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Jinyang Guo, Youngju Kim, Zhi Kai Chong, Ayah Alassali, Jose Pablo Chacon, Dieter Gottschalk, Magdalena Kitzberger, Kerstin Kuchta

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Low-density polyethylene is the most applied packaging plastic. The recycling rate of LDPE is low, especially for the material from the post-consumer source. The quality of the LDPE post-consumer recyclates is influenced by different contaminations, either intentionally or non-intentionally. Printing ink is one of the substances that is regarded as having a negative influence on the quality of LDPE recyclates. This study used a new type of nonionic surfactant oxirane, methyl-, polymer for de-inking as a pretreatment. Afterward, the LDPE film flakes were mechanically recycled in the laboratory. A melt flow index (MFI), differential scanning calorimetry (DSC), and tensile strength analysis were carried out. This work reported the de-inking effect of a new type of nonionic surfactant. However, no evidential correlation between the rate of color removal and the improvement in the rheological, thermal, and mechanical properties can be identified from the result of this study.

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Article

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Jinyang Guo [1,*], Youngju Kim [1], Zhi Kai Chong [1], Ayah Alassali [1], Jose Pablo Chacon [1], Dieter Gottschalk [2], Magdalena Kitzberger [1] and Kerstin Kuchta [1]

[1] Circular Resource Engineering and Management (CREM), Hamburg University of Technology (TUHH), 21079 Hamburg, Germany
[2] Wildplastic GmbH, 20457 Hamburg, Germany
* Correspondence: jy.guo@tuhh.de

Abstract: Low-density polyethylene is the most applied packaging plastic. The recycling rate of LDPE is low, especially for the material from the post-consumer source. The quality of the LDPE post-consumer recyclates is influenced by different contaminations, either intentionally or non-intentionally. Printing ink is one of the substances that is regarded as having a negative influence on the quality of LDPE recyclates. This study used a new type of nonionic surfactant oxirane, methyl-, polymer for de-inking as a pretreatment. Afterward, the LDPE film flakes were mechanically recycled in the laboratory. A melt flow index (MFI), differential scanning calorimetry (DSC), and tensile strength analysis were carried out. This work reported the de-inking effect of a new type of nonionic surfactant. However, no evidential correlation between the rate of color removal and the improvement in the rheological, thermal, and mechanical properties can be identified from the result of this study.

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1. Introduction

Flexible plastics such as films and bags are widely used because of their lightness, flexibility and protective properties. LDPE (low-density polyethylene) is the most commonly used polymer for producing flexible films and bags for various packaging, building materials, and agriculture applications [1,2]. The total demand for LDPE in the EU was 8.85 Mt in 2020. LDPE is the most consumed plastic for packaging applications [3]. However, the post-consumer recycling of LDPE still poses a problem leading to downcycling, incineration, being landfilled or even leaking into the environment [4,5]. In the EU, approximately 9.6% of LDPE was recycled into secondary material in 2017, and less than 20.0% of the market demand for LDPE LDPE recyclate was met in 2018 [6].

Plastic packaging waste, including film, can be separately collected via the source segregation system or extracted from municipal solid waste (MSW) [7,8]. From the separately collected or extracted plastic waste stream, plastic film can be sorted out according to its physical property by a wind shifter and ballistic separator [9–12]. LDPE can also be optically identified and sorted by Fourier Transform Near Infrared Spectroscopy (FT-NIR) [13–15]. Moreover, with recent advancements, some limitations of the NIR-sensor-based sorting, such as multilayer construction and differentiating between LDPE and HDPE, can be overcome [16–20].

After the sorting process, the LDPE enters the mechanical recycling process. First, t films go undergo shredding, density separation, washing (optional case hot washing with 1–2 wt.% NaOH) and drying before the extrusion process [21,22].

Similar to other types of plastics, the recyclate quality of LDPE is limited by degradation and contamination [23]. The degradation of LDPE is caused by heat, mechanical, UV, and chemical stresses in the life cycle of the polymer from production to mechanical recycling. The contamination can be from intentionally added substances (IAS) and non-intentionally added substances (NIAS). The IAS can be further categorized as additives, processing agents, printing ink, and adhesion (i.e., solvent-based polyurethane (PU) in multilayer packaging) [24]. NIAS includes the degradation of products of IAS, impurities from IAS, side reaction products, and contaminations from recycling processes. The analysis of impurities from IAS and side reaction products are reported to be hard to measure [25]. Horodytska et al., 2020, analyzed 134 different substances detected in post-consumer HDPE and LDPE samples. More than 50% of the substances in recycled LDPE were reported to be contamination from the recycling process, typically the from the packaging content [26]. A brief overview of different categories of contamination is shown in Table 1.

Table 1. A brief overview of intentionally and non-intentionally added substances in recycled plastic.

Category	Origin	Source
Intentionally added substances (IAS)	Additives	[27–30]
	Processing agents	[26,31]
	Printing ink	[32,33]
	Adhesion	[24,26]
Non-intentionally added substances (NIAS)	Degradation of products of IAS	[25,26]
	Impurities from IAS	[25,31]
	Side reaction products	[25]
	Contamination from recycling processes	[32]

This study aims to investigate the de-inking effect of a nonionic surfactant (oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol) that has not been covered by previous studies. Secondly, this study also aims to establish a method to measure the rate of color removal for inhomogeneous, post-consumer plastic waste. Lastly, additional quality parameters, including the melt flow index (MFI), crystallinity and mechanical property of the recycled plastic after the surfactant washing pretreatment, are investigated.

2. Backgrounds

2.1. Cleaning with Surfactants as Pretreatment Process

To improve the quality of the recyclates, the contamination must be reduced. If contaminative substances have already entered the macromolecular matrix of a polymer, the options for removal in the mechanical recycling process are (1) extrusion degassing [34]; (2) melt filtration; and (3) solid phase extraction. However, different researchers have reported the limitation in the efficiencies of these decontamination technologies. Alshahrani et al. reported that odor was decreased by 37% after three-stage degassing [35]. The efficiency of melt filtration in contaminant removal is not well studied. Nevertheless, mineral and polymeric contaminations were detected by ATR-FTIR imaging from an industrially produced recyclate with melt filtration [36]. The solid phase extraction process is currently limited to PET [26].

Therefore, decontamination and quality upgrades in mechanical recycling should be considered an integrated approach in the pretreatment, extrusion, and post-extrusion stages [37]. The upstream cleaning process before extrusion is one of the crucial steps in reducing contamination (odor and printing ink) [38]. However, washing with hot water at 60 °C is limited to odor removal only for LDPE [39]. Washing the plastic flakes with 1–2 wt.% caustic soda (NaOH) at a high temperature (70–80 °C) can partly remove the printing ink [21,38,40,41].

Even for advanced recycling technologies such as dissolution recycling processes and pyrolysis, effective washing is still a necessary pretreatment. For instance, the study by Genuino et al. reported that effective washing could remove biogenic, inorganic and halogen content as a pretreatment for chemical recycling with pyrolysis [42].

Cleaning plastic film with nonionic surfactants, including ethoxylated nonylphenol, ethoxylated alcohol and ethoxylated amine, was reported as an effective method with a pH over 10 and heating under 55 °C [40]. A series of studies were conducted to test different surfactants (anionic, cationic, nonionic, and zwitterionic) on water-based and solvent-based ink systems with different colors. These researchers reported that water-based ink could be dissolved without surfactant at a high pH value (11–13) [40]. Quandary ammonium cation presents the best de-inking performance, especially cetrimonium bromide (CTAB) [41]. This research series also proved that de-inking could improve the mechanical property of the recyclates [43]. Based on these results, Chiptong et al. compared the de-inking effect of different quandary ammonium cations and reported that a longer hydrophobic chain favors the de-inking effect [44]. Ye et al. researched converting used cooking oil to produce surfactants for de-inking [45]. The produced de-inking agent from the waste cooking oil was not reported to have a counter-ion. Therefore, it might be extrapolated that a nonionic surfactant was produced.

The patented de-inking process of Cadel from Spain uses surfactants, including cetrimonium bromide (CTAB), dodecyl trimethylammonium bromide (DTAB), and dodecyl sulfate (SDS) [46]. Another de-inking process was developed by the German printing ink producer Siegwerk, focusing only on UV-cured ink [47].

2.2. Printing Ink Compositions

The surface energy of plastics is low, which is difficult for adhesion on the surface. For example, E film has a surface energy of 22–32 mN/m without any treatment [48–50]. Therefore, a surface treatment is required as the pretreatment step before printing in order to break the crystallized structure on the surface and enable the adhesion of the printing ink [51,52]. Plastic films can be printed by offset or flexography [53]. The printing ink can be categorized into water-based, solvent-based, and UV-cured printing inks [54]. The printing ink is composed of binder, pigment, carrier and additives [55]. The carrier (water or organic solvent) evaporates during the drying process. The pigment delivers the desired color of the printing. The binder provides almost all the mechanical characteristics of the printed area, including strength, flexibility, and mechanical and chemical stability. A list of different binders is shown in Table 2, from [53,55–57].

Table 2. Binder chemicals for water-based and solvent-based ink systems.

Water-Based Ink System	Solvent-Based Ink System
Methyl methacrylate	nitrocellulose (NC)
Acrylates, i.e., styrene, butyl acrylates	polyamide (PA)
Acrylic and methacrylic acid	polyvinyl butyral (PVB)
methacrylates	polyvinyl chloride (PVC)
acrylic acid	polyurethane (PU)

As shown in Table 2, the binder resins of the printing inks are from different polymers than the plastic packaging. Hence, it is reasonable to consider the printed area of the plastic packaging as a multilayer structure. The technologies for separating a multilayer structure were reported with different methods [58–62]. For example, in Yousef et al. and Mumladze et al., ultrasonic washing was employed as a pretreatment [59,60]. As ultrasonic washing is generally applied to a high-efficiency cleaning process, it might also help remove the printing ink from the plastic surface.

The adhesion mechanism between the binders and the polymer surface varies between different types of binder materials, which are often combined in the printing ink [54,63]. As a result, multiple common binder materials could be present in the post-consumer LDPE stream, which poses difficulties in designing the cleaning process. Therefore, the ink system in post-consumer flexible packaging is considered as a black box.

3. Materials and Methods

3.1. Materials

LDPE film samples were collected from post-consumer sources. First, partially printed transparent and monolayer materials were selected. Then, the samples were cut into 20 × 20 mm pieces for cleaning (Figure 1). The non-printed pieces were separated by hand from the printed pieces during the sample preparation. The nonionic surfactant oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol (CAS No. 204336-40-3, commercial name: TEGO Surten W111) and a dispersant DA850 were acquired by Evonik Operations GmbH. The cationic surfactant cetrimonium bromide (CTAB CAS No. 57-09-0) was purchased from VWR International GmbH as a reference. The chemical structures of the two surfactants used in this study are shown in Figure 2.

Figure 1. Shredded LDPE samples for the washing experiment.

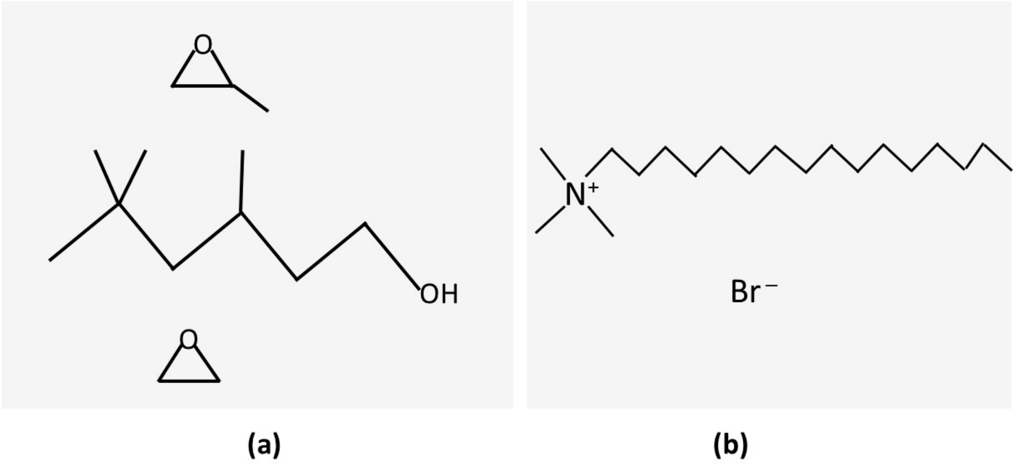

Figure 2. Chemical structures of the surfactants used in this study. (**a**) Oxirane, methyl-, polymer with oxirane, mono(3,5,5-trimethylhexyl) ether. (**b**) Cetrimonium bromide. Source: [64,65].

3.2. Cleaning Experiment Setup

The cleaning experiment was carried out in two stages. In the first stage, 5 g of the samples were washed in a 500 mL Erlenmeyer flask to select the most suitable surfactant. In the second stage, based on the result from the first stage, 100 g of the samples were cleaned to evaluate the impact of different washing agents on the quality of the recyclates. The cleaning process is shown in Figure 3.

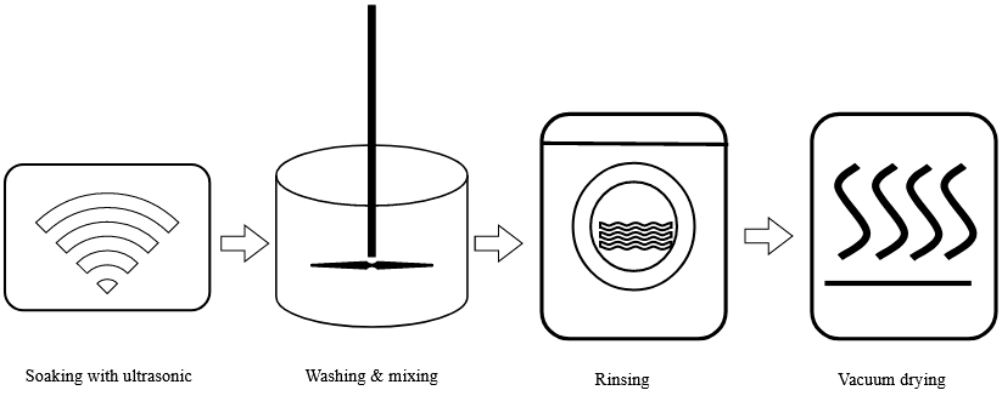

Figure 3. The cleaning process.

3.2.1. Small-Scale Experiment

The cleaning experiment follows the methods reported in the studies of Gecol and colleagues [40,41,43]. A quantity of 5 g of printed film samples was randomly selected for the washing test. The selected samples were first placed in a 250 mL bottle; the bottle was filled with 1 wt.% surfactant solution and treated in an ultrasonic bath. NaOH solution was added to adjust the targeted pH value. Then, the samples were transferred into a 500 mL Erlenmeyer flask with a 40 °C water bath, and an agitator with an electronic motor was employed to generate turbulence. The pH of the solution under agitation was kept identical to that of the pre-soaking process. After the main de-inking experiment, the samples were placed in a household washing machine with a flushing program for 13 min to remove the residue ink and surfactants. Finally, two golf balls were placed in a washing net along with the plastic film samples to simulate the friction washing process in industrial recycling plants. A list of the different experiment sets used in this study is shown in Table 3.

Table 3. Experiment sets for the small-scale cleaning process.

Sample Name	Surfactant	pH
S1	CTAB, 1 wt.%	12
S2	CTAB, 1 wt.%	10
S3	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.%	10
S4	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.%	11
S5	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.%	12
S6	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.% plus 1 wt.% macroemulsifier DA850	10
S7	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.% plus 1 wt.% macroemulsifier DA850	11
S8	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.% plus 1 wt.% macroemulsifier DA850	12

3.2.2. Scale-Up

The experiment was scaled up to produce a sufficient amount (100 g) for laboratory-simulated mechanical recycling under selected conditions from the setups in Section 2.1. The printed and unprinted parts of the sample were separated by hand to compare the washing performance. The unprinted part was used as the blank sample. All the other samples were randomly selected. In the upscaling, the soaking process was kept identical. In addition, a 1 L PE bottle was added to provide a suitable volume during the soaking. For the agitation process, a 5 L reactor with two baffle plates installed inside was applied to generate turbulence during the washing process. A water bath was used to heat the reactor by using a submersible heater (Unold 58915). The rinsing process was kept identical as well. After washing, the plastic film samples were dried in a vacuum drying cabin (Steinberg SBS-ADO-2500) at 60 °C for 12 h. One sample with printed LDPE film (P) and one with selected unprinted LDPE film (NP) were washed at 80 °C with 2 wt.% NaOH as control groups. An overview of the different setups is presented in Table 4.

Table 4. Experiment setups of the scaled-up cleaning experiment.

Sample Name	Surfactant	T (°C)	pH
SU **-1	CTAB, 1 wt.%	40	10
SU-2	CTAB, 1 wt.%	40	12
SU-3	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.%plus 1 wt.% macroemulsifier DA850	40	10
SU-4	oxirane, methyl-, polymer with oxirane, iso-alkyl alcohol 1 wt.%plus 1 wt.% macroemulsifier DA850	40	12
P *	-	80	12.7 (RT)
NP *	-	80	12.7 (RT)

* Sample P and NP were not treated with ultrasonic pre-soaking. ** SU = Scaled-up.

3.3. Cleaning Performance Evaluation

3.3.1. CIELAB (International Commission on Illumination) Method

The de-inking performance calculation method is adapted from Gecol et al., 2001 [40], and Ye et al., 2022 [45]. The de-inked sample, the unprinted part, and the original samples were measured together under the same light surrounded by a colorimeter. The DE* (de-inking rate) value was calculated by comparing the three samples based on the measured CIELAB values (L*a*b). The unprinted sample was defined as 100% DE*, and the original sample was defined as 0% DE*.

The DE* value is calculated as shown in Equation (1).

$$\mathrm{DE}^* = \sqrt[2]{\Delta \mathrm{L}^2_{\mathrm{sample-blank}} + \Delta \mathrm{a}^2_{\mathrm{sample-blank}} + \Delta \mathrm{b}^2_{\mathrm{sample-blank}}} \times \mathrm{DE}^{*}_{\mathrm{blank}}{}^{-1} \quad (1)$$

where:

DE*: de-inking rate;
$\mathrm{DE}^*_{\mathrm{blank}}$: DE* value of reference sample (virgin polymer)
L: the value of lightness;
a: green-red opponent colors, a negative value for greenish and a positive value for reddish;
b: blue-yellow opponent colors, a negative value for blueish and a positive value for yellowish.

The washed samples from the small-scale washing experiment were evenly distributed onto a 150 × 100 mm area in a closed room without a window under a constant light source. Then, 6 points were selected to measure the L*a*b values. Finally, the CIELAB values were averaged as the final value of the sample.

The color of the washed samples from the scaled-up experiment was measured by the same method on the test specimens after extrusion and injection molding.

3.3.2. Flake Sorting Method

Due to the inhomogeneity of the post-consumer plastic waste, the cleaned samples were manually selected into three categories: completely transparent, partly transparent, and not cleaned. Then, the weight of the three categories were measured, and the DE* was calculated by Equation (2).

$$\mathrm{DE}^* = \left(\mathrm{m_{ct}} + \mathrm{m_{pt}} * 0.5\right) / \mathrm{m_{sample}} \quad (2)$$

where:

DE*: de-inking rate
$\mathrm{m_{ct}}$: mass of completely transparent flakes (g);

m_{pt}: mass of partly transparent flakes (g);
m_{sample}: mass of the whole selected flakes (g).

For the samples from the scaled-up experiment, 5 samples were selected according to the sampling method in the LAGA PN 98 [52] to ensure the selected samples were representative. Then, the DE* was calculated by averaging the values obtained from each sample.

3.4. Quality Analysis of Recyclate

After obtaining a sufficient amount of the LDPE film flakes, the samples were extruded by a 16 mm double-screw extruder (Thermo Fischer Scientific, Polylab OS) with an L/D ratio of 25/1. The extrusion temperature of the different zones is 160 °C, 175 °C, 205 °C, 210 °C, and 220 °C (from the funnel to die). Next, a laboratory shredder cut the extruded filaments into granulates (Retsch SM 300). The test specimens were produced by an injection–molding machine (Babyplast 6/10) according to the standard ISO 294-1, type 5A [53]. The melt flow index (MFI) of the recyclates was measured using a melt flow indexer (Zwick & Roell Mflow) at 190 °C with 5.0 kg of weight according to the standard ISO 1133-1 [54]. The DSC analysis was carried out with a differential scanning calorimeter (Mettler Toledo DSC 1) under a nitrogen atmosphere with a 20 mL/min flow. The operation cycle started with heating up from room temperature to 200 °C in 17 min and cooling down from 200 °C to room temperature in 16 min. The crystallinity of the LDPE was calculated using Equation (3) [55].

$$\chi_c = {}^{\Delta H}/_{\Delta H_{c,\ 100\%}} \tag{3}$$

where:

χ_c: Crystallinity in %;
ΔH: Enthalpy of fusion (J/g);
$\Delta H_{c,100\%}$: Enthalpy of fusion assuming 100% crystallinity, here 293 J/g [66].

A tensile strength testing machine (Zwick & Roell Z2.5) was applied to measure the elongation modulus, the tensile strength, and strain at the break of the recyclates according to ISO 527 [67]. Each sample was measured with five replications.

3.5. Evaluation of the Substitutability

The quality parameters obtained from Ch.2.4 were evaluated together and adapted from the method developed by Demets et al [68]. The detailed calculation is presented in Table 5.

Table 5. Calculation method for evaluating the quality of the recyclates, adapted from [68].

Recycling Quality (RQ)	Parameter	Range/Equation for RQ				
RQ^{proc}	MFI	0–0.1875 RQ = 0	0.1875–0.25 RQ = 16 * MFI−3	0.25–4 RQ = 1	4–5 RQ = −MFI + 5	>5 RQ = 0
RQ^{mech}	E-Modulus (E)	0–89.6 RQ = 0.011161E	89.6–300 RQ = 1	300–600 RQ = −0.00333 + 2	>600 RQ = 0	
	Yield strength (σ_y)	0–10 RQ = 0.1 σ_y	>10 RQ = 1			
	Strain at break (ε_b)	0–300 RQ = 0.00333 ε_b	>300 RQ = 1			

The RQ^{mech} was calculated by Equation (4).

$$RQ^{mech} = \left(RQ^{E} + RQ^{\sigma_y} + RQ^{\varepsilon_b}\right)/3 \tag{4}$$

where:

RQ^{mech}: recycling quality score of the mechanical property;
RQ^{E}: recycling quality score of the E-modulus;
RQ^{σ_y}: recycling quality score of the yield strength;
RQ^{ε_b}: recycling quality score of the strain at break.

The substitutability score $\alpha^{rec:vir}$ of the recyclate was calculated with Equation (5).

$$\alpha^{rec:vir} = \min\left(RQ^{mech}, RQ^{proc}\right) \tag{5}$$

4. Results

4.1. De-Inking Effect of Small-Scale Experiment

The CIELAB values of each sample and the DE* values are presented in Figure 4a,b.

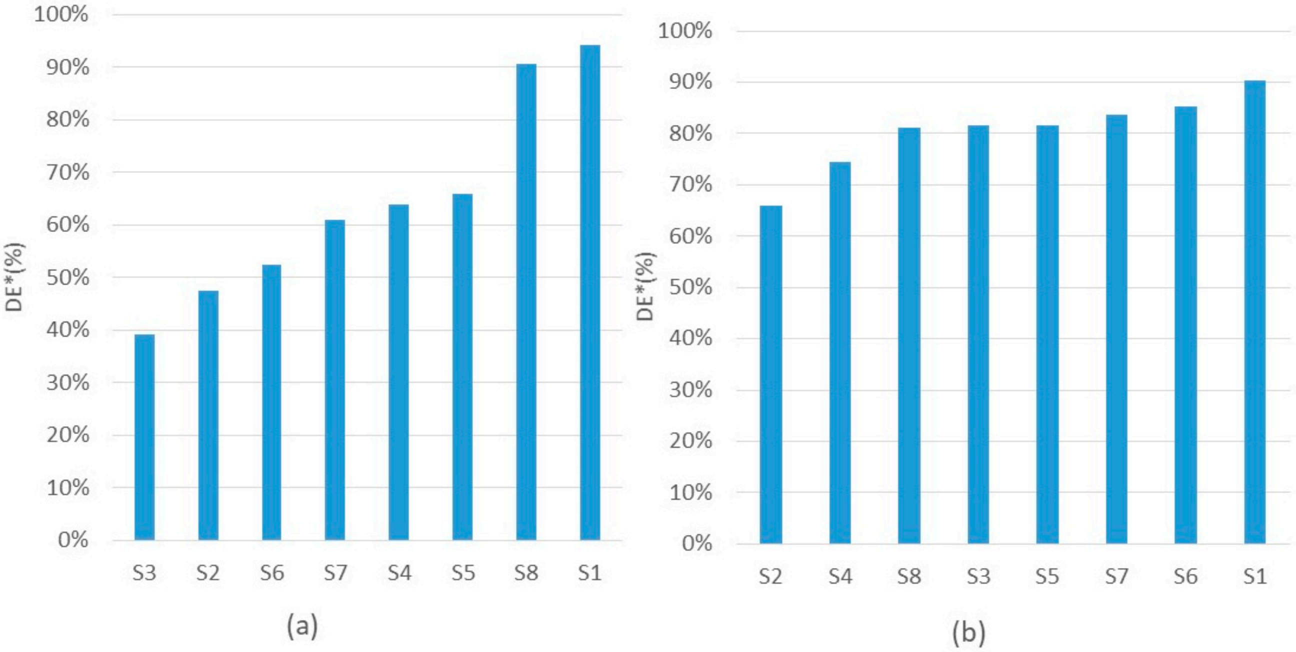

Figure 4. De-inking effect from the small-scale experiment. (**a**) CIELAB values of the small-scale samples. (**b**) De-inking rate by sorting the un-deinked flakes.

From the color measurement of the de-inking effect, CTAB at pH 12 (Sample S1) was measured with the best performance from both measurements of a de-inking effect of 94.19% and 90.26%, respectively. In the colorimeter analysis, sample S8 showed the second-best de-inking performance of 90.63%. With the flake sorting method in Figure 4b, samples S6 and S7 showed the second better de-inking result. From the small-scale experiment, it can be concluded that applying the macroemulsifier DA850, present in samples S6, S7, and S8, can enhance the de-inking performance. However, the influence of different pH values on the de-inking effect of the nonionic surfactant was not evident.

4.2. De-Inking Effect of Large-Scale Experiment

The six samples from the scaled-up experiment after extrusion are shown in Figure 5. The CIELAB values of each sample after extrusion and the DE* values are presented in Figure 6a,b.

Figure 5. The recyclates of the six scaled-up samples after extrusion.

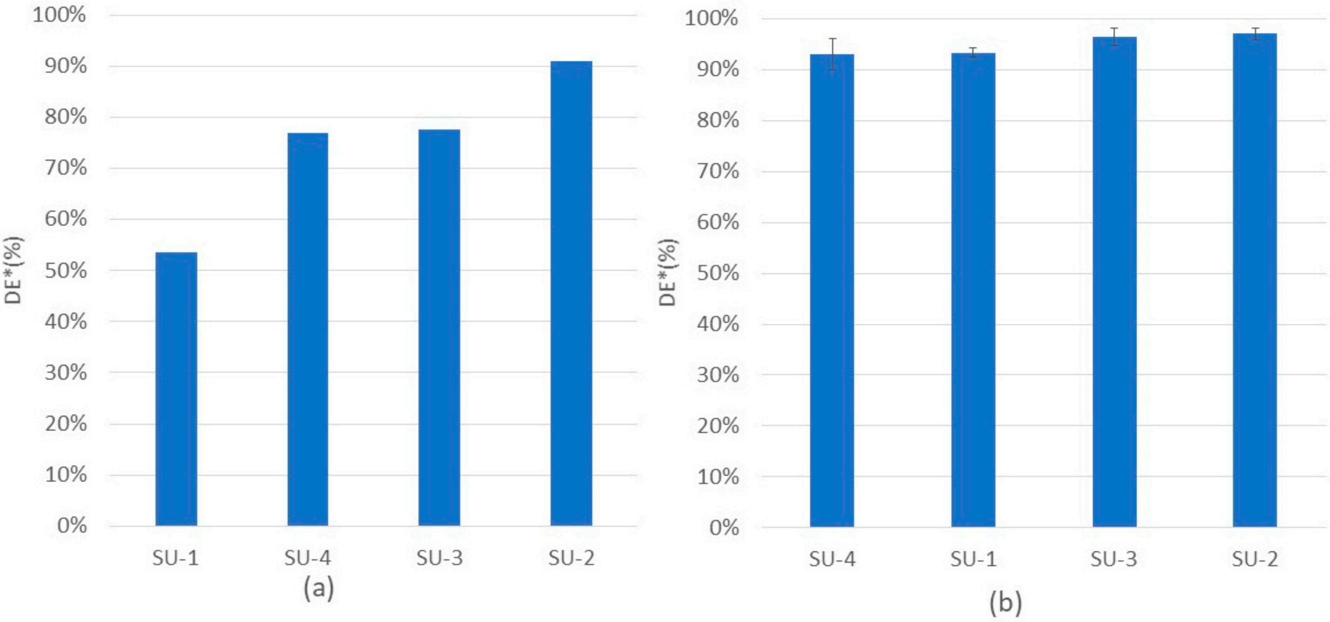

Figure 6. De-inking effect from the scaled-up experiment. (**a**) CIELAB values of the scaled-up samples by measuring the test specimen after extrusion. (**b**) De-inking rate by sorting the un-deinked flakes.

In the scaled-up experiment, comparable de-inking effects were obtained from the CIELAB method (Figure 6a) and flake sorting methods (Figure 6b). However, after extrusion and test specimen production by injection molding, the sample SU-2 showed the best de-inking performance under the CIELAB method. In contrast, the sample treated under pH 10 showed a brownish color, which might have resulted from the degradation product of nitrocellulose. The samples SU-3 and SU-4 showed a slightly worse de-inking effect than SU-2. This difference is also amplified by the CIELAB method, as the color remains evenly distributed in the sample in the extrusion process. No evidential difference in the de-inking performance between pH 10 and 12 was inspected for the samples treated with nonionic surfactant, which confirms the result from the small-scale experiment.

4.3. Material Test

4.3.1. Melt Flow Index (MFI)

From Figure 7, samples SU-2, SU-3 and SU-4 showed a similar melt flow index of 0.245, 0.225, and 0.220 g/10 min, respectively, which were lower than the non-printed sample (NP, MFI = 0.31). On the other hand, the printed sample (P) under mechanical recycling showed the highest melt flow index (MFI) of 0.435 g/10 min at 5 kg and 190 °C, which was even higher than the extruded non-printed part. According to the literature, the MFI value of LDPE for film blowing should have a minimum MFI of 0.25 g/10 min at 190 °C and 5 kg, indicated in Figure 7 with a blue line as a benchmark [68].

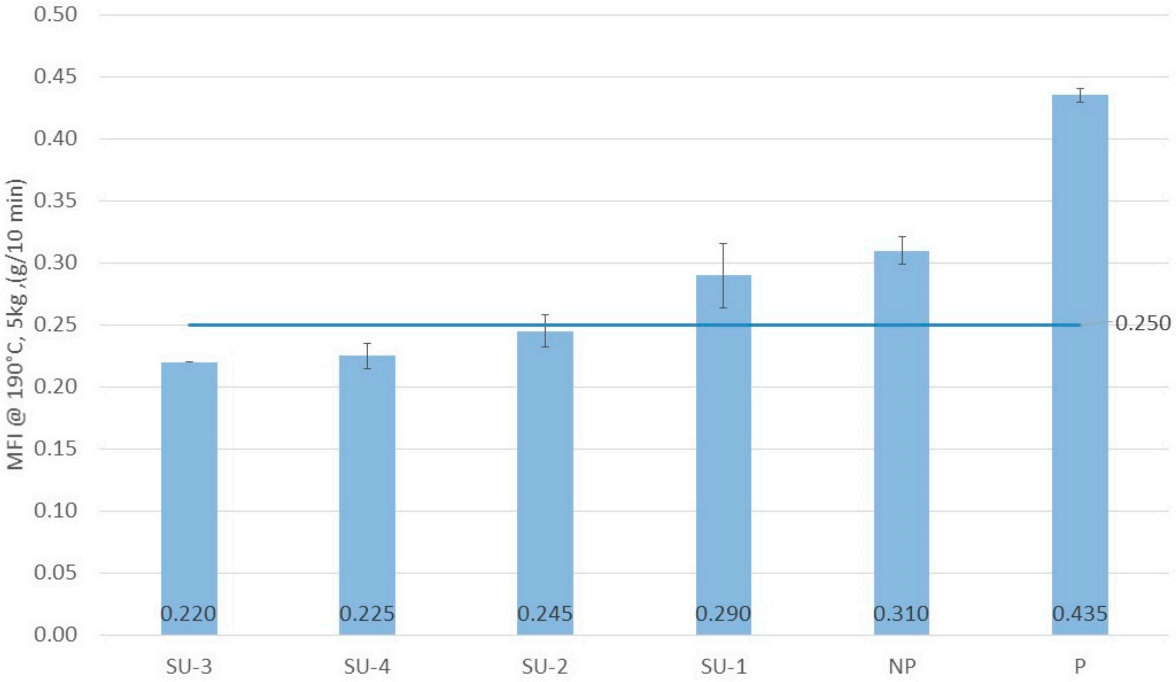

Figure 7. The melt flow index (MFI) of the samples from the scaled-up experiment.

4.3.2. DSC Analysis

The melting points, crystallization temperature, and crystallinity of the scaled-up test samples are shown in Table 6; the DSC curves of all the scaled-up samples are shown in Figure 8.

Table 6. Melting temperature, crystallization temperature, and crystallinity of each sample according to the differential calorific scanning (DSC) analysis.

Sample Names	T_m (°C)	T_c (°C)	X_c (%)
SU-1	112.12	126.17	44.92
SU-2	111.15	126.58	44.63
SU-3	110.71	127.38	50.14
SU-4	112.45	125.44	49.72
NP	111.15	126.58	46.52
P	110.29	124.16	40.67

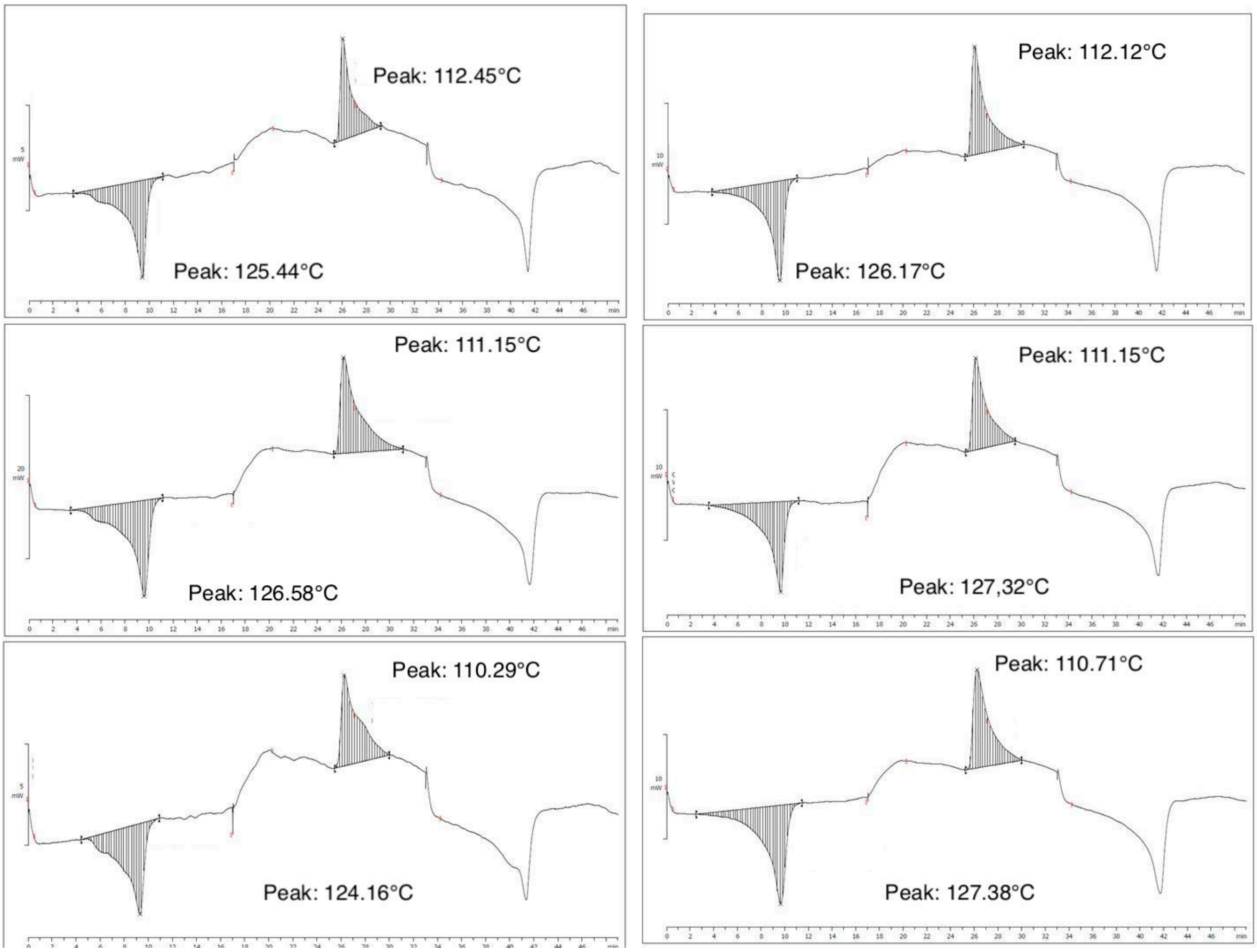

Figure 8. The DSC curves of all the scaled-up samples.

From the DSC analysis, all the samples kept similar melting and crystallization temperatures. However, the crystallinity of the samples showed a different trend. The samples treated with the oxirane nonionic surfactant had a higher crystallinity (50.14% and 49.12%), followed by the samples treated with cationic surfactant (approx. 45%). The non-printed LDPE film showed a slightly higher crystallinity of 46.52%. The printed film without de-inking had the lowest crystallinity of 40.67%.

4.3.3. Tensile Strength

The E-modulus, tensile strength, and strain at the break of each sample are shown in Figure 9a–c. The partly de-inked sample SU-1 showed the highest E-modulus of 521.72 ± 43.32 MPa. All the other samples were measured with comparable tensile strengths as the standard deviation ranges overlapped each other. For example, the lowest E-modulus was 334.84 ± 41.91 Mpa, measured from sample SU-2; the second highest E-modulus was 383.51 ± 44.13 Mpa, measured from sample SU-4.

From the tensile strength values, sample SU-1 also showed the highest value of 28.77 ± 3.2 Mpa, while sample NP showed the lowest value of 19.54 ± 3.16 Mpa. The tensile strengths of all the printed and de-inked samples are comparable, as depicted by the overlapped standard deviations.

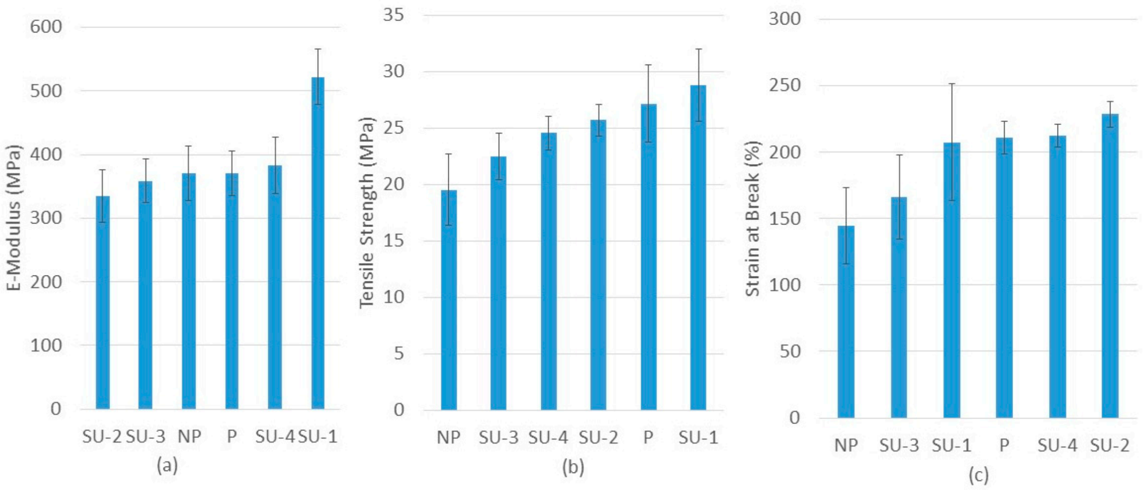

Figure 9. Results of the tensile test. (**a**) E-modulus; (**b**) tensile strength; and (**c**) strain at break.

From the strain at break, samples SU-1, SU-2, SU-4, and P showed comparable values in the strain at the break due to the overlapping of standard deviations. Samples NP and SU-3 showed a reduced strain at break compared with the other samples.

5. Discussion

5.1. Cleaning Effectiveness of Different Conditions

From Figure 4b, the washing setup of SU-2 and the setup of SU-4 showed the best de-inking effect of 94.19% and 90.63%, respectively. This confirms a previous study that a pH value between 11–13 contributes positively to the de-inking effect in general [41]. However, the flake counting method showed different results, except for SU-2 whichstill shows the best de-inking effect of 90.26%, followed by the setup SU-3. This result contradicts the color measurement effect, which showed a relatively high standard deviation (11% to 76%), indicating that the CIELAB might not be the most suitable method to be applied for measuring the de-inking effect of post-consumer plastic flakes.

From the results of the small-scale experiment, the combination of the nonionic surfactant plus a macroemulsifier (S6, S7 and S8) showed a slightly worse de-inking effect than the reference method S1 using the cationic surfactant CTAB. Moreover, this de-inking effect was shown independently of the pH values with the nonionic surfactant applied in this study.

In Figure 6a, all the setups showed an over 90% de-inking effect, with a maximum of 3% standard deviations, manually selecting the flakes with printing ink remaining (flake sorting). By measuring the color of the test specimens, it was shown that samples SU-3 and SU-4 had similar colors. However, a brown color was observed from sample SU-1. This color might have resulted from the nitrocellulose as the binder material decomposes during mechanical recycling or from the pigment residue. The difference between the SU-1 sample and the printed sample P—by simulating the state-of-art mechanical recycling—indicated that the washing at pH 10 with the cationic surfactant did not remove the color completely.

5.2. De-Inking Effect and Recyclate Quality

All de-inked polymer samples showed a decrease in the melt flow index, which indicated a degradation in the pretreatment process. Since the degradation of LDPE results in the cross-linking of the degradation products, which decreases the melt flow index (MFI) [69], the degradation could be caused by the ultrasonic washing applied in the pretreatment for samples SU-1 to SU-4 [70]. Another phenomenon is that the undeinked sample (P) showed a higher MFI than the non-printed sample (NP). One possible explanation is that the rheology modification additives in the printing ink changed the melt flow behavior of the LDPE recyclates [71]. Alternatively, this might have resulted from the air bubbles in the recyclates, possibly generated from printing ink decomposition under the heat from extrusion [33]. This trend could also confirm that the difference between the three more completely de-inked samples (SU 2–4) showed lower MFIs than the partly de-inked sample SU-1. From the different MFIs measured for each sample, the following conclusion can be drawn:

(1) Ultrasonic pretreatment degraded the LDPE polymer, resulting in the reduction of MFI;

(2) The remaining ink resulted in an increase in MFI either from the rheological additives or from the air bubbles in the granulates.

The two samples (SU-3 and SU-4) treated with nonionic surfactant showed a higher crystallinity than the non-printed polymer and a slightly darker color than an ideally complete de-inking. These two phenomena illustrate that the printing ink residue functioned as a nucleating agent in the recyclates and induced crystallinity. On the other hand, the sample treated with cationic surfactant showed a slightly lower crystallinity, which means that the cross-linking from the degradation had a more dominant effect on the crystallinity.

Gecol et al. reported that de-inking improved tensile strength and decreased the crystallinity of the recyclates. The mechanical property and crystallinity also decreased compared with virgin plastic [43].

In this study, the printed and extruded samples showed contradictory results. The printed and extruded samples (P) were measured with the lowest crystallinity of 40.67%. This might have resulted from the ink residue reducing the close packing ability of the polymer molecular chains. Typically, a low crystallinity contributes to reduced tensile strength and increased strain at break [72]. Another phenomenon was that the partly de-inked sample SU-1 showed the best mechanical property, possibly due to the remaining acrylic acid that enhanced the mechanical property of the plastic [73].

5.3. Substitutability Evaluation

The substitutability of the recyclates is presented in Figure 10a. However, the non-printed plastic showed better substitutability than the printed sample. A similar effect also could be derived by calculating the RQ^{mech} from the mechanical property data reported in the study of Gecol et al., shown in Figure 10b [43]. The sample extruded with printing ink outperformed the de-inked and non-printed recyclates. This is not congruent with the motivation of de-inking since the de-inking should not only deliver a recyclate with a color similar to the virgin plastic but also similar properties. However, these results may be limited due to the application of ultrasonic washing in the soaking process and the lack of melt filtration and degassing in the extrusion.

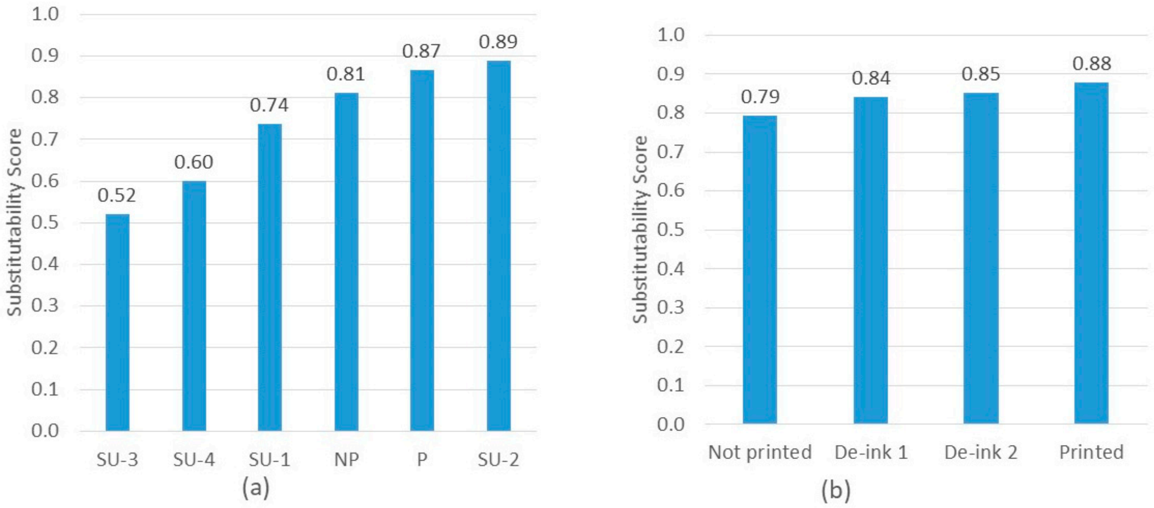

Figure 10. Substitutability score of the LDPE recyclates from (**a**) the scaled-up experiment of this study and (**b**) calculated from the data in [40].

For substituting the virgin polymers, all the de-inked samples showed lower MFI values for film production (0.25–4 g/10 min at 190 °C, 5 kg) [69]. However, it is possible to apply the de-inked recyclates to produce big bags, i.e., construction waste. Furthermore, the de-inked recyclates can be mixed with other granulates to increase the stiffness.

5.4. Limitations of This Study

The first limitation of this study is the color measurement. The color measurement should be carried out under a normed light source (D65K). However, this was not available. Therefore, the color measurement was conducted in a room without a window under a constant light source.

The second limitation of this study is the printing ink composition analysis. However, the analytic methods for the chemical composition of ink are more focused on the liquid ink before printing. An analysis method for printed ink, especially for a mixture from a post-consumer source, has not yet been developed, which could become one aspect of future work.

6. Conclusions

This study reported the de-inking effect of a nonionic surfactant assembled with a macroemulsifier. However, to measure the color change of recycled plastic, CIELAB is still the most suitable method. The precondition is that the material has a homogeneous color. The washing process with a surfactant can also be applied as an alternative or a bypass to the current hot washing process with 2 wt.% NaOH. This process also provides potential advantages from operational safety and energy-saving aspects.

Nevertheless, it is also evident that the printing ink influenced the quality of the recyclate to a more significant extent than previously reported, most likely due to the complexity of the printing ink composition, especially for the waste from post-consumer sources. The ultimate goal of applying surfactant for cleaning the plastic flakes upstream of the extrusion is to obtain an improved quality of recyclates to substitute the virgin polymer. Therefore, the influence of the printing ink compositions, their degradation products, and their possible side reaction products should be systematically investigated in the future.

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