



Use of recycled materials as mid layer in three layered structures-new possibility in design for recycling

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ABSTRACT

Challenges of using recycled polymer materials in food packaging are related both to the properties of the material and demanding safety issues for food contact materials. The aim of this research was to detect possible risks using recycled post-consumer plastic waste in three-layered polyethylene (PE) films produced with virgin outer layers and a mid-layer from recycled flexible PE material. Migration tests were performed as risk assessment for identifying possible migrating substances from the mid-recycled layer. Overall migration was determined on various 3-layered structures made from virgin low-density PE (LDPE) and virgin linear low-density PE (LLDPE), respectively, with two different recycled PE from post-consumer waste (R1 and R2). 95% ethanol was used as a simulant for screening at 20, 40 and 60 °C for 10 days. Specific migration was performed using sophisticated analytical techniques (liquid and gas chromatography). Overall migration values for all temperatures were from $< 2 \text{ mg/dm}^2$ at lowest and $5.6 \pm 0.1 \text{ mg/dm}^2$ at highest temperature, thus well below the overall migration limit (OML) of 10 mg/dm^2 . Specific migration of intentionally and non-intentionally added substances were detected, identified and quantified. Similarly, specific migration values were under specific migration limit (SML) defined in legislation. Optical properties of the reference samples were significantly different in appearance and on cross section for samples with recycled materials R1 compared to recycled material R2 which caused only slight changes in the appearance of both reference films. This research presents a valuable study on migration from recycled flexible PE films and use of recycled materials in food packaging applications. Use of virgin materials as outer layer in combination with commercially available recycled material can increase the use of recycled materials in packaging as well as reduce the total amount of flexible plastic solid waste.

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1. Introduction

The food packaging global market has been growing rapidly in the last decades, leading to the development of diverse packaging materials, packaging solutions as well as new technologies and innovations (Ahvenainen, 2003; de Oliveira and de Melo, 2019; Marsh and Bugusu, 2007; Robertson, 2005; Siracusa and Lotti, 2019; Vilela et al., 2018; Yildirim et al., 2018). Up to date, most of the technologies and processes were designed as linear models

from production, their use to disposal. The linear model has led to a huge amount of food packaging materials ending up as municipal solid waste, not reused or recycled but sent to landfill or incineration (Brouwer et al., 2018; Martinho et al., 2017; van Velzen et al., 2019).

Nowadays the circular economy concept has been spread as a quality concept followed in all spheres of life, as well as in food and packaging industry (Geueke et al., 2018; Van Eygen et al., 2018). According to circular economy principles, food packaging materials should be brought back into the stream, as new packaging solutions -either reused or recycled (MacArthur et al., 2016). Due to reduction of environmental impact of food packaging, different efforts are already undertaken with the aim to increase recycling rates

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(Commission, 2018; Groh et al., 2019). Nevertheless, the concern of food safety increased with specific focus on diverse chemical compounds as potential migrants from food packaging materials (Geueke et al., 2018; Groh et al., 2019; Ibarra et al., 2019; Muncke et al., 2017).

Food contact materials are strictly regulated with limitations in use of recycled materials (Commission, 2008). Regulation EU 2008/282 sets up criteria on evaluation of feedstock and recycling processes intended for use as food contact materials. To date more than 100 recycling companies have obtained a temporary approval from EFSA, and waiting for final decision from European Commission (EC). Most of these companies are dealing with PET bottles as feedstock, and only a few with rigid polyolefin packaging (HDPE and PP) (EFSA Panel on Food Contact Materials and Aids, 2013; EFSA Panel on Food Contact Materials et al., 2018). However, to our knowledge, they are not dealing with flexible PE packaging, and there are no recycling companies with a temporary approval of a feedstock-process combination for PE film materials. Overcoming these challenges could be possible with new approach and design for recycling, as contamination of the food through migration of chemical substances could be reduced by using alternative design of packaging itself. One of the possibilities of using recycled materials in food packaging applications is to avoid direct contact of recycled materials with food, but rather use it as core layer in a multilayered structure (Chytiri et al., 2006). Material safety as well as its compliance with regulations includes monitoring of overall migration and specific migration of different chemicals (Geueke et al., 2018; Groh et al., 2019; Reinas et al., 2012). These data are usually obtained through experiments of migration from food contact materials into food simulants under controlled time and temperature. Possible migration of different chemical compounds from food packaging to packed food can be related to food type. However, non-intentionally added substances (NIAS) are present also because of degradation or interaction of additives commonly added to polymers in the production process (Geueke et al., 2018; Groh et al., 2019; Ibarra et al., 2019; Leeman and Krul, 2015; Nerin et al., 2013). NIAS are usually detected with sophisticated analytical techniques (such as gas and liquid chromatography) and any potential risk in the final material or article coming from NIAS must be considered (Ibarra et al., 2019; Sanchis et al., 2017).

Polyolefins (PO) represent almost a 40% share in the total packaging market. They are recyclable and can easily be brought back to the packaging stream, so their reuse and recycling is in the focus of polymer producers nowadays (Hu et al., 2013; Luijsterburg and Goossens, 2014; Serranti et al., 2011, 2015).

It is well known that polyolefins are prone to degradation during compounding, converting to final products and end use, resulting in a change in molecular weight and molecular weight distribution (Coulter et al., 2007). For this reason, they are usually stabilized with different additives like anti-oxidants, UV absorbers, light stabilizers and processing stabilizers (Ambroggi et al., 2016; Coulter et al., 2007; Sanchis et al., 2017). These substances are intentionally added substances (IAS) in polymer production and are listed in Commission Regulation (EU) No 10/2011 in Annex I in the positive list of substances with defined specific migration limits (Commission, 2011). However, substances that are not listed in the regulations, are non-intentionally added substances, and thus must be considered in risk assessment analysis (Coulter et al., 2007; Geueke et al., 2018; Groh et al., 2019; Ibarra et al., 2019; Lau and Wong, 2000; Leeman and Krul, 2015; Muncke et al., 2017; Nerin et al., 2013). Such substances are very often expected to be found as they are present as e.g. degradation products of different additives. Presence of oligomers (molecules that consist of relatively few repeating units) in polymeric materials are also common and expected thus also considered as NIAS (Hoppe et al., 2016).

There are several studies on use of multilayered structures as functional barriers with artificial contaminants in mid layer (Arvanitoyannis and Bosnea, 2004; Franz et al., 1994), use of PP as functional barrier film to recycled paperboards (Song et al., 2003) and PE as functional barrier in multilayered structures (Badeka et al., 2003; Chytiri et al., 2006). Even though polyolefins are prone to diffusion (e.g. migration is higher for these materials), they can act as protective layers in multilayered structures both for non-contact use of recycled materials for protection of unwanted migrant. However, to date these researches were not focused on the detailed risk assessment analysis and determination of both intentionally and non-intentionally added substances and their effect.

In line with state-of-the-art on circular economy and the use of recycled materials, trends are moving towards design for recycling. The aim is to upgrade the recycled materials so that they can be used in food packaging applications (Steenis et al., 2018).

To the best of our knowledge there are no available migration data from recycled flexible PE films sourced from post-consumer waste (PCW). The goal of this study was to assess the risk for multilayered polyethylene films with post-consumer polyethylene as an internal layer. Technical suitability of PCW as well as its safety in use as food packaging material was evaluated. Migration tests were performed on the prepared films as risk assessment for identifying possible migrants from the recycled mid-layer. This research presents a valuable study on migration from recycled PE flexible films and clear insight in the level of compliance we can achieve with recycled plastics with regard to OM, SM and NIAS. In addition, it clearly shows limitations of circular economy for recycling of flexible PE used in food packaging with currently available technologies. This particular research can provide guidelines in design for recycling and reduction of flexible plastic waste, ensuring responsible consumption and production in line with United nations development goal 12 (Assembly, 2015). Use of virgin materials as outer layer in combination with commercially available recycled material can increase the use of recycled materials in packaging as well as reduce the total amount of flexible plastic solid waste.

2. Materials and methods

2.1. Materials

Multilayered films were prepared from two virgin and two recycled PEs available on the market. Both virgin linear low-density polyethylene (LLDPE) and low-density polyethylene (LDPE) are commercially available products.

The virgin LDPE, (density 923 kg/m³) is developed for packaging film applications, like shrink film (lamination films) and carrier-bag film and pouches (food packaging). It contains no additives. The product is suitable for recycling.

The virgin LLDPE, (density 931 kg/m³) is developed for packaging application. It contains additives and is suitable for recycling.

Recycled material 1 (R1) is commercially available recycled material from PCW (density 930 kg/m³). According to the producer it is produced using a special blend of LDPE and LLDPE post-consumer recycled plastic; well suited for blown film processing and recommended for applications like carrier bags and other flexible packaging applications.

Recycled materials 2 (R2) is commercially available custom-made blend of PCW and post-industrial polyethylene (density 927 kg/m³).

Eight different films were made from virgin and recycled materials with 20 µm on the outer layers and 40 µm in the mid-layer,

Table 1
Three-layer films: list of samples and construction of multilayered PE films.

Sample mark	Outer layer 1 (20 µm)	Mid layer (40 µm)	Outer layer 2 (20 µm)
LDPE	LDPE	LDPE	LDPE
LLDPE	LLDPE	LLDPE	LLDPE
LDPE/R1	LDPE	R1	LDPE
LDPE/R2	LDPE	R2	LDPE
LLDPE/R1	LLDPE	R1	LLDPE
LLDPE/R2	LLDPE	R2	LLDPE
R1	R1	R1	R1
R2	R2	R2	R2

each having a total film thickness of 80 µm (Table 1). Three layered films used in the experiments were produced on a Varex 50/60/50 3-layer coextrusion film line from Windmüller & Hölscher, with a 200 × 1.2 mm die.

2.2. Determination of chemical compounds present in multilayered PE films

Detection and quantification of additives and other substances present in the samples were performed by microextraction of grinded pellets (data are presented as mean values of three replicates with standard deviations) and Soxhlet extraction of the prepared films (one measurement per sample for confirmation from microextraction).

For the microextraction procedure pellets were milled under cryogenic cooling by liquid nitrogen to powder. The powder (0.5 ± 0.0001 g) was extracted in a mixture of two organic solvents at their boiling temperature, for 90 min. The samples were left to cool down, filtered and transferred to vials for further analysis.

Soxhlet extraction principle is continuous extraction of the solid by repeated boiling-condensation of a solvent in such a way that the extraction fluid is continuously refreshed. During extraction, the target analytes are exposed to pure, clean, hot solvent on each pass, and the soluble components leach out of the polymer. Three-layered films were placed in a metal filter and extracted with chloroform as solvent for 5 h at the boiling point. The equipment was left to cool down for 5 h, then sample volumes were measured, and transferred to vials for further analysis. List of chemical compounds detected by microextraction and Soxhlet extraction is presented in Table 2.

2.3. Migration analyses

Migration testing was done according to Commission

Regulation EU 10/2011 (Commission, 2011) for plastic materials and articles intended to come into contact with food. Migration experiments were carried out by cell method of 1.9 dm² (both sided measurement) of samples in 100 ml of 95% ethanol simulant recommended for screening (by EFSA) at 20, 40 and 60 °C for 10 days (EFSA Panel on Food Contact Materials and Aids, 2016).

To carry out the migration analysis the films were cut into circular pieces with diameter 11 cm and transferred into migration cells with a volume of 100 ml. The migration cells were filled to the brim with 95% ethanol as a food simulant, bringing it in contact with the films at both sides. The migration cells were then sealed and kept in an oven at constant temperature (20, 40 and 60 °C) for 10 days. Blank samples were exposed to the same procedure and test conditions. All migration experiments were carried out in triplicate, presented as mean values with standard deviations.

2.3.1. Overall migration test

Overall migration test was determined according to European standard method EN 1186. After the storage period the migration extracts were evaporated to dryness and the residues were weighed. The overall migration from the films is determined as the mass of residue after evaporation of the food simulant and reported as the mean of three determinations on separate tests. Overall migration was expressed in mg/dm² for samples where measured values were below 2 the results are presented as <2

2.3.2. Specific migration test

Specific migration analysis was carried out according to European Standard EN 13130. The residues obtained after overall migration were dissolved in 3 ml chloroform and transferred to suitable vials for further chromatographic analysis. Chromatographic analysis was performed to identify the migrants from the films for the specific migration. High Performance Liquid Chromatography (HPLC) separation was carried out on Agilent HP1100 Series system with Agilent 1200 UV detector (DAD), ZORBAX Eclipse XDB-C18 column operated at 50 °C by gradient elution. The flow rate was 1 ml/min and injection volume 10 µl. An Agilent 6890N coupled to an Agilent 5973 Network Mass Selective Detector and a Gerstel MPS2 Autosampler was used for the Gas Chromatography-Mass Spectrometry (GC-MS) analysis. For separation Zebtron ZB-5MSPlus column (30m × 0.25 mm × 0.25 µm, Phenomenex) was used split injection (2:1 ratio) with temperature range from 60 °C to 300 °C at a heating rate of 10 °C/min, total run time 40 min.

The chromatograms obtained from the extracts, reference materials and materials with recycled mid layer were compared to find

Table 2
List of chemical compounds with codes detected by microextraction and Soxhlet extraction.

Commercial name/standard name	IUPAC name of the substance	Codes used	CAS number	Listed in Annex I	SML (mg/dm ²)	SML defined (mg/kg)
Antioxidant P168	Tris (2,4-di- <i>tert</i> -butylphenyl)phosphite	P168	31570-04-4	Yes	10	60
Antioxidant AO1010	Pentaerythritol tetrakis (3-(3,5-di- <i>tert</i> -butyl)-4-hydroxyphenyl)-propionate	AO1010	6683-19-8	Yes	10	60
Antioxidant AO1076	Octadecyl-3-(3,5-di- <i>tert</i> -butyl-4-hydroxyphenyl)-propionate	AO1076	2082-79-3	Yes	1	6
P168 oxidized	Tris (2,4-di- <i>tert</i> -butylphenyl)phosphate	P168	95906-11-9	No	/	/
Arvin 4	2,4-di- <i>tert</i> -Butylphenol	Arvin 4	96-76-4	No	/	/
Arvin 8	7,9-Di- <i>tert</i> -butyl-1-oxaspiro (4,5)deca-6,9-diene-2,8-dione	Arvin 8	82304-66-3	No	/	/
Diethylhexyl phthalate (DEHP)	Phthalic acid, Bis (2-ethylhexyl) phthalate	DEHP	117-81-7	Yes	0.25	1.5
Diethylhexyl terephthalate (DEHT)	Terephthalic acid, Bis (2-ethylhexyl) terephthalate	DEHT	6422-86-2	Yes	10	60
Oligomers	Standard for oligomers C20 Eicosane C ₂₀ H ₄₂	Oligomers	112-95-8	No	/	/

differences between extracts and film samples. Quantification was done using standards and according to calibration curves and response factors for each identified additive and their degradation products. Identification or classification was carried out with obtained mass spectra and available databases. Specific migration was expressed in mg/kg of food.

2.4. Microscopy cross section of the film sample

Light microscopy measurements of cross sections of all prepared three-layered films were performed by using LM-light microscope Axiopot (Carl-Zeiss, Oberkochen, Germany). The microscopy light is transmitted light with contrast technique bright field/pol. Prior to the microscopy measurements all samples were prepared on a rotary microtome Leica RM 2165 (Meyer Instruments, Inc; Houston) under ambient temperature (23 °C). Cross section of films is presented in 500X magnification.

2.5. Surface color

Professional photographs of 8 3-layered samples has been conducted for visual impression and evaluation.

Visual color was measured for all the prepared samples with a Konica Minolta Chroma Meter (Tokyo, Japan). The L^* (lightness), a^* (red-green) and b^* (yellow-blue) were read using a D_{65} light source against the white calibration plate. Color difference between two samples (ΔE_{ab}^*) was calculated using the following equation:

$$\Delta E_{ab}^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \quad (1)$$

where ΔL^* is the difference in L^* (lightness) between two samples (comparison between reference materials and different multilayered structures with recycled materials) while, similarly, Δa^* and Δb^* are the differences in the coordinates a^* (green/red) and b^* (blue/yellow), respectively. For each film, 5 readings were taken, and the average values were determined. ΔE_{ab}^* below 0.5 indicates an imperceptible difference in color, 0.5–1.5 a slight difference, 1.5–3.0 a noticeable difference, 3.0–6.0 a marked difference, 6.0–12.0 an extremely marked difference, and above 12.0 a color of a different shade (Belović et al., 2014).

2.6. Statistical analysis

Analysis of variance has been conducted in Minitab 18. One-way ANOVA was performed for overall and specific migration values (at each temperature) and color measurement. A general linear model (GLM) including temperature and sample and their interactions, was applied for the overall migration (OM). In addition, not standardized PCA analysis on covariance matrix was performed by using the statistical data analysis software system XLSTAT, version (2018.7).

3. Results and discussion

Prior to migration test two extraction techniques were used for determination of chemical compounds present in virgin and recycled PE. From the extraction analysis of all samples chemical compounds that are present have been detected and quantified on HPLC and GC-MS analytical equipment.

Intentionally added substances (IAS), additives P168, AO1010, AO1076 are detected and listed in Annex I. The two phthalates DEHP and DEHT are also listed, but in recycled polyethylene they are considered as NIAS. All SML for detected chemical compounds listed in regulation are presented in Table 2.

P168-ox, Arvin 4 and Arvin 8 as well as oligomers are not listed

in Annex I and are considered as NIAS. Microextraction method has been developed for determination of additives present in different polymeric samples.

Chemical compounds detected and quantified (ppm) by HPLC and GCMS from microextraction of LDPE, LLDPE, R1 and R2 pellets are presented in Table 3.

The LDPE reference material did not contain any chemical compounds in extracted samples as expected, because this material does not contain any additives (according to the technical data sheet). The LLDPE sample contained additives that are usually added in production of polyethylene such as the antioxidants P168 and AO1010. Besides the additives degradation product of additive P168 (P168-oxidized) as well as oligomers were extracted from the samples. Recycled samples (both R1 and R2) showed presence of additives usually added to polyolefins during productions (P168, AO1010 and AO1076). In addition, presence of some non-intentionally added substances (NIAS) was also detected. From GC-MS analysis presence of the so-called Arvin substances (Arvin 4 and 8) and two phthalates (Diethylhexyl phthalate (DEHP) and Diethylhexyl terephthalate (DEHT)) were detected. Additives detected by HPLC (P168, P168-oxidized and AO1076) were confirmed by GCMS.

Additionally, Soxhlet extraction of films has been carried out as confirmation of microextraction results, but also for precision in the determination of possible migrants from specifically constructed multilayered structures (Table 4).

Representative chromatograms (chosen sample LLDPE/R2) from HPLC and GC-MS are presented in Figs. 1 and 2 respectively.

Standard stock solutions for specific chemical compounds P168, AO1010, AO1076, oligomers (Eicosane standard), P168-ox and Arvin 4 were prepared from commercially available standards. The response factor from the standard Bis(2-ethylhexyl) isophthalate was used as ad hoc quantification of DEHT and DEHP and they were identified by internal library; Arvin 8 was identified by internal library.

Calibration curves and response factor were calculated from different dilutions from stock solutions and used to calculate the concentrations of specified substances.

3.1. Overall migration

Overall migration values for all samples at 20, 40 and 60 °C after 10 days exposure to 95% ethanol are presented in Table 5. OM values for all measured samples were below 10 mg/dm² implying that all samples are in compliance with regulatory limit for overall migration.

Virgin LDPE and LLDPE material had similar OM values at 20 °C (at lower temperatures it's expected to have lower migration and less pronounced influence of polymer type) while at 40 and 60 °C the OM values of the LLDPE were considerably higher than for LDPE. This dependence can be partly assigned to the polymer characteristic as LDPE reference material contained no additives, while LLDPE was produced with addition of antioxidants. Also, harsh conditions that were applied (95% EtOH) can influence higher levels of oligomers as the polymer is partly dissolved under these conditions (Geueke et al., 2018). This influence is more pronounced for LLDPE compared to LDPE due to different polymer types and sensitivity to specific solvent and conditions (95% EtOH and high temperature) (Arvanitoyannis and Bosnea, 2004; Nerin et al., 2013). Both polymer systems with recycled R1 (LDPE/R1 and LLDPE/R1) showed higher OM values than with recycled material R2 (LDPE/R2 and LLDPE/R2) meaning that the migration levels are related to type and quality of recycled polymer too. Temperature increase is influencing the OM levels in all samples. Overall migration values for all temperatures were from <2 at lowest and 5.6 ± 0.1 mg/dm²

Table 3

Chemical compounds detected and quantified (ppm) by HPLC and GCMS from microextraction of LDPE, LLDPE, R1 and R2 pellets.

	P168 HPLC	P168-ox HPLC	AO1010 HPLC	AO1076 HPLC	Oligomers GCMS	Arvin4 GCMS	Arvin8 GCMS	DEHP GCMS	DEHT GCMS
LDPE	nd	nd	nd	nd	nd	nd	nd	nd	nd
LLDPE	920 ± 5	30 ± 0	210 ± 0	nd	2200 ± 113	nd	nd	nd	nd
R1	50 ± 5	540 ± 2	30 ± 0	200 ± 0	300 ± 66	1 ± 1	7 ± 1	14 ± 0	35 ± 15
R2	40 ± 0	520 ± 7	20 ± 0	280 ± 4	230 ± 16	1 ± 1	2 ± 1	2 ± 0	4 ± 1

Table 4

Chemical compounds detected by HPLC and GCMS from Soxhlet extraction of three -layered films.

	P168 HPLC	P168-ox HPLC	AO1010 HPLC	AO1076 HPLC	Oligomers GCMS	Arvin4 GCMS	Arvin8 GCMS	DEHP GCMS	DEHT GCMS
LDPE	nd*	nd	nd	nd	nd	nd	nd	nd	nd
LLDPE	d*	d	d	nd	d	nd	nd	nd	nd
LDPE/R1	nd	d	nd	d	d	nd	d	d	d
LDPE/R2	nd	d	d	d	d	nd	d	d	d
LLDPE/R1	nd	d	nd	d	d	d	d	d	d
LLDPE/R2	d	d	d	d	d	d	d	d	d
R1	nd	d	nd	nd	d	nd	nd	d	d
R2	d	d	nd	d	d	nd	nd	d	d

*nd-not detected.

d*-detected/confirmed.

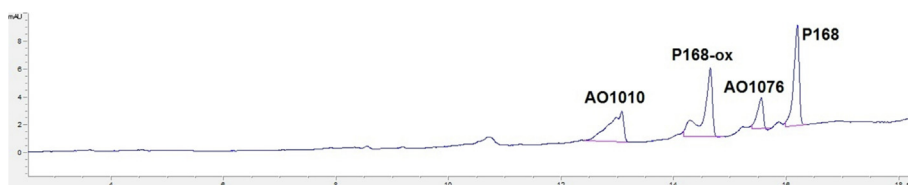


Fig. 1. HPLC of selected sample (LLDPE/R2) from Soxhlet extraction.

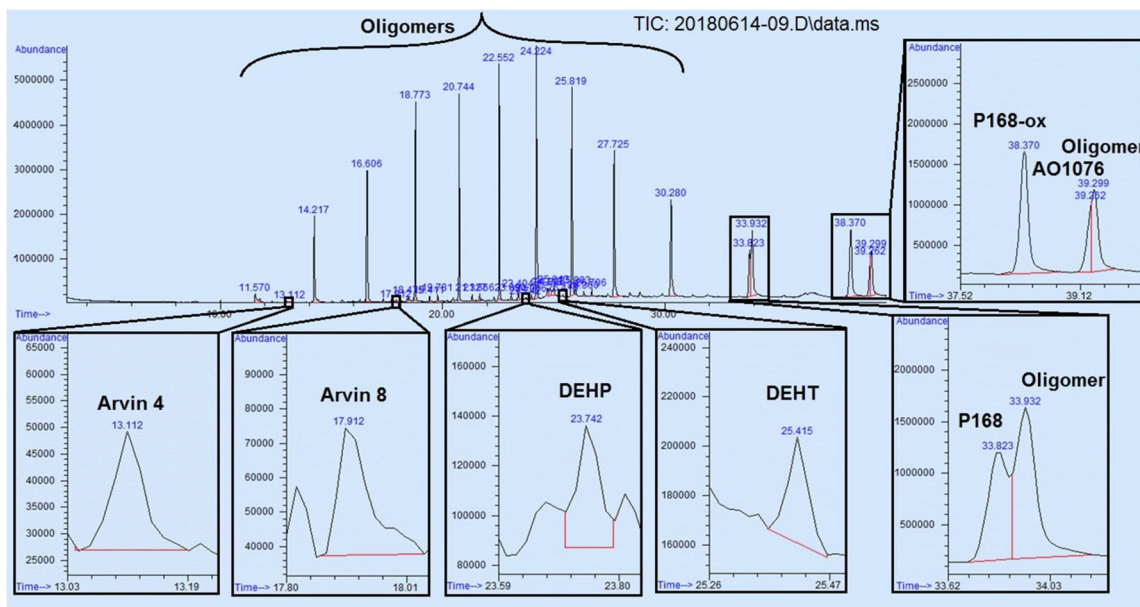


Fig. 2. GC-MS of selected sample (LLDPE/R2) from Soxhlet extraction.

at highest temperature implying that all values for overall migration were well below the overall migration limit of 10 mg/dm²

For statistical analysis of the OM, a GLM with interactions was used. Two effects were considered: Material and temperature (main effect plots are presented in Fig. 3a) and b)). Lowest value of overall migration was recorded for LDPE/R2 and R2 sample (under

detection limit). Recycled sample R2 had less influence on OM values compared to R1. Migration from LDPE compared with R2 was slightly higher at 20 °C and at lower temperatures influenced by migration from LDPE. At higher temperatures, more distinguished difference has been measured implying that influence of temperature is more pronounced for the recycled sample then for

Table 5

Overall migration (mg/dm²) for three-layered samples at 20, 40 and 60 °C after 10 days exposure to 95% ethanol; given as average \pm SD (n = 3).

Sample	Overall migration (mg/dm ²)		
	20 °C	40 °C	60 °C
LDPE	<2 ^B	<2 ^B	<2 ^D
LDPE/R1	<2 ^B	2.7 ^{AB} ±0.1	4.1 ^B ± 0.1
LDPE/R2	<2 ^B	<2 ^B	2.4 ^D ±0.3
LLDPE	<2 ^B	3.1 ^{AB} ±0.8	3.2 ^C ± 0.3
LLDPE/R1	<2 ^B	3.0 ^{AB} ±0.4	4.5 ^B ± 0.2
LLDPE/R2	<2 ^B	<2 ^B	3.1 ^C ±0.3
R1	2.1 ^A ± 0.3	3.9 ^A ± 0.8	5.6 ^A ± 0.1
R2	<2 ^B	2.0 ^{AB} ±1.0	3.1 ^C ±0.3

*Means that do not share a same letter within the same column are significantly different.

the virgin one. Migration from LLDPE/R2 is mainly related to migration from LLDPE as values were on the lower level compared to both R2 and LLDPE/R2 at 20 °C. At 40 °C LLDPE showed higher migration values then both LLDPE/R2 and R2 while at 60 °C these values were almost at the same level.

OM value for R1 were two times higher than from recycled sample R2.

3.2. Specific migration

Specific migration limit (SML) is defined in Commission Regulation EC 10/2011 as maximum permitted amount of a given substance released from a material or article into food or food simulants (Commission, 2011); In Annex I of the same regulation, the positive list of approved substances for food contact materials (FCM) is given and additives commonly used in plastic materials and articles are present on this list. Targeted chemical compounds were determined from the microextraction of pellets and Soxhlet extraction of films. Specific migration results are presented in Table 6 and Fig. 4. If there is no specific limit defined in this list or other restriction are provided in Annex I, a generic specific migration limit of 60 mg/kg shall be applied. AO1076 is set up in the list with SML = 6 mg/kg. For the other two additives generic SML will be used. Specific migration values of additive AO1076 analyzed by both HPLC and GC-MS techniques were below the approved SML (mg/kg). Highest detected value of 0.56 \pm 0.05 mg/kg was measured at 60 °C for the LLDPE/R2 sample, thus ten times lower than actual SML for AO1076 defined in Annex I. Highest measured value of additive P168 was 4.92 \pm 0.11 mg/kg for the LLDPE sample

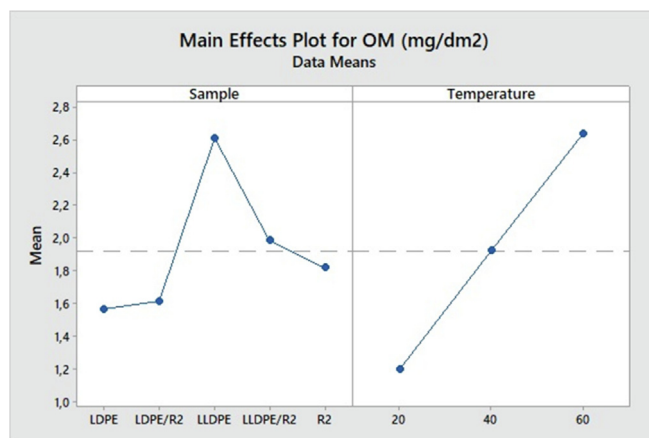


Fig. 3b. Main effect plot for recycled sample R2.

at 60 °C, while highest value of AO1010 of 0.73 \pm 0.05 mg/kg was measured for the LLDPE sample at 40 °C. Specific migration values of these chemical compounds are significantly below SML (60 mg/kg), implying that specific migration values of 3-layered structures designed with the selected recycled core layer are in accordance with regulation (Commission, 2011).

Nevertheless, it is important to consider all NIAS that are detected and quantified in this research (P168-ox, Arvin substances, phthalates and oligomers). P168-ox is an oxidation product of the antioxidant P168 commonly used in LLDPE. It was detected in LLDPE material as well as in R1 and R2. As expected, P168-ox was not found in the LDPE film, whereas a considerable amount was found in the LLDPE film (e.g 5.8 mg/kg at 40 °C). This is in accordance with the results from the microextraction. P168-ox was found in all the films with recycled mid-layer. In the LDPE films, the levels were in the same range as in the pure recycled films, R1 and R2. In the LLDPE films, the levels were higher, due to that both the mid-layers and the outer layers contain P168-ox. Arvin substance 4 and 8 (found both in R1 and R2 by micro extraction of pellets) were detected and quantified in the specific migration analysis (Table 6). Reaction or degradation products are generated during processing of polymers at high temperatures. Forming of these chemical compounds depends on processing, handling and storage conditions (Beldi et al., 2012; Biedermann-Brem et al., 2012). Organic chemical compounds known as Arvin substances have been detected for the first time in polyethylene pipes by Arvin et al. (Brocca et al., 2002). The Threshold of Toxicological Concern (TTC) approach has been developed to qualitatively assess the risk of low-level substances in the diet (Committee, 2012; Roberts et al., 2015). The Cramer classification scheme (decision tree) is the best-known approach to estimate TTC for a chemical substance based on its chemical structure (Roberts et al., 2015). There are three Cramer classes with Class III representing the most severe toxic hazard. According to TTC and Cramer classification Arvin 4 (2,4-di-tert-Butylphenol, CAS 96-76-4) has a Cramer Class I, giving it an SML of 1.8 mg/kg and Arvin 8 (7,9-Di-tert-butyl-1-oxaspiro (4,5)deca-6,9-diene-2,8-dione, CAS 82304-66-3) has a Cramer Class III, giving it an SML of 0.09 mg/kg. Arvin 4 has been detected at 40 °C and 60 °C for LLDPE samples (0.02 \pm 0.00 mg/kg and 0.01 \pm 0.01 mg/kg respectively) and LLDPE/R2 (0.01 \pm 0.00 mg/kg) while highest value of Arvin 8 was detected for sample LLDPE/R1 at 60 °C (0.05 \pm 0.02 mg/kg) and for sample LDPE/R1 and LLDPE/R1 at 40 °C (0.03 \pm 0.02 mg/kg). These results are implying that all levels of detected Arvin substances are below SML defined in Cramer classification. The analyzed concentration of DEHP were below the SML

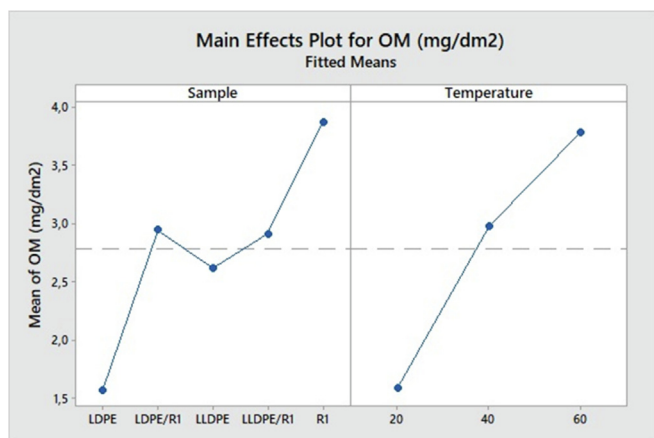


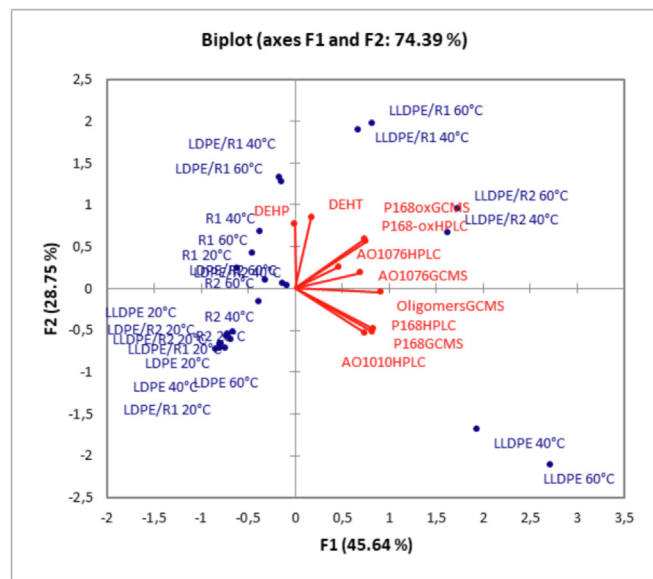
Fig. 3a. Main effect plot for recycled sample R1.

Table 6Specific migration (mg/dm²) for three-layered samples at 20, 40 and 60 °C after 10 days exposure to 95% ethanol; given as average \pm SD (n = 3).

Sample	Specific migration at 20 °C (mg/kg)									
	P168 HPLC	P168-ox HPLC	AO1076 HPLC	AO1010 HPLC	DEHP GCMS	DEHT GCMS	Oligomers GCMS	Arvin 4 GCMS	Arvin 8 GCMS	
LDPE	Nd*	nd	nd	nd	nd	nd	nd	nd	nd	
LDPE/R1	0.01 ^C \pm 0.02	0.69 ^C \pm 0.02	nd	nd	nd	nd	nd	nd	nd	
LDPE/R2	nd	0.71 ^C \pm 0.02	0.01 ^A \pm 0.02	nd	nd	nd	nd	nd	nd	
LLDPE	0.16 ^A \pm 0.07	2.13 ^A \pm 0.24	nd	0.01 ^A \pm 0.02	nd	nd	nd	nd	nd	
LLDPE/R1	nd	2.22 ^A \pm 0.40	nd	nd	nd	nd	nd	nd	nd	
LLDPE/R2	0.04 ^{BC} \pm 0.01	2.56 ^A \pm 0.20	0.04 ^A \pm 0.02	nd	nd	nd	nd	nd	0.01 ^A \pm 0.00	
R1	0.09 ^{AB} \pm 0.00	1.35 ^B \pm 0.17	nd	0.03 ^A \pm 0.00	0.08 ^A \pm 0.00	0.01 ^A \pm 0.00	0.180 ^A \pm 0.02	nd	nd	
R2	0.06 ^{BC} \pm 0.00	1.14 ^{BC} \pm 0.08	nd	nd	nd	nd	0.02 ^B \pm 0.01	nd	nd	
Sample	Specific migration at 40 °C (mg/kg)									
	P168 HPLC	P168-ox HPLC	AO1076 HPLC	AO1010 HPLC	DEHP GCMS	DEHT GCMS	Oligomers GCMS	Arvin 4 GCMS	Arvin 8 GCMS	
LDPE	nd	nd	nd	nd	nd	nd	0.102 ^{BC} \pm 0.005	nd	nd	
LDPE/R1	0.09 ^C \pm 0.01	3.87 ^D \pm 0.52	0.04 ^{AB} \pm 0.07	nd	0.13 ^A \pm 0.02	0.03 ^{AB} \pm 0.01	0.378 ^{BC} \pm 0.030	nd	0.03 ^A \pm 0.00	
LDPE/R2	0.04 ^C \pm 0.04	3.94 ^D \pm 0.09	0.11 ^{AB} \pm 0.11	nd	0.01 ^C \pm 0.00	nd	0.219 ^{BC} \pm 0.027	nd	0.01 ^B \pm 0.00	
LLDPE	3.31 ^A \pm 0.41	5.75 ^C \pm 0.47	nd	0.73 ^A \pm 0.05	nd	nd	0.948 ^A \pm 0.659	0.02 ^A \pm 0.00	nd	
LLDPE/R1	0.07 ^C \pm 0.01	9.08 ^A \pm 0.22	nd	nd	0.08 ^B \pm 0.01	0.04 ^A \pm 0.01	0.770 ^{AB} \pm 0.232	nd	0.03 ^A \pm 0.02	
LLDPE/R2	1.40 ^B \pm 0.08	7.55 ^B \pm 0.33	0.43 ^A \pm 0.38	0.24 ^B \pm 0.04	0.02 ^C \pm 0.00	0.01 ^{CD} \pm 0.00	0.573 ^{BC} \pm 0.049	0.01 ^B \pm 0.00	0.01 ^B \pm 0.00	
R1	0.24 ^C \pm 0.02	3.30 ^D \pm 0.49	nd	0.03 ^C \pm 0.00	0.08 ^B \pm 0.03	0.02 ^{BC} \pm 0.01	0.037 ^C \pm 0.013	nd	0.01 ^B \pm 0.00	
R2	0.29 ^C \pm 0.00	3.57 ^D \pm 0.39	0.05 ^{AB} \pm 0.01	nd	0.01 ^C \pm 0.00	nd	0.012 ^C \pm 0.001	nd	nd	
Sample	Specific migration at 60 °C (mg/kg)									
	P168 HPLC	P168-ox HPLC	AO1076 HPLC	AO1010 HPLC	DEHP GCMS	DEHT GCMS	Oligomers GCMS	Arvin 4 GCMS	Arvin 8 GCMS	
LDPE	nd	nd	nd	nd	nd	nd	1.58 ^{CD} \pm 0.23	nd	nd	
LDPE/R1	0.11 ^C \pm 0.04	3.82 ^B \pm 0.44	nd	nd	0.12 ^A \pm 0.00	0.04 ^A \pm 0.00	2.38 ^C \pm 0.24	nd	0.01 ^B \pm 0.00	
LDPE/R2	0.09 ^C \pm 0.01	3.57 ^B \pm 0.09	0.18 ^B \pm 0.07	nd	0.01 ^{BC} \pm 0.00	0.01 ^{BC} \pm 0.00	1.50 ^{CD} \pm 0.15	nd	0.01 ^B \pm 0.00	
LLDPE	4.94 ^A \pm 0.11	4.38 ^B \pm 0.51	nd	0.53 ^A \pm 0.16	nd	nd	13.08 ^A \pm 1.72	0.01 ^A \pm 0.00	nd	
LLDPE/R1	0.11 ^C \pm 0.04	9.95 ^A \pm 0.18	nd	nd	0.07 ^{AB} \pm 0.01	0.05 ^A \pm 0.00	6.27 ^B \pm 0.45	nd	0.05 ^A \pm 0.00	
LLDPE/R2	0.90 ^B \pm 0.05	8.76 ^A \pm 1.96	0.56 ^A \pm 0.05	0.07 ^B \pm 0.03	0.02 ^C \pm 0.00	0.01 ^{BC} \pm 0.00	5.60 ^B \pm 1.11	0.01 ^A \pm 0.00	0.01 ^B \pm 0.00	
R1	0.18 ^C \pm 0.15	3.68 ^B \pm 0.44	nd	0.03 ^B \pm 0.00	0.07 ^{AB} \pm 0.05	0.02 ^B \pm 0.01	0.21 ^D \pm 0.15	nd	0.01 ^B \pm 0.00	
R2	0.26 ^C \pm 0.00	3.85 ^B \pm 0.217	0.21 ^B \pm 0.01	nd	0.02 ^B \pm 0.01	0.01 ^{BC} \pm 0.00	0.28 ^{CD} \pm 0.05	nd	nd	

**Nd means that is not detected or value below 0.01 mg/kg.

*Means that do not share a same letter within the same column are significantly different.

**Fig. 4.** PCA plot: specific migration values from HPLC and GC-MS for 3-layered films.

1.5 mg/kg for this chemical compound in all film samples. The DEHT concentration found was also at a low level (below 0.05 mg/kg) and the SML value stated in Annex I is 60 mg/kg. Neither DEHP nor DEHT are expected to be present in polyolefins recycling streams. These components are usually used as plasticizers in polyvinyl chloride (PVC) production as well as in adhesives and printing inks. Their presence is thus related probably to printing

inks, adhesives, labels etc. (Geueke et al., 2018; Pivnenko et al., 2016). The two phthalates come from the recycled mid-layer, considering that it was not detected in the virgin materials by micro extraction.

Migration of oligomers were not detected at 20 °C, except for samples R1 and R2 (0.180 and 0.02 mg/kg respectively). At 40 °C migration was detected for all samples with highest level for LLDPE sample (0.948 mg/kg). However, at 60 °C these values increased more, with highest level for LLDPE sample (13.08 mg/kg). Oligomers are defined as molecules consisting of a few monomers repeating units (from 2 up to 40). Generally, oligomers with molecular weight below 1000 Da are considered as potential migrants as smaller molecules are more prone to diffusion compared to bigger ones (Hoppe et al., 2016). It is well known that oligomers are always present in polymeric structures, however, they have gained more attention as possible migrants especially in food packaging application (Hoppe et al., 2016). Oligomers are not listed in the Annex I of the regulation 10/2011/EU and thus are treated as NIAS (Commission, 2011).

In food legislation oligomers are not treated separately as chemical compounds, however according to the article 3 of the EU framework Regulation no. 1935/2004 they must be risk assessed as occurring in plastics (Regulation, 2004). Nelson et al., 2011 presented that only a limited number of toxicity studies on oligomeric materials were found in both the publicly-available databases and internal FDA files (Nelson et al., 2011). This group has reviewed the available data and concluded that among 11 oligomeric substances with toxicity data in internal FDA files, 17 genetic toxicity tests were performed and were all negative (Nelson et al., 2011). Risk assessment for oligomers is still set through overall migration limit as 10 mg/dm² or 60 mg/kg. Highest measured value of specific

migration of oligomers in our research was 13.08 mg/kg for LLDPE at 60 °C (this value is still well below SML of 60 mg/kg). It is also found that migration of oligomers was highly dependent on temperature. In this case influence of harsh conditions probably caused dissolving of polymer at 60 °C and thus more oligomers from migration results. Oligomers are not very clearly classified as chemical compounds migration from food contact materials. Their presence is related to processing of polymeric materials and thus should be considered as polymer specific substances rather than NIAS. Still, the risk appears also if recycling process is considered as oligomers can also appear because of thermal processes or hydrolysis in recycling process (Hoppe et al., 2016; Nelson et al., 2011).

PCA biplot for SM results are presented in Fig. 4. The two first principal components explained 74.39% of the total variability in the data. Several groups can be observed on the PCA plot.

Most of the evaluated parameters are in good correlation with PC1, while DEHT and DEHP span PC2. Group formation is detected for all samples at 20 °C. LDPE sample with R1 and R2 recycled mid-layer at 40 and 60 °C formed one group and showed similar behavior. Most distinguished sample was LLDPE at 40 and 60 °C.

Increase in migration levels for all measured samples is related to increase in temperature. However, sample type is also influencing SM levels. Highest influence on migration values were from samples containing LLDPE and recycled R1 and R2 (LLDPE, LLDPE/R1 and LLDPE/R2) at 40 and 60 °C (Fig. 4). This is partly influenced by the LLDPE composition (additives that are present regularly in LLDPE sample), but also related to influence of migration levels from recycled R1 and R2 as mid layer. According to the PCA biplot diagram (Fig. 4) it can be noted that both DEHP and DEHT migration is related to migration from LLDPE/R1 and LDPE/R1 at 40 and 60 °C, antioxidants AO1076 are mainly migrating from LLDPE structures, while AO1010 and P168 are migrating from LLDPE virgin material at 40 and 60 °C. Nevertheless, all samples showed significantly lower specific migration values than stated in Annex I of legislation for IAS. If we consider 10 mg/dm² for NIAS too, presented SM values are on much lower levels than stated in legislation documents (Commission, 2011).

Results from both overall and specific migration in this study are implying that by diluting a plastic packaging film with another material that contain less molecular contaminants less of these contaminants are present in the overall film structure and less will migrate to the food it contains.

3.3. Optical properties

Microscopy cross-section, optical measurements and visual tests were performed to illustrate the influence of recycled mid layer on appearance of 3-layered structures. Pictures of the cross-sections are presented in Fig. 5.

Films containing recycled mid-layer have a certain amount of impurities (visible as particles and gels in microscopy cross section) compared to virgin samples (Fig. 5-samples R1 and R2). This effects of recycled R1 are visible on the film samples (Fig. 6a) and b) as opaque sandy-brownish colour, implying that the recycled mid-layer with R1 causes significant changes in visual color in both reference materials. Recycled material R2 had less visible influence on the visual color and cross-section compared to R1, however, changes are still visible as bubbles and impurities on cross section as well as visual changes on the samples. Considering that outer layers of virgin reference materials were 20 µm and transparent it can be expected to visually recognize impurities from the recycled core layer.

Visual colour changes caused by incorporation of two different recycled polymers and virgin LDPE and LLDPE has been measured and presented in Tables 7 and 8.

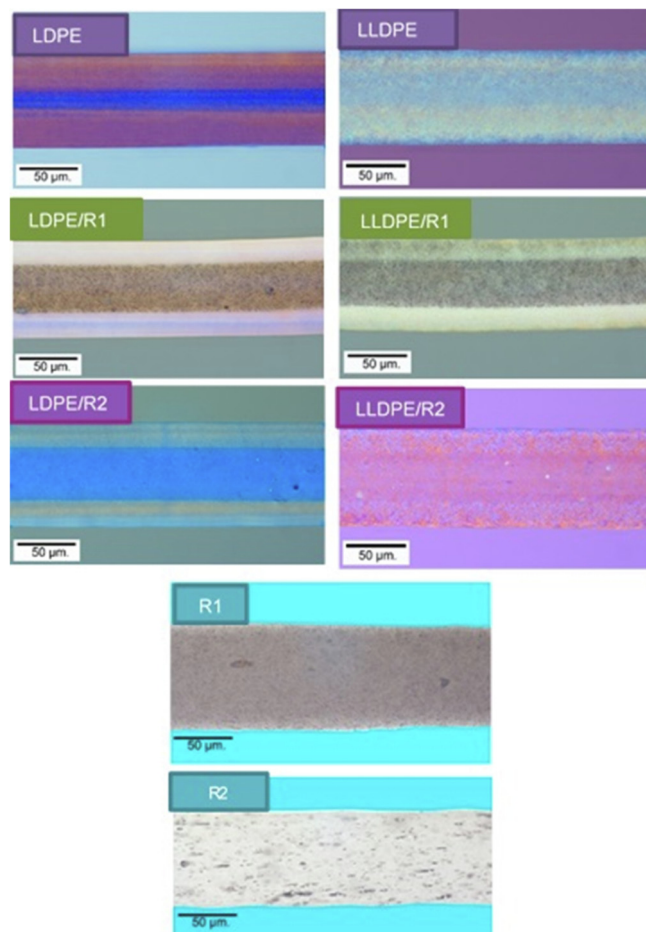


Fig. 5. Microscopy cross-section of 3-layered films.

L* values of virgin reference LDPE compared to multilayered structure containing recycled polymer R1 shows higher L* values, indicating darker sample. For sample containing recycled R2 the L* values were measured to be almost on same level as the reference (Table 7). This is because L* for R1 is almost 12 units lower than LDPE while R2 is less than 1 unit lower. According to calculated ΔE^*_{ab} value an extremely marked difference between LDPE and LDPE/R1 (9.08) was detected, while if compared with recycled R1 it is defined as the color of a different shade (17.68). For samples LDPE/R2 and recycled R2 it is described as sample with a noticeable difference (1.92 and 2.69 respectively).

An extremely marked difference between LLDPE reference and multilayered structure containing R1 (10.27) was detected, while the addition of R2 acted in slight difference between samples (1.29).

Changes in color, both measured and visually, are causing changes in the materials appearance. This doesn't have to be a drawback; however, it can limit the applications in where the recycled materials may be used. Particles and gels that are visible on the microscopy cross section are causing these changes, influencing not just visual effect, but also materials properties.

4. Conclusion

Microextraction of polymer pellets and Soxhlet extraction of three-layer films were carried out and allowed the detection and quantification of chemical substances migrating from designed 3-layered structures.

Overall migration data reported in this research are implying



Fig. 6a. 3-layered film samples: LDPE, LDPE/R1, LDPE/R2, R1 and R2.



Fig. 6b. 3-layered film samples: LLDPE, LLDPE/R1, LLDPE/R2, R1 and R2.

that 3-layered films (LDPE and LLDPE) with recycled mid-layer of all samples were below OML (10 mg/dm^2). The specific migration results point out that both IAS and NIAS are present in the investigated samples. However, most of NIAS, except oligomers, originated from the recycled mid-layer (as they were not detected in reference material). Oligomers are present as migrants (NIAS) and were found in higher amount for LLDPE virgin materials than for samples with recycled mid layer, suggesting that they originate mainly from the LLDPE layer (reference/virgin materials).

The highest migration values from overall and specific migration were observed in LLDPE/R1 materials while the lowest values were for LDPE/R2 materials, hence this was the best solution for the use of recycled mid layer. However, both OM and SM values were below limits stated in legislation, pointing out their possible use as food contact materials.

Visual color as well as optical properties of the films were evaluated, as these properties may be decisive when choosing the adequate packaging material. Samples with R1 mid layer had less transparency and visually appeared more sandy and brownish compared to R2 which stayed transparent and showed only slight changes in visual color.

This scientific research presents a unique data set as a starting point in approval of use of flexible recycled PE films in food contact application.

According to the presented data further research will be focused on more use of recycled materials through their design and suitability for food packaging application. Nevertheless, focus on recycling processes and possibilities of their approval by regulatory

Table 7

Color indexes ($L^*a^*b^*$) of three-layered films of: LDPE, LDPE/R1, LDPE/R2, R1 and R2: given as average \pm SD (n = 5).

	$L^*(D65)$	$a^*(D65)$	$b^*(D65)$	ΔE^*_{ab}
LDPE	$93.97^A \pm 0.08$	$-1.87^C \pm 0.04$	$2.00^E \pm 0.08$	–
LDPE/R1	$88.64^C \pm 0.16$	$-0.05^B \pm 0.03$	$9.12^B \pm 0.10$	9.08
LDPE/R2	$93.39^B \pm 0.12$	$-0.26^C \pm 0.04$	$2.89^D \pm 0.14$	1.92
R1	$81.09^D \pm 0.35$	$1.15^A \pm 0.05$	$14.09^A \pm 0.09$	17.68
R2	$93.12^B \pm 0.05$	$-0.25^C \pm 0.02$	$3.97^C \pm 0.16$	2.688

*Means that do not share a same letter within the same column are significantly different.

Table 8

Color indexes ($L^*a^*b^*$) of three-layered films of: LLDPE, LLDPE/R1, LLDPE/R2, R1 and R2: given as average \pm SD (n = 5).

	$L^*(D65)$	$a^*(D65)$	$b^*(D65)$	ΔE^*_{ab}
LLDPE	$95.30^A \pm 0.06$	$-0.15^C \pm 0.01$	$2.20^E \pm 0.03$	–
LLDPE/R1	$88.04^D \pm 0.25$	$0.10^B \pm 0.02$	$9.46^B \pm 0.11$	10.27
LLDPE/R2	$94.58^B \pm 0.036$	$-0.23^D \pm 0.01$	$3.26^D \pm 0.04$	1.29
R1	$81.09^E \pm 0.35$	$1.15^A \pm 0.05$	$14.09^A \pm 0.09$	18.56
R2	$93.12^C \pm 0.05$	$-0.25^D \pm 0.02$	$3.97^C \pm 0.16$	2.84

*Means that do not share a same letter within the same column are significantly different.

authorities (EFSA and EC) should be conducted. Influence of recycled materials on visual properties of prepared films is still a challenge and should be in a focus of the future research. Already,

these data are giving valuable insight into the possibilities of recycled materials application, reduction of flexible plastic waste, thus having a strong impact on sustainability and new sustainable goals for the future.

CRedit authorship contribution statement

Tanja Radusin: Writing - original draft, Investigation, Formal analysis, Validation. **Jorunn Nilsen:** Conceptualization, Methodology, Writing - review & editing. **Sondre Larsen:** Investigation, Formal analysis. **Steffen Annfinn:** Investigation, Data curation. **Charlotte Waag:** Investigation, Data curation. **Marianne Sørflaten Eikeland:** Supervision. **Marit Kvalvåg Pettersen:** Writing - review & editing, Formal analysis, Supervision. **Siw Bodil Fredriksen:** Conceptualization, Supervision.

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