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# Investigating the Effect of Liquid State Decontamination on the Material Properties of Post-Consumer High Impact Polystyrene Recyclate

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# Investigating the Effect of Liquid State Decontamination on the Material Properties of Post-Consumer High Impact Polystyrene Recyclate

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**Abstract:** The global focus on environmental sustainability has intensified the need for innovative plastic waste management solutions, especially in the food packaging industry. This study explores liquid state decontamination processes of recyclates produced by post-consumer waste recycling of polystyrene yogurt cups and their effects on the material properties of the recycled material. For this purpose, different decontamination processes based on a twin-screw extruder with vacuum degassing and an industrial-scale recycling machine were applied. Tests such as tensile testing, Charpy impact testing, melt flow rate (MFR) measurement, plate-plate rheometry, high-pressure capillary rheometry (HCR), and differential thermal analysis (DTA) were conducted to assess the mechanical and thermo-rheological properties as well as the oxidation induction temperature as an indicator of polymer degradation. The findings provide a comprehensive understanding of the achievable properties of polystyrene from post-consumer waste and reveal insights into the different process-induced degradation processes of the recycled polystyrene.

**Keywords:** Recyclate, Post-Consumer, Polystyrene, High Impact Polystyrene, Liquid State Decontamination, Food Packaging

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# Introduction

The global challenge of plastic waste management has gained increasing attention. In 2021, 390.7 million tons of plastic were produced worldwide, and this trend is on the rise. The environmental impact of plastic pollution, particularly from the food packaging industry, highlights the need for recycling. The European Union is aiming for a 50% recycling rate for plastic packaging by the end of 2025, up from 32% in 2020. In addition, the new targets for the recycling rates consider new points of calculation, which leads to the even bigger challenge of meeting these targets. Sustainable food packaging must have high mechanical integrity and comply with food safety standards, which requires innovative recycling solutions [1,2].

High impact polystyrene (HIPS) is commonly used in the food packaging industry due to its robustness and versatility [3]. However, recycling post-consumer HIPS, such as polystyrene yogurt cups, is challenging due to contamination issues. Recycled plastic contamination can affect both mechanical properties and potential health risks [4]. One promising approach is the decontamination process in the liquid state (melt phase super-cleaning), which offers the possibility of improving the quality and safety of recycled plastics [5].

This study examines the effects of different decontamination processes and their vacuum influences on the mechanical and thermo-rheological properties of recycled post-consumer HIPS. A twin-screw extruder with a vacuum degassing unit and an industrial scale recycling machine were used for liquid state decontamination and the properties of the post-consumer recycled HIPS produced were studied.

A comprehensive series of tests were carried out to investigate the influence of liquid state decontamination on the properties of recycled material and to gain insight into its capabilities for closed loop recycling. These tests included tensile testing, Charpy impact testing, melt flow rate (MFR) measurement, plate-plate rheometry, high pressure capillary rheometry (HCR), and differential thermal analysis (DTA).

# Material and Methods

## Material and Decontamination Processes

A company partner processed material from the German waste fraction DSD331. This waste fraction is defined as used, residue-free, rigid, system-compatible plastic items made of polystyrene, volume ≤ 1 liter, such as cups and trays, including ancillary components such as closures, labels, etc. [6]. The waste fraction was sorted on an object-related scale, shredded, hot-washed (>80°C, at least 2.5% NaOH), and sorted on a flake scale.

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After the pre-treatment, the material was extruded with a mass flow of 1300 kg/h and granulated. The extrusion step included a laser filtration and a vacuum degassing system. This was the initial material batch, henceforth referred to as "input".

In the following processing steps, different liquid state decontamination (LSD) processes were applied. On the one hand, a Leistritz ZSE 18 MAXX twin-screw extruder (Leistritz AG, Germany) was used, equipped with two vacuum ports connected to a pump. The vacuum levels of the two ports, set to "High" (300 mbar) and "Low" (700 mbar), were varied to assess their effect on the material. Other machine parameters remained unchanged: 240°C, 258 rpm, no filtration, and a mass flow of 3 kg/h. Three balanced vacuum settings were used:

- High-High
- Low-Low
- Non-Non

Two other settings were used to determine the effects of a dual versus single vacuum degassing system. In this setting, only the vacuum port on the die side of the extruder was used, leaving the other port open to draw in air.

- High-Non
- Low-Non

Additionally, an industrial-scale super-cleaning process derived from PET recycling was used. The process used high vacuum to extract volatiles, with a residence time of ~25 min to achieve a "super-cleaned" material batch [5].

## Characterization

Mechanical properties were examined by conducting tests on injection-molded multipurpose and Charpy specimens following ISO 294-1 and ISO 19063-2. A victory 60 injection molding machine (Engel, Austria) was used to produce the specimen. Specimens were stored for 48 hours at testing conditions prior to testing.

Tensile tests were performed with a Z005 universal testing machine (Zwick Roell, Germany) with a 5 kN load cell and strain gauges, following ISO 527-2. Young's modulus was measured within a strain range of 0.05 to 0.25% at a testing speed of 1 mm/min. The testing speed was then increased to 50 mm/min until failure. Tests were conducted at 23°C and 50% relative humidity.

Charpy impact tests were carried out using an HIT25P impact tester (Zwick Roell, Germany), according to ISO 179-1. Specimens were notched with a type A notch utilizing an RM2265 microtome (Leica, Germany). Unnotched and notched tests used 5 J and 0.5 J pendulums, respectively, at 23°C and 50% relative humidity.

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MFR tests used an Mflow Extrusion Plastometer (Zwick Roell, Germany) at just one temperature of 200°C with a 5 kg weight, following ISO 1133-1. The other rheological measurements (plate-plate rheometry and HCR) were performed at 200°C, 220°C, and 240°C.

Plates for plate-plate rheometry were pressed at 230°C and 1 bar using a Manual Hydraulic Press 15 Ton (Specac Ltd, UK). For the measurements, an MCR 502e (Anton Paar, Austria) following ISO 3219-2 was used. Tests covered a frequency range from 628 to 0.05 rad/s at the three temperatures mentioned above.

HCR viscosity measurements on a Rheograph 25 (Göttfert, Germany) used pellets and a 1 mm die of either 20 or 0.1 mm length, allowing the Bagley correction to be applied. The tests were carried out according to ISO 11443 with three different material batches (input, super-cleaned, and high-high) in the shear rate range between $10^4$ and $10^1$ $s^{-1}$. Results were corrected using the Weißenberg-Rabinowitsch method.

DTA measurements to examine material degradation were performed to characterize oxidation induction temperature (OIT) following ISO 11357-6. Specimens were cut into plates, put into aluminum pans, and heated from 30°C to 240°C at 10 K/min using a DSC 4000 (Perkin Elmer, USA). The onset of the oxidation peak was measured.

# Results and Discussion

## Mechanical Properties

The tensile properties and in particular the tensile modulus ($E_t$) are very important for a thermoformed product, such as a cup [7]. The input batch reached a modulus of 2176 MPa, which was not significantly different from the other batches, shown in Table 1. Characterization of the yield stress ($\sigma_y$) indicated that processing had a positive influence, especially at higher vacuum levels, with an almost 10% improvement due to the super-cleaning process. All the extruded material batches showed higher values for the strain at break ($\varepsilon_b$). The influence of vacuum setting during extrusion showed only a slight trend towards higher strain at break for higher vacuum levels. The most significant differences can be observed in the super-cleaned batch with high yield and post-yield stresses but low strains at break values. The respective stress-strain curves are depicted in Figure 1. Only small standard deviations were detected in tensile testing.

The Charpy impact tests of the unnotched specimens showed high standard deviations ($\sigma\_a_{cU}$) due to imperfections in the material. The input material achieved the lowest Charpy impact strength of the unnotched specimens ($a_{cU}$) as shown in Table 1. Due to the high spread of the result values, notched specimens ($a_{cN}$) were additionally characterized. The results obtained revealed a trend that was opposite to that of the unnotched specimens (see Figure 1[b]). With these specimens, the

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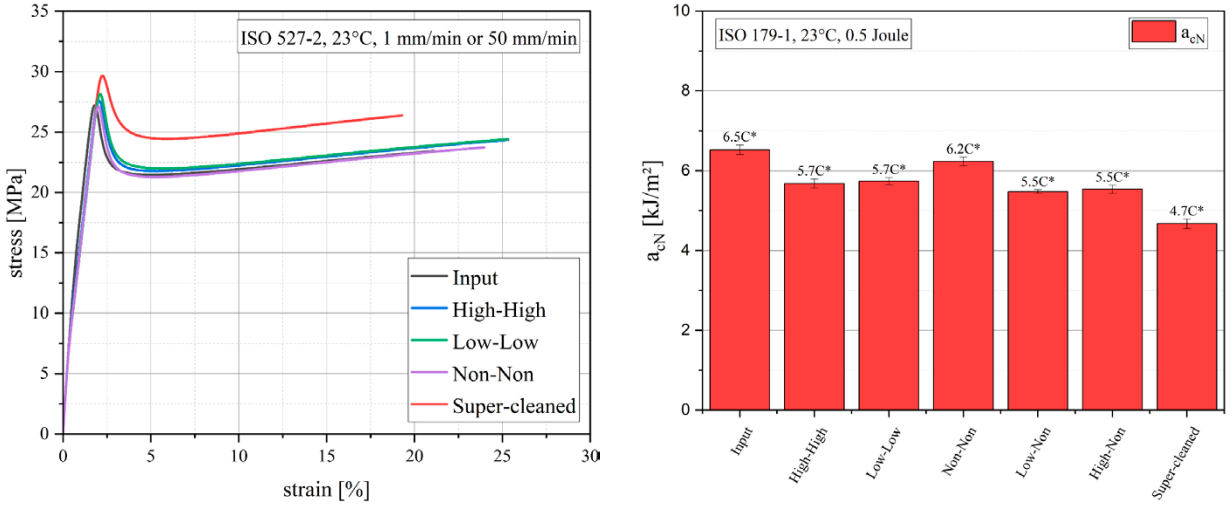
input batch shows the highest impact strength values followed by the extruded material batches, where the vacuum level had no significant influence. The lowest impact strength was achieved by the super-cleaned batch.

**Table 1.** Mechanical properties of the tensile testing and of the Charpy unnotched & notched impact testing.

| | $E_t$ | $\sigma_y$ | $\varepsilon_b$ | $a_{cU}$ | $\sigma\_a_{cU}$ | $a_{cN}$ | $\sigma\_a_{cN}$ |
|---|---|---|---|---|---|---|---|
| | MPa | MPa | % | [kJ/m²] | [kJ/m²] | [kJ/m²] | [kJ/m²] |
| Input | 2176 | 27.3 | 21.1 | 38.3C* | 6.1 | 6.5C* | 0.12 |
| High-High | 2178 | 28.1 | 25.3 | 56.6C* | 14.5 | 5.7C* | 0.11 |
| Low-Low | 2163 | 28.2 | 25.3 | 53.7C* | 16.0 | 5.7C* | 0.09 |
| Non-Non | 2174 | 27.2 | 24.0 | 56.2C* | 17.4 | 6.2C* | 0.11 |
| High-Non | 2169 | 29.0 | 24.2 | 54.0C* | 11.6 | 5.6C* | 0.06 |
| Low-Non | 2164 | 28.9 | 24.9 | 50.6C* | 10.6 | 5.5C* | 0.10 |
| Super-cleaned | 2153 | 29.7 | 19.3 | 52.3C* | 11.3 | 4.7C* | 0.12 |




(a) (b)

**Figure 1: (a) Stress-strain-curves from tensile testing and (b) Charpy notched impact strengths.**

## Rheological Properties

First, the MFR was measured. The input batch achieved an MFR of 4.8 g/10min that corresponds to a melt volume rate (MVR) of 5.0 cm³/10min, resulting in a melt density of 0.97 kg/dm³. The melt density remained unchanged, but every extruded material batch achieved higher MFR values than

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the input batch, regardless of vacuum level, as shown in Table 2. The lowest MFR value can be observed in the super-cleaned batch.

The oscillating plate-plate rheological measurement showed no major differences in the vacuum setting while extruding. The differences between the input and extruded material batches were also not significant. The comparison between the input and the super-cleaned batch confirmed the results of the MFR. With small differences, the super-cleaned batch achieved a higher complex viscosity at all temperatures and all circular frequencies.

The rheological measurements were completed by the HCR viscosity analysis to show the behavior of the materials at higher shear rates. The input and the high-high batches revealed the same result as in the plate-plate rheological measurements: no significant differences. The super-cleaned batch again achieved the highest viscosities, with differences approaching zero at the highest shear rates.

Combining the plate-plate and HCR rheological measurements revealed the possibility of superpositioned curves to provide information over a wide shear rate range, indicating the applicability of the Cox-Merz rule [8]. After correcting the HCR results, all curves showed a good overlap between the two different test systems, as illustrated in Figure 2(a). The curves were merged at the intersection, shown in Figure 2(b). This gave us single curves from shear rates of 0.5 $s^{-1}$ to 20,000 $s^{-1}$.

Looking at the rheological properties revealed a controversial change in properties caused by the processing. The viscosity, in particular the MFR, showed that the extrusion process increased the MFR, and the LSD process decreased it. The increase might have been due to degradation of the polystyrene matrix by chain scission, and the decrease indicated cross-linking of the polybutadiene particles in the polystyrene matrix due to the increased process time and thermal stress on the material [9].

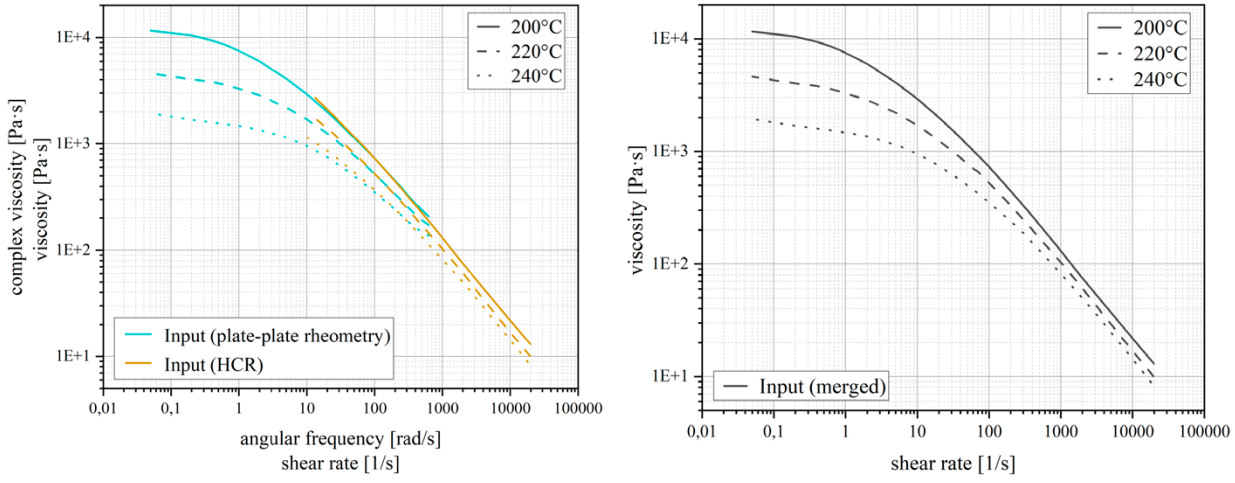


**Figure 2: Viscosity curves of the input material: (a) overlapping curves from plate-plate rheometry & HCR rheometry and (b) merged viscosity curves.**

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## Thermal Properties

DTA characterization revealed several transition temperatures: polystyrene glass transition at 95-100°C, small peaks at 125°C and 165°C from traces of foreign polymers (polyethylene and polypropylene), and the crucial oxidation peak onset. The input batch achieved an OIT of 181.6°C. The extruded material batches, independent of the vacuum level, showed similar values within the standard deviation. The super-cleaned batch, on the other hand, only reached 173.4°C, indicating measurable degradation through the LSD process. This demonstrated a clear difference in degradation between the extrusion and the LSD processes due to different processing times at high thermal stresses. This finding is consistent with the duality of degradation in the rheological properties mentioned above.

**Table 2.** Rheological properties of MFR testing and thermal properties of DTA measurements.

| | MFR | σ_MFR | MVR | σ_ MVR | $\rho_m$ | OIT | σ_OIT |
|---|---|---|---|---|---|---|---|
| | g/10min | g/10min | cm³/10min | cm³/10min | kg/dm³ | [°C] | [°C] |
| Input | 4.8 | 0.07 | 5.0 | 0.08 | 0.970 | 181.6 | 1.67 |
| High-High | 5.2 | 0.11 | 5.3 | 0.11 | 0.968 | 179.9 | 0.81 |
| Low-Low | 5.6 | 0.09 | 5.8 | 0.09 | 0.968 | 180.3 | 0.60 |
| Non-Non | 5.2 | 0.07 | 5.4 | 0.08 | 0.969 | 182.2 | 1.27 |
| High-Non | 5.2 | 0.09 | 5.4 | 0.09 | 0.968 | 181.1 | 0.64 |
| Low-Non | 5.4 | 0.09 | 5.5 | 0.10 | 0.968 | 180.3 | 1.39 |
| Super-cleaned | 4.3 | 0.09 | 4.5 | 0.09 | 0.969 | 173.4 | 1.15 |

## Conclusion

The vacuum setting during twin-screw extrusion had an almost negligible influence on the mechanical, rheological, and thermal properties. Comparing input with any extruded material batches, the extrusion itself showed only a minor influence on the mechanical properties. The rheological or thermal properties were not significantly affected, except for a higher MFR due to the chain scission degradation caused by extrusion. The super-cleaned batch showed the greatest changes in properties. Here, the mechanical properties displayed that it could withstand higher tensile stresses but lost some of its impact strength. The rheological properties revealed higher viscosities in each test method due to the cross-linking degradation of the polybutadiene in the LSD process, which also correlates with the higher tensile strength and lower ductility measured in the mechanical tests. The OIT results showed almost no degradation in the extruded material batches but high degradation in the super-cleaned batch, in line with the degradation hypothesis. In conclusion, multiple closed-loop recycling of HIPS with an LSD process could cause problems due to material degradation and, therefore, this type of recycled product requires good quality control.

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