

Thermal and Physical Performance of Recycled PET and EPS at Different Recycling Levels

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ABSTRACT

The continuous increase in waste production, driven by economic growth and population expansion in Indonesia, highlights the urgent need for a deeper understanding of sustainable waste management practices. This study aims to analyze the impact of different recycling levels on the thermal and physical properties of polyethylene terephthalate (PET) and expanded polystyrene (EPS), both of which significantly contribute to environmental pollution. This research examines recycling frequencies of one, three, and five cycles, utilizing advanced testing methodologies such as Differential Scanning Calorimetry (DSC) for thermal property analysis and Melt Flow Index (MFI) for physical characterization. The study seeks to uncover the structural transformations that occur in these plastics throughout successive recycling processes and their subsequent implications. The PET test results indicate that an increased recycling frequency tends to elevate the melting point, reduce crystallinity, and enhance melt flowability.

INTRODUCTION

Economic growth and population expansion in Indonesia have led to a proportional increase in waste generation. Plastic waste, particularly polyethylene terephthalate (PET) and expanded polystyrene (EPS), has become a dominant presence in landfills. PET is widely used in manufacturing single-use containers for carbonated beverages, mineral water, and milk. Due to its short lifespan and high production volume, PET packaging presents significant challenges in determining the most effective disposal method. Various approaches exist for managing post-consumer PET bottles, with landfilling being the most cost-effective yet least environmentally and economically viable option (Jakić et al., 2023).

Expanded polystyrene (EPS) is commonly used for take-out food containers due to its lightweight, rigidity, excellent insulation properties, and high impact resistance (Ho, 2017). However, post-consumer EPS containers often end up in municipal landfills, contributing to the growing volume of waste and posing environmental concerns. Additionally, the extended degradation time of most plastics—often spanning several centuries—further exacerbates the ecological burden (Hidalgo-Crespo et al., 2022). Notably, over the past decade, polystyrene (PS) has accounted for approximately 10% of global plastic waste by weight (Aljabri et al., 2017), making it a major post-consumer waste issue due to its low recycling rates (Ankesh et al., 2021). Recycling remains a crucial strategy for mitigating plastic waste's environmental impact by reintegrating materials into the production cycle.

Currently, recycled PET is commercially available and used in various products, though the number of recycling cycles it has undergone is often unknown. Despite numerous studies investigating the thermal and flowability properties of recycled PET, data correlating these properties with repeated recycling cycles remain scarce (Nafis et al., 2023)(Roungpaisan et al., 2023)(Nik Hassan et al., 2018). Previous research indicates that recycling generally reduces the melting temperature and crystallinity of PET. Additionally, recycled PET exhibits lower viscosity than virgin PET, reflecting its molecular weight reduction (Cusano et al., 2023).

Recycling expanded polystyrene (EPS), commonly known as Styrofoam, poses challenges due to its lightweight and voluminous nature. However, several methods and initiatives exist to facilitate EPS recycling. EPS can be densified by heating and mechanically shredding the foam, converting it into a rigid material suitable for further processing through moulding techniques (Dennis & Dennis, 2022)(Waste Diversion Ontario, 2018). Studies suggest that the glass transition temperature of recycled EPS is lower than that of virgin EPS (Samper et al., 2010).

To date, no research has specifically examined the impact of recycling frequency on the thermal and rheological properties of both PET and EPS. This study aims to explore the intricate relationship between recycling cycles and the thermal and flowability characteristics of these plastics. As waste management becomes increasingly crucial to Indonesia's socio-economic development, understanding the structural transformations occurring during repeated recycling processes is essential.

Our research adopts a tiered methodology, analysing plastics subjected to recycling frequencies of one, three, and five cycles. Differential scanning calorimetry (DSC) is utilized to assess thermal property changes, while the melt flow index (MFI) measures rheological aspects, particularly flowability, of the recycled plastics. Gaining insight into the thermal behaviour and flowability of these materials will provide a foundation for optimizing product applications and processing parameters.

LITERATURE REVIEW

1. Recycling of PET and EPS: Challenges and Opportunities

Polyethylene Terephthalate (PET) and Expanded Polystyrene (EPS) are widely used in packaging and industrial applications, yet their recycling rates remain low (Hidalgo-Crespo et al., 2022). PET possesses excellent mechanical properties but undergoes molecular degradation during recycling, leading to changes in melting point, crystallinity, and melt flow index (Masmoudi et al., 2018). Meanwhile, EPS is an amorphous polymer whose glass transition temperature (T_g) shifts with increasing recycling cycles.

PET recycling is commonly performed through mechanical methods due to economic feasibility (Jakić et al., 2023). However, studies show that repeated recycling reduces PET's intrinsic viscosity due to polymer chain scission (Roungpaisan et al., 2023). Similarly, EPS loses its thermal properties over multiple reprocessing cycles, impacting its industrial applications (Dennis & Dennis, 2022).

2. Thermal and Crystallinity Changes in Recycled PET

Increasing recycling cycles in PET generally leads to reduced crystallinity and higher melting points (T_m) due to molecular structural changes caused by repeated heating. Jakić et al. (2023) found that recycled PET exhibits higher T_g due to increased mobility of degraded polymer chains. Cusano et al. (2023) further confirmed that molecular weight reduction in recycled PET affects its viscoelastic properties.

Additionally, Piccarolo et al. (2002) reported that PET undergoing more than three recycling cycles exhibits an exothermic cold crystallization peak due to amorphous structure modifications induced by reheating. These changes highlight the importance of processing parameter control to maintain optimal mechanical properties in recycled PET.

3. Impact of Recycling on EPS Thermal Properties

Unlike PET, EPS is an amorphous polymer with no distinct melting point. Studies by Samper et al. (2010) indicate that recycled EPS experiences a decline in T_g with increasing recycling cycles, suggesting polymer degradation. Ankesh et al. (2021) also observed that the initial softening temperature of EPS decreases with more recycling cycles, reflecting reduced thermal stability.

Hidalgo-Crespo et al. (2022) further reported that recycled EPS shows reduced mechanical strength due to molecular network degradation. To address this issue, research has suggested reinforcing EPS with inorganic fillers or blending it with other polymers to improve mechanical properties (Andena et al., 2019).

4. Melt Flow Index (MFI) and Processability of Recycled PET and EPS

MFI is a critical parameter in assessing the flowability of molten polymers during processing. Masmoudi et al. (2018) demonstrated that increased recycling cycles in PET raise its MFI due to molecular weight reduction. Similarly, EPS exhibits higher MFI with more recycling cycles, indicating improved flowability but potential mechanical strength loss (Bustos Seibert et al., 2022).

METHODOLOGY

Materials

The materials used in this experiment consisted of waste PET sourced from mineral water bottles. The shredding process involved breaking down PET bottles into smaller fragments. During the washing stage, contaminants were removed using hot water and detergents. The shredded PET was then dried at a temperature of approximately 120–150°C for four hours (Bustos Seibert et al., 2022), ensuring the elimination of any residual moisture.

The waste EPS used in this study was obtained from electronic appliance packaging, with bulk densities ranging from 18.5 to 14.5 kg/m³ (Acierno et al., 2010). EPS is an inert, low-density thermoplastic derived from hydrocarbons, composed of numerous spherical beads containing 2% polystyrene and 98% air (Andena et al., 2019; Ankesh et al., 2021).

Specimen Preparation

The heating process densified the shredded EPS waste at 120°C for 15 minutes. This heating phase resulted in a 95% decrease in foam dimensions by causing the softening of EPS, a phenomenon triggered within the temperature range of 100°C to 120°C (Budiyantoro & Yudhanto, 2023). Samples of PET and EPS waste are melted at a temperature of 260°C, and the melting process is conducted in four variations, namely one time, three times, and 5 times recycling. Table 1 illustrates the sample codes corresponding to their respective recycling variations.

Table 1. Sample codes corresponding recycling variations

Sample	Recycling number	Code
PET	1 time	PET-1
PET	3 times	PET-3
PET	5 times	PET-5
EPS	1 time	EPS-1
EPS	3 times	EPS-3
EPS	5 times	EPS-5

The specimen preparation process is illustrated in Figure 1, where the samples were subjected to three different heating variations and subsequently tested using DSC and MFI.

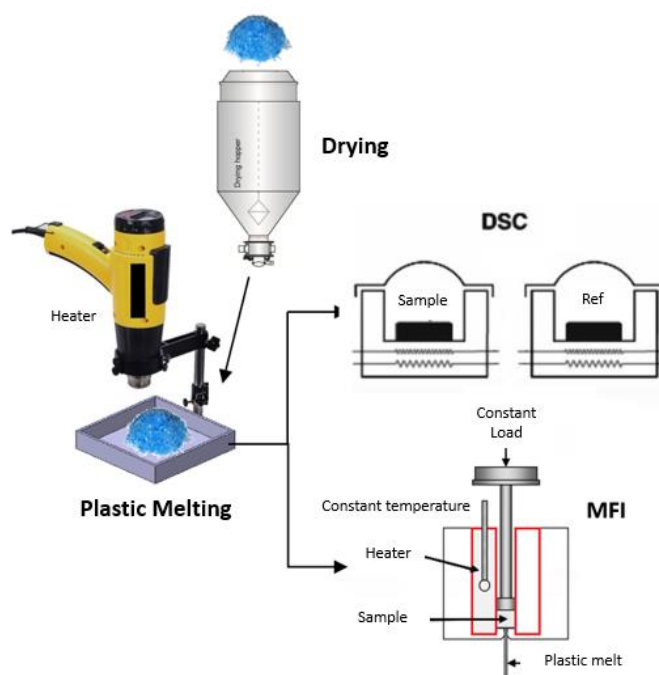


Figure 1. Sample preparation and testing

Thermal Test

The temperature-dependent analysis of the samples was conducted using a DSC-60 Plus Shimadzu instrument. The experiment was performed at a heating rate of 10 K min^{-1} , within a temperature range of $30\text{--}600^\circ\text{C}$, under a nitrogen (N_2) atmosphere. The sample weights were 4.28 mg for EPS and 6.3 mg for PET, following the ASTM D3418 standard (Pereira et al., 2020)(Budiyantoro et al., 2019).

The determination of melting point and crystallinity content applies exclusively to PET, as it is a semi-crystalline thermoplastic. In contrast, for EPS, only the glass transition temperature (T_g) was analyzed, as it is classified as an amorphous thermoplastic. The melting point (T_m) and glass transition temperature (T_g) were identified from the peaks of the DSC test curve. Additionally, the crystallinity content of PET was estimated using DSC enthalpy data and calculated based on Equation (1) (Roungpaisan et al., 2023).

$$X_c = \frac{\Delta H_i}{\Delta H_0} \times 100\% \quad (1)$$

Where:

- X_c = Degree of crystallinity (%)
- ΔH_i = Enthalpy change of heat from the tested sample (J/g)
- ΔH_0 = Enthalpy change of heat from a perfect crystalline structure. (J/g)

ΔH_i is indicated by the area under the DSC curve. ΔH_0 is the crystallinity of pure PET (140,1 J/g) (Jakić et al., 2023).

Flowability Test

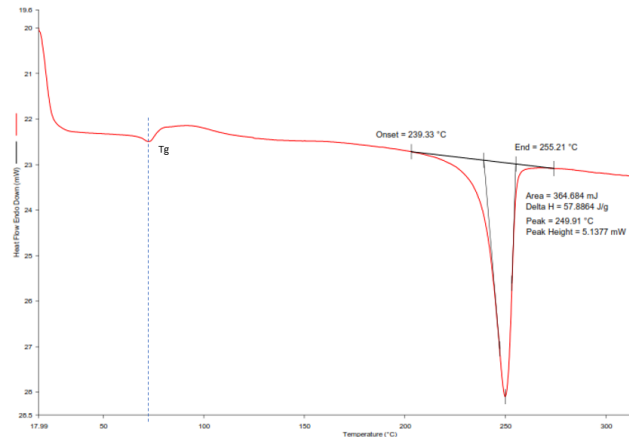
The physical property, specifically the flowability of molten PET and EPS, was evaluated using the Melt Flow Index (MFI) test, following the ASTM D1238

standard (Fernandes et al., 2014; Junaedi et al., 2022). MFI serves as an indicator of a molten material's ability to flow under pressure. During the test, the plastic is melted inside a cylinder under a 3.8 kg load. The melting temperature is set at 260°C for PET and 230°C for EPS. The MFI results are expressed in g/10 minutes.

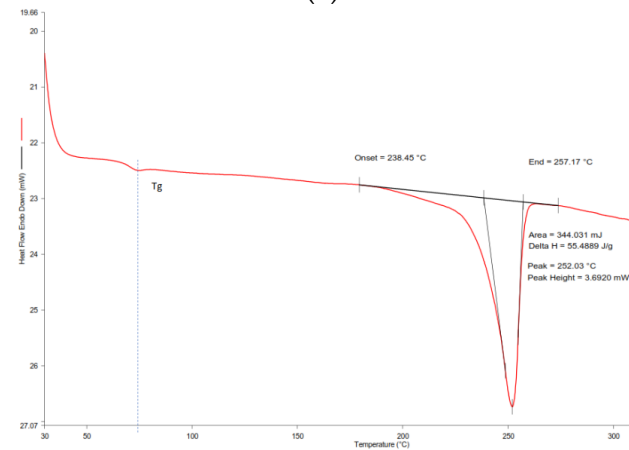
RESEARCH RESULTS AND DISCUSSION

Thermal Properties

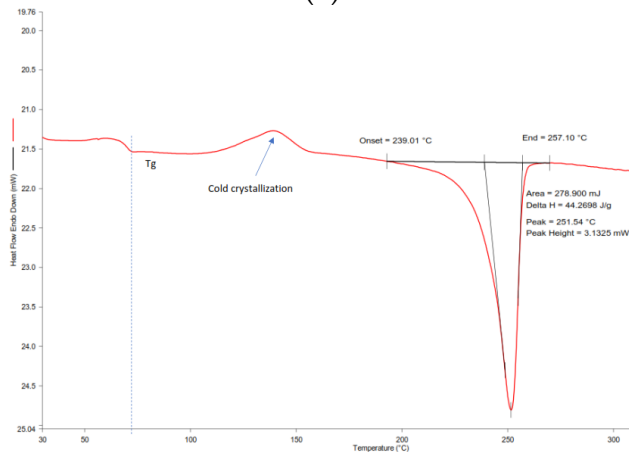
The DSC test results for PET-1, PET-3, and PET-5 are presented in Figure 2. The test results above are summarized in Table 2.



(a)



(b)



(c)

Figure 2. DSC test results: (a) PET-1; (b) PET-3; (c) PET-5.

The peaks on the DSC curve represent the melting temperature (T_m), with no significant differences observed among the three PET variations. Additionally, the DSC results indicate a slight variation in glass transition temperature (T_g).

In the DSC curve for PET-5, a cold crystallization exotherm is observed near 140.8°C. The repeated heating and cooling cycles during recycling influence the thermal history of PET. Cold crystallization involves structural modifications, leading to an increase in crystallization degree, lamella thickness, and crystal structure perfection (Piccarolo et al., 2002). This process occurs as newly formed crystallites develop at the interface between pre-existing crystalline structures and amorphous regions (Zhu et al., 2013).

As the recycling cycles increase, there is a potential for molecular degradation. Chain scission or other structural modifications may take place, fostering conditions favourable for crystalline structure formation during the cooling phase.

Table 2. DSC test results

Code	T _g	Onset	Endset	Peak
PET-1	71.5	239.33	255.21	249.91
PET-3	75.6	238.45	257.17	252.03
PET-5	74.3	239.01	257.10	251.54

The degree of crystallinity can be determined from the area under the DSC curve and calculated using Equation (1), as presented in Table 3. An increase in recycling cycles leads to a decrease in crystallinity.

The recycling process, particularly the repeated heating and cooling cycles, can disrupt and degrade the crystalline structure of PET. As a result, the degree of crystallinity tends to decline with an increasing number of recycling cycles (Hadi et al., 2014).

Table 3. Degree of crystallinity of PET

Code	Degree of crystallinity (%)
PET-1	41.31
PET-3	39.6
PET-5	31.59

The degree of crystallinity is closely linked to product quality. PET with higher crystallinity tends to be harder and exhibits greater strength, making it more suitable for solid and pressure-resistant products. Conversely, PET with lower crystallinity tends to be more transparent, which is preferable for containers requiring clarity or enhanced aesthetic appeal.

The DSC curves of EPS and PET display distinct characteristics. As a semi-crystalline thermoplastic, PET exhibits both a glass transition temperature (T_g) and a melting temperature (T_m) associated with its crystalline regions. In contrast, EPS, being an amorphous thermoplastic, shows a glass transition temperature on its DSC curve, indicating its transition from a glassy to a rubbery state, but lacks a distinct melting temperature.

Figure 3 presents the DSC curves of EPS for three different recycling cycle variations.

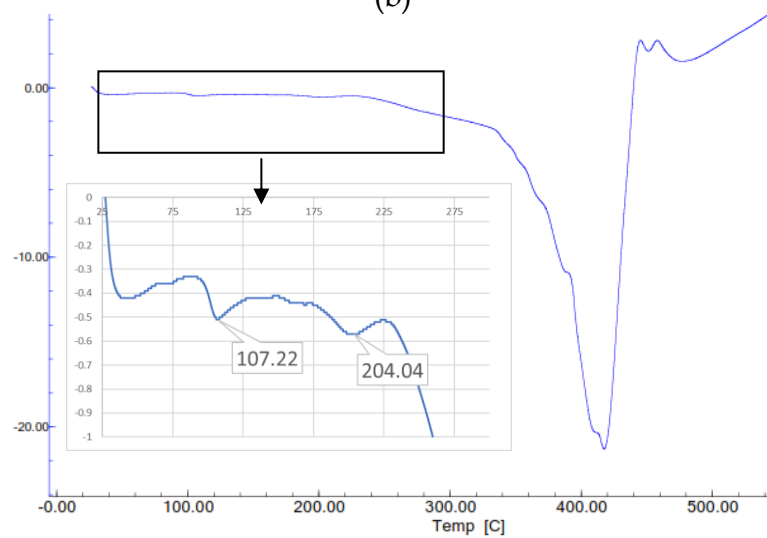
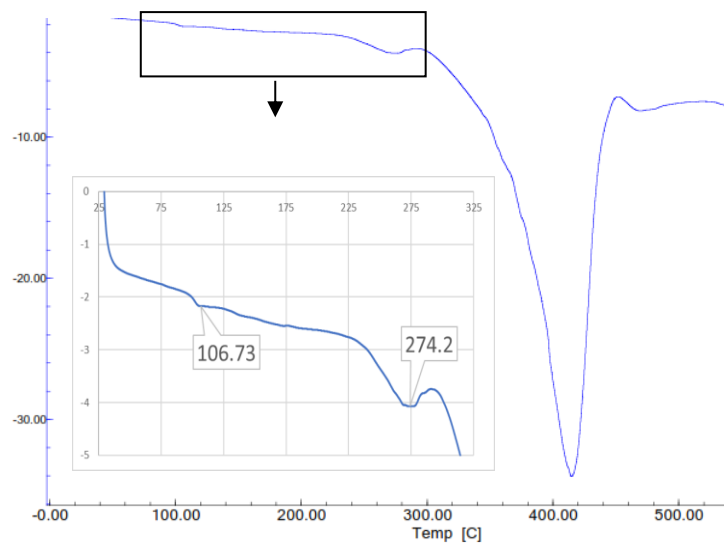
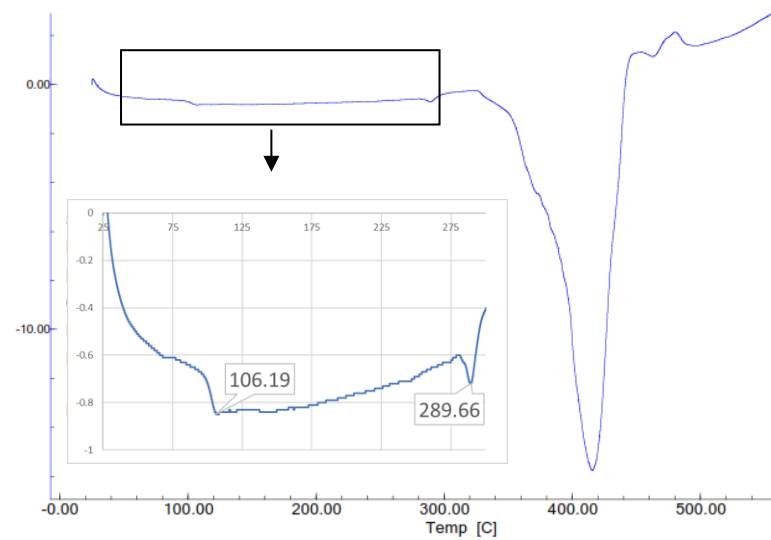


Figure 3. DSC test results: (a) EPS-1; (b) EPS-3; (c) EPS-5.

Figure 3(a) illustrates a noticeable descent at 106.73 °C, corresponding to the glass transition temperature (T_g). During the third recycling cycle, the T_g is recorded at 106.19 °C, while in the fifth recycling cycle, it slightly increases to 107.22 °C.

As the temperature rises, all recycling conditions exhibit a distinct peak, indicating thermal decomposition at 417 °C (the temperature at the minimum of the endotherm).

The peak between T_g and the decomposition temperature represents the initial transition from soft plastic to liquid, a crucial phase that enables EPS molding in its molten state. This initial transition temperature is observed at 289.66 °C in the first recycling cycle, decreases to 274.2 °C in the third recycling cycle, and drastically drops to 204 °C in the fifth recycling cycle.

Flowability

The Melt Flow Index (MFI) represents the flowability of waste PET and waste EPS. The MFI data is derived from an average of 3–5 specimens for each recycling level variation.

As shown in Table 4 and Figure 4, the MFI values increase with a higher number of recycling cycles, suggesting that waste PET becomes more flowable as recycling progresses.

Table 4. MFI of recycled PET

Sample	PET virgin	PET-1	PET-3	PET-5
1	32	44.16	54	86.16
2	33	44.04	50.64	90.96
3	33	43.44	53.28	84.72
4	34	45.84	51	88.2
5	32	46.32	46.68	90.96
Avrg	32.8	44.76	51.12	88.2
SD	0.75	1.12	2.57	2.51

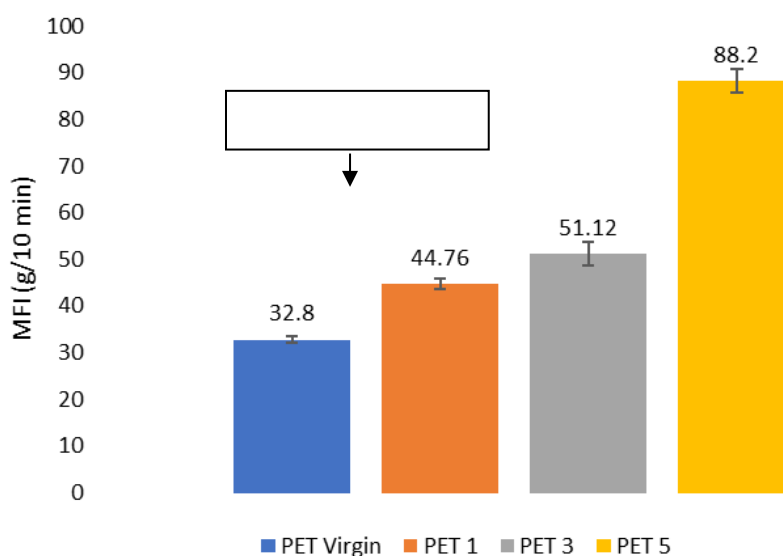


Figure 4. The increased flowability in recycled PET

The trend of increasing flowability is also observed in recycled EPS, where the MFI values rise with a higher number of recycling cycles, as illustrated in Table 5 and Figure 5.

Table 5. MFI of recycled EPS

Sample	EPS virgin	EPS-1	EPS-3	EPS-5
1	25	51.96	57.43	61.04
2	25	52.15	57.28	64.71
3	30	51.73	58.37	61.09
Avrg	26.7	51.9	57.69	62.3
SD	2.36	0.17	0.48	1.72

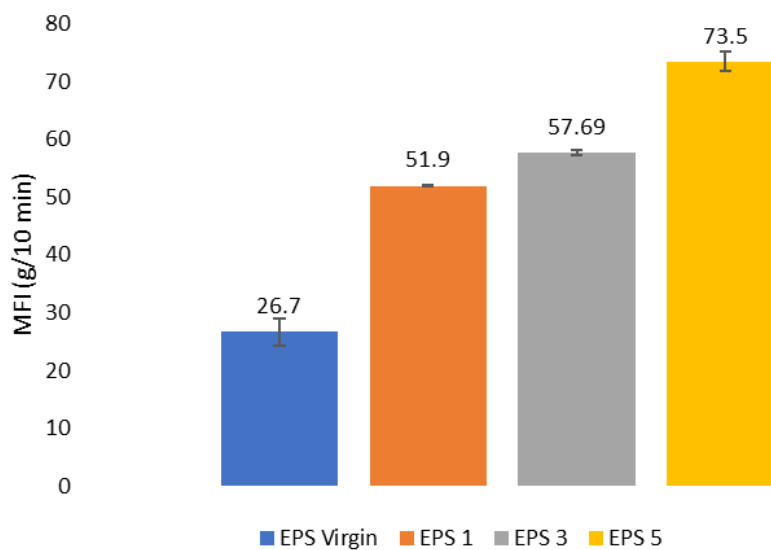


Figure 5. The MFI of recycled EPS

Repeated heating and cooling during recycling may lead to a decrease in the molecular weight of PET and EPS. The recycling process exposes the materials to elevated temperatures, and over multiple cycles, thermal degradation can occur (Masmoudi et al., 2018). The random chain scission caused by thermal degradation results in a reduction of molecular weight, which subsequently affects both melt viscosity and intrinsic viscosity (Cusano et al., 2023).

CONCLUSIONS AND RECOMMENDATIONS

In conclusion, this research provides quantitative insights into the thermal properties and flowability of PET and EPS plastics across multiple recycling cycles. The decline in PET crystallinity, subtle shifts in glass transition temperatures, and notable changes in flowability emphasize the measurable effects of recycling on material properties. For EPS, variations in glass transition temperatures across recycling cycles reflect changes in its thermal behaviour. Moreover, the lower transition temperature for initial softening before thermal decomposition in EPS-5 (204 °C) signifies a significant shift in material characteristics.

The study consistently demonstrates an increase in flowability for both PET and EPS with successive recycling cycles. These quantitative findings are crucial for optimizing recycling processes, maintaining product quality, and making informed decisions regarding product applications.

ADVANCED RESEARCH

Future research should delve deeper into the molecular-level mechanisms driving these thermal and rheological transformations in PET and EPS across multiple recycling cycles. Advanced spectroscopic techniques, such as FTIR and Raman spectroscopy, could elucidate chemical bond alterations, while DSC and TGA can further refine our understanding of degradation kinetics. Additionally, computational modeling and molecular dynamics simulations may provide predictive insights into polymer chain scission, crosslinking tendencies, and the impact of residual additives. Investigating the interplay between thermal history and mechanical performance will be essential for developing tailored reprocessing strategies, enhancing recycle performance, and expanding the applicability of recycled PET and EPS in high-performance applications.

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