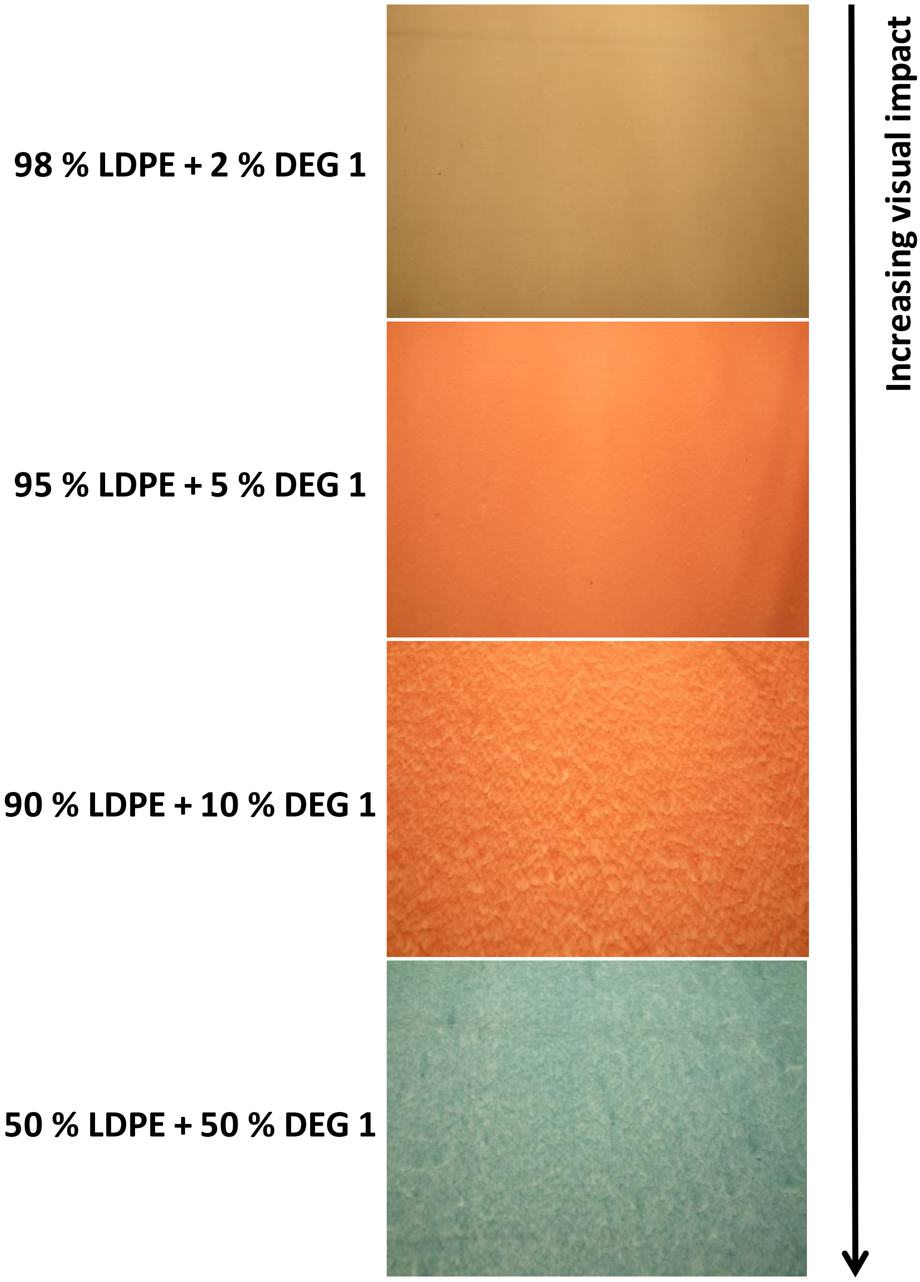

Fig. 10: Visual impact of DEG 1

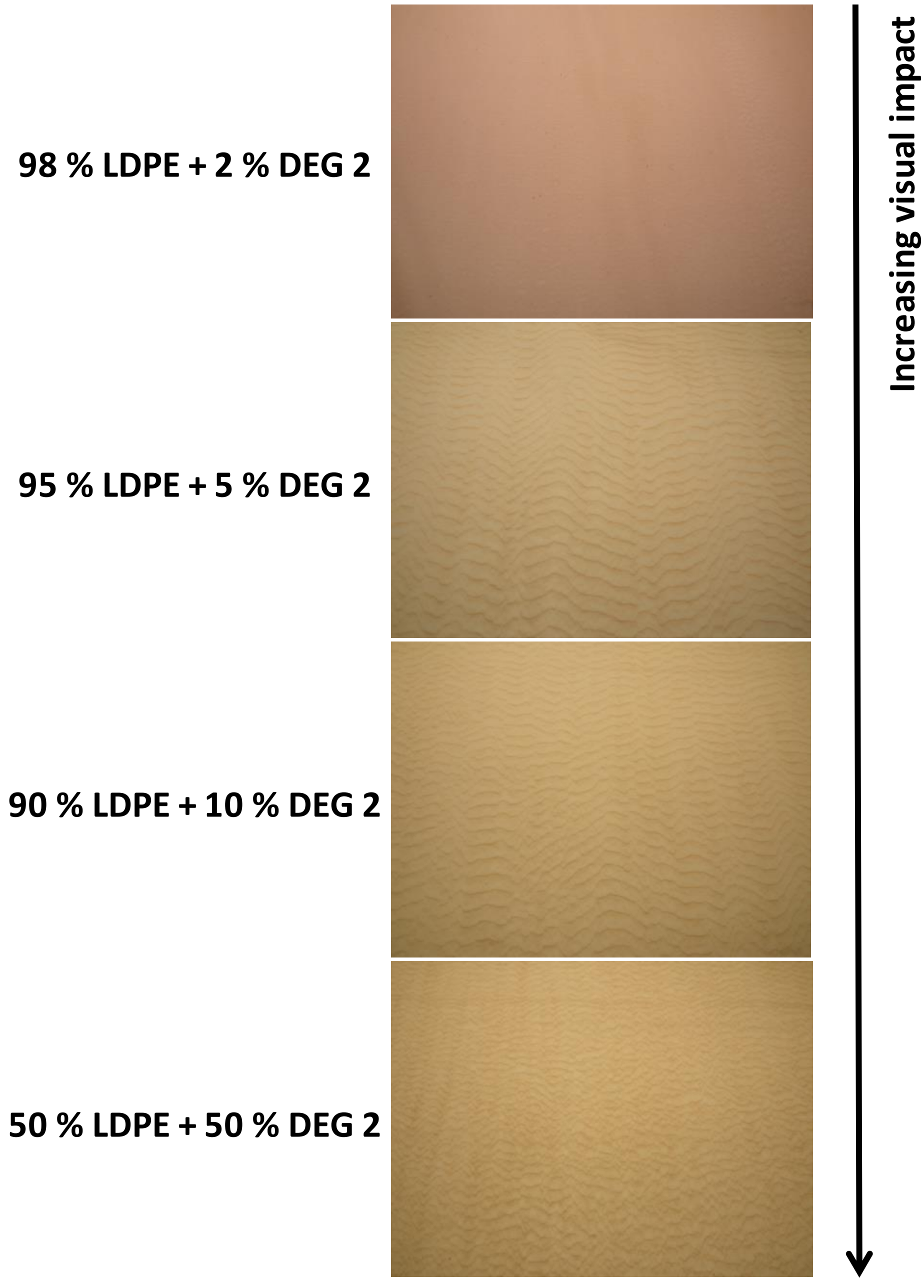

Fig. 11: Visual impact of DEG 2

Fig. 12: Visual impact of DEG 3

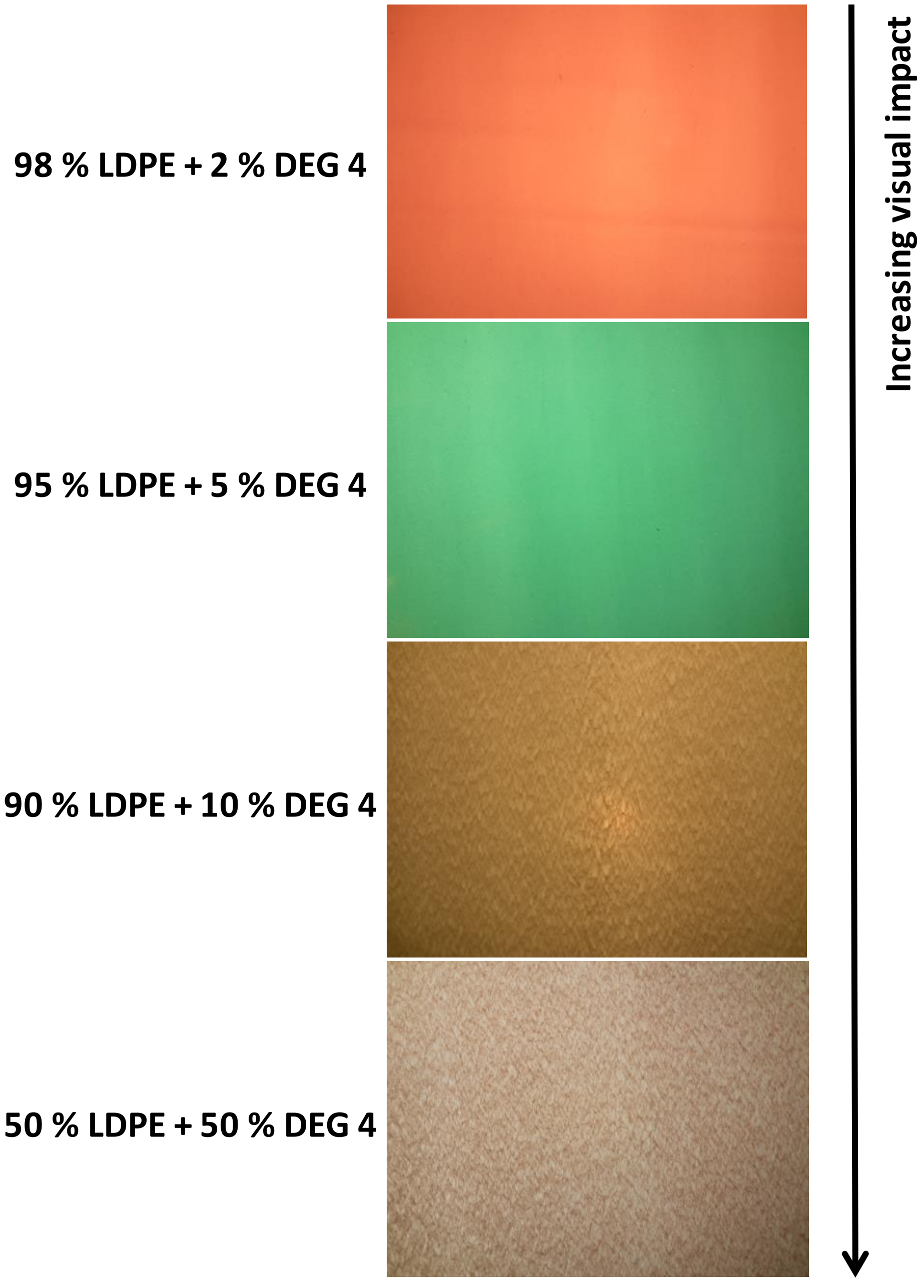

Fig. 13: Visual impact of DEG 4

Testing of mechanical properties

In the next testing steps (testing of mechanical properties), specimens for mechanical tests were punched out of the produced “blown” films.

The mechanical tests were performed at the OFI Technologie & Innovation GmbH. This test series (Fig. 14) included the measurement of the tensile properties (ISO 527-3), the tear resistance (DIN 53363) and the puncture impact (dart drop test ISO 6603-2).

Fig. 14: machines for tensile, tear resistance and puncture impact tests (left to right)

The test results for the different mixtures in comparison with the 100 % LDPE-blown films are shown in Table 1 to Table 4. The light blue coloured values are the starting values, red coloured cells indicate a decrease of the values and dark blue coloured cells point out an increase of the values.

Table 1: results mechanical tests – DEG 1

			100 % LDPE	LDPE +2% DEG1	LDPE + 5 % DEG1	LDPE +10% DEG1	LDPE +50% DEG1
Tensile properties	Machine Direction	σM [MPa]	100%	96%	97%	72%	80%
		εB [%]	100%	131%	141%	76%	178%
	Cross Direction	σM [MPa]	100%	101%	99%	70%	56%
		εB [%]	100%	95%	99%	96%	64%
Tear resistance	Machine Direction	Fmax [N]	100%	100%	99%	85%	73%
		Thickness [mm]	100%	93%	91%	93%	84%
		Tear resistance [N/mm]	100%	105%	107%	93%	88%
	Cross Direction	Fmax [N]	100%	97%	94%	70%	75%
		Thickness [mm]	100%	96%	92%	96%	89%
		Tear resistance [N/mm]	100%	104%	103%	75%	87%
Puncture Impact		Fm [N]	100%	84%	80%	65%	68%
		Thickness [mm]	100%	94%	92%	95%	87%
		Fm/thickness [N/mm]	100%	90%	86%	68%	78%
		Work Wp [J]	100%	73%	76%	62%	140%

Table 2: results mechanical tests – DEG 2

			100 % LDPE	LDPE +2% DEG2	LDPE + 5 % DEG2	LDPE +10% DEG2	LDPE +50% DEG2
Tensile properties	Machine Direction	σM [MPa]	100%	80%	62%	80%	71%
		εB [%]	100%	137%	265%	313%	256%
	Cross Direction	σM [MPa]	100%	90%	56%	83%	77%
		εB [%]	100%	83%	105%	113%	105%
Tear resistance	Machine Direction	Fmax [N]	100%	63%	64%	58%	67%
		Thickness [mm]	100%	78%	108%	86%	85%
		Tear resistance [N/mm]	100%	82%	60%	69%	81%
	Cross Direction	Fmax [N]	100%	73%	93%	76%	77%
		Thickness [mm]	100%	80%	108%	91%	86%
		Tear resistance [N/mm]	100%	93%	89%	86%	91%
Puncture Impact		Fm [N]	100%	74%	35%	56%	50%
		Thickness [mm]	100%	79%	83%	91%	85%
		Fm/thickness [N/mm]	100%	93%	42%	62%	58%
		Work Wp [J]	100%	114%	43%	82%	59%

Table 3: results mechanical tests – DEG 3

			100 % LDPE	LDPE +2% DEG3	LDPE + 5 % DEG3	LDPE +10% DEG3	LDPE +50% DEG3
Tensile properties	Machine Direction	σM [MPa]	100%	77%	62%	79%	82%
		εB [%]	100%	181%	265%	95%	222%
	Cross Direction	σM [MPa]	100%	83%	56%	84%	91%
		εB [%]	100%	91%	105%	85%	101%
Tear resistance	Machine Direction	Fmax [N]	100%	85%	91%	75%	80%
		Thickness [mm]	100%	91%	99%	89%	89%
		Tear resistance [N/mm]	100%	95%	93%	85%	90%
	Cross Direction	Fmax [N]	100%	94%	94%	80%	84%
		Thickness [mm]	100%	98%	103%	92%	91%
		Tear resistance [N/mm]	100%	99%	95%	89%	95%
Puncture Impact		Fm [N]	100%	75%	91%	83%	77%
		Thickness [mm]	100%	95%	97%	91%	91%
		Fm/thickness [N/mm]	100%	79%	94%	91%	85%
		Work Wp [J]	100%	89%	112%	119%	110%

Table 4: results mechanical tests – DEG 4

			100 % LDPE	LDPE +2% DEG4	LDPE + 5 % DEG4	LDPE +10% DEG4	LDPE +50% DEG4
Tensile properties	Machine Direction	σM [MPa]	100%	90%	93%	98%	60%
		εB [%]	100%	120%	220%	72%	71%
	Cross Direction	σM [MPa]	100%	88%	75%	72%	22%
		εB [%]	100%	89%	92%	117%	1%
Tear resistance	Machine Direction	Fmax [N]	100%	93%	94%	122%	83%
		Thickness [mm]	100%	97%	103%	108%	92%
		Tear resistance [N/mm]	100%	97%	92%	114%	91%
	Cross Direction	Fmax [N]	100%	80%	85%	95%	12%
		Thickness [mm]	100%	97%	102%	108%	94%
		Tear resistance [N/mm]	100%	85%	87%	91%	13%
Puncture Impact		Fm [N]	100%	85%	85%	50%	21%
		Thickness [mm]	100%	98%	103%	109%	85%
		Fm/thickness [N/mm]	100%	87%	83%	46%	24%
		Work Wp [J]	100%	76%	80%	59%	27%

Mechanical properties - DEG1:

The material mixtures with 2 and 5 % DEG 1 show only a minimum decrease in tensile strength and an appreciable increase of the breaking elongation in machine direction, but also the mixtures with 10 and 50 % DEG1 show reasonable tensile testing properties.

The tear resistance test shows similar properties; so the mixtures with 2 and 5 % DEG1 have similar tensile properties to the virgin LDPE. The values from the 10 and 50 % mixtures decrease up to 30 %.
A decrease of the puncture impact values, according to the amount of DEG 1, is shown by trend – except the mixture with 50 % DEG1.

Mechanical properties – DEG2:

Mixtures with DEG2 show a decrease of the tensile strength, but a huge increase of the breaking elongation in machine direction.

The tear resistance values decreases between 20 and 40 % in machine direction and about 10 % in cross direction in comparison to the neat polymer.

The puncture impact value of the mixture with 2 % DEG 2 increases, the values from all other mixtures decrease up to 43 %.

Mechanical properties – DEG3:

The tensile properties of these materials are similar to the values of the mixtures of DEG2, but the tear resistance values range from 85 to 99 % in comparison to the neat polymer.

The puncture impact value of the 5, 10 and 50 % mixtures rises up to 119 %, only the 2 % mixture shows a lower value.

Mechanical properties – DEG4:

The results of the tensile tests of the 2, 5 and 10 % DEG 4 mixtures shows only a small decrease of the tensile strength, but a significant increase in the breaking elongation. The 50 % mixture exhibits low tensile properties.

The tear resistance values in machine direction are located in the range of the neat polymer (from all mixtures). Also the tear resistance values in cross direction, from the mixtures with 2, 5 and 10 %, DEG 4 show only a small decrease, but the 50 % DEG 4 mixture significantly drops.

At the puncture impact tests the values of the 50 % DEG 4 mixture also drop, the values of all other mixtures ranges between 59 and 80 % in comparison to the benchmark (neat polymer).

Conclusion

Finally it can be concluded that in general, the recycling process of the different materials worked without major complications. However, there were difficulties (unsteady bubble and melt pressure varied during the extrusion process) when it came to the extrusion of new blown film with materials containing more than 5% DEG material. The fluctuations of the process parameters during the recycling process and the blown film extrusion might also be due to the inhomogeneties in the input materials.

The mechanical properties off all mixtures decrease in most cases, except the breaking elongation which is, especially in machine direction, much higher than the breaking elongation of the virgin LDPE. It should be noted that measures on mixtures with 50% DEG material had significant lower mechanical properties. However, the visual impact and the mechanical properties already decreased at the 2 % mixtures – at all formulations.
Whatever degradable or oxo-fragmentable material enters the recycling stream; these tests show that these materials are conflicting with the recycling process of conventional material.

So in conclusion, it can be said that as little as 2% oxo-fragmentable or degradable materials in the recycling streams can affect the quality of recyclates, hence the production of a new PE film.

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