UNIVERSITI MALAYSIA PERLIS

International Journal of Nanoelectronics and Materials

IJNeaM

ISSN 1985-5761 | E-ISSN 2232-1535

Melt Behavior of Soil-Buried Polypropylene/Polylactic Acid Blends Reinforced with Kenaf Particles

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Received 6 July 2025, Revised 14 August 2025, Accepted 8 September 2025

ABSTRACT

The increasing demand for sustainable materials has intensified research into the flow behavior of biodegradable polymer blends reinforced with natural fillers such as kenaf particles. Understanding their rheological performance, especially after environmental exposure like soil burial, is crucial for optimizing polymer processing and for recycling purposes. Thus, this study was set up to investigate the flow behavior of polypropylene (PP)/polylactic acid (PLA) blends filled with kenaf particles before and after the soil burial testing. The polymer blends were formulated with PP/PLA ratios of 90/10 and 70/30, incorporating 5 and 10 phr of kenaf particles. The compounding process was carried out using a heated two-roll mill at 190 °C. Meanwhile, the rheological behavior was evaluated using the melt flow index (MFI) and a capillary rheometer to examine the effects of soil degradation and aging on the prepared blends and composites. After 3 months of soil burial, the MFI of the 90/10 blend decreased from 2.23 to 2.01 g/min, suggesting the formation of crosslinked structures, possibly due to the presence of ethylene copolymer segments in the PP chains. In contrast, the 70/30/10 composite exhibited a substantial increase in MFI, from 3.34 to 5.50 g/10 min, indicating significant PLA degradation caused by polymer chain scission, which led to an increased fraction of low molecular weight PLA. A similar trend was observed in the apparent viscosity (η_{app}) of the 70/30/10 composite that showed a consistent decline ofter 3 months of soil degradation period. This was further supported by a reduction in extrudate die swell, confirming molecular weight loss due to chain scission during soil burial. Overall, the results suggest that a higher content of kenaf particles in PP/PLA blends accelerates the composites degradation under soil burial conditions. This is attributed to the water retention capability of incorporated kenaf particle, which promotes hydrolysis and consequently causing molecular weight reduction for the PLA chain during the degradation process.

Keywords: *Biodegradable polymers, polypropylene, polylactic acid, kenaf particles, soil burial, rheology, melt flow*

1. INTRODUCTION

The development of biodegradable polymer composites has gained significant attention due to the increasing demand for sustainable materials. Polyolefins such as polypropylene (PP) and polyethylene (PE) have been widely used due to their exceptional strength, toughness, and processability; however, their non-biodegradable nature poses significant challenges in waste management and raises serious concerns about long-term ecological impact. While polylactic acid (PLA) has the ability to yield high mechanical performance with environmental benefits, its inherent brittleness, low thermal stability, and limited impact resistance restrict its broader application in demanding engineering and packaging environments [1,2]. In the past decade, many industries have blended polypropylene (PP) with polylactic acid (PLA) to balance the overall polymer melt strength while introducing biodegradability to their target product [3,4,5]. In addition to the blending of polypropylene (PP) and polylactic acid (PLA), the incorporation of natural fillers such as kenaf particles plays a multifunctional role in enhancing the overall properties of the resulting biocomposites. Beyond contributing to improved biodegradability, kenaf serves as an effective reinforcing agent by providing mechanical stability and structural integrity to the polymer matrix. Moreover, the presence of kenaf particles influences the rheological behavior of the melt, acting as a viscosity modifier that can tailor the flow characteristics during processing. According to Irfan et al., the length of kenaf particles plays a crucial role in the extrusion process of kenaf-filled polypropylene composites [4]. These particles influence the viscosity of PLA blends by introducing interfacial interactions and altering the structural integrity of the polymer matrix [5,6].

Besides mechanical performance, the degradation of thermoplastic composites significantly impacts the product's physical appearance and molecular structure, leading to a reduction in molecular weight and alterations in flow characteristics. When PLA and PP composites filled with natural fillers undergo soil burial degradation, they experience substantial structural and chemical transformations due to microbial activity, moisture absorption, and prolonged environmental exposure. These degradation processes often lead to polymer chain scission, increased porosity, and polymer-filler detachment, which directly impact the material's viscosity and melt flow characteristics [7,8]. As the polymer matrix deteriorates, variations in viscosity and melt flow index (MFI) influence the ease of reprocessing and recyclability [9]. Atalay et al. previously reported that elevated temperature and humidity further accelerate the hydrolytic degradation of PLA, causing a dramatic reduction in the melted PLA complex viscosity [10].

As PP, PLA, and their blends are frequently disposed of in landfills, concerns regarding their environmental impact have become increasingly distressing; thus, understanding their flow behavior before and after the degradation is crucial for evaluating the polymer biodegradation potential and recyclability. Recent studies by Kwon et al. have revealed that enzymatic degradation notably influences the mechanical properties of PLA/PPC-based blends, with chain scission identified as the dominant degradation mechanism [11]. Besides, the rheological analysis at different degradation stages offers valuable information on molecular structural changes, enabling the formulation of strategies to enhance or control the stability of PLA-based composites. Such insights are vital for the development of sustainable, biodegradable polymer systems suitable for practical implementation across diverse industrial sectors [12]. In this study, the flow behavior of the soil burial of 90/10 and 70/30 PP/PLA blends reinforced with 5 and 10 phr kenaf particles was investigated. The purpose of the work is to gain a deeper understanding of the composite soil degradation pattern and behavior with the aim of tailoring end performance for targeted product applications.

2. MATERIALS AND METHODS

2.1 Materials

Polypropylene (PP) copolymer with ethylene (Pro-Fax SM240) with a melt flow index of 25g/10 min (2.16 kg at 230 °C) was obtained from Titan PP Polymer Malaysia. Polylactide PLA (Native Pultrusion Sdn. Bhd) with a density of about 1.21 g/cm^3, was dried in an oven for 24 hours (40 °C). The kenaf particle ranging from 500 nm to 50 µm was provided by Lembaga Kenaf dan Tembakau Negara, headquarters Malaysia.

2.2 Sample Preparation

The materials were premixed manually before mixing it using a heated two-roll mill (Guangdong Xihua Machinery Co. Ltd - RM8175A) to produce the compound based on the formulation stated in Table 1. All samples were compounded at 190 °C for 20 minutes. The samples were prepared by using a Gotech Hot-press (GT-014-H) machine at 195 °C for 8 minutes.

Table 1 Design formulations for PP/PLA blends and PP/PLA/kenaf composites

Samples		PP (wt %)	PLA (wt %)	Kenaf (phr)
PP/PLA blend				
1.	90/10	90	10	0
2.	70/30	70	30	0
PP/PLA/kenaf composite				
3.	90/10/5	90	10	5
4.	90/10/10	90	10	10
5.	70/30/5	70	30	5
6.	70/30/10	70	30	10

2.3 Testing and Characterisation

The specimen with a size of 125 mm x 12.7 mm x 3.2 mm was buried in peat soil with an organic carbon content (C) of 55 %, pH of 4.9 and a low salt solubility content of 0.04 %. The samples were buried at an open space area of Arau, Perlis, Malaysia, where the weather condition is such that the temperature and moisture fluctuate from 24 to 37 °C and 70 to 90 % for the duration of 3 months. The melt flow index (MFI - GT-7100-MI) test was executed at 190 °C based on the modified ASTM D1238-90b. The die diameter and length were 2.095 and 8 mm, respectively. The preheat time was regulated for 3 minutes and the calculated flow rate is by gram per 10 minutes based on the cut extrudate interval. The extrudate swell was measured using a calliper after the extrudate collected from the MFI was fully solidified. The calculation of die swell is using the following Equation 1.

$$B = \frac{D_e}{D_d} \tag{1}$$

Next, the rheological characteristics of the selected compound were attained by using the single-bore capillary rheometer with the LCR7001 series from Dynisco, Malaysia. A capillary die with a diameter of 1.5 mm and L/D of 12 was used for the test and the data was plotted without further correction. The power law model was used to describe the relationship between shear stress, apparent shear rate, and flow index n (non-Newtonian index), and it is expressed in Equation 2. The viscosity (η) is obtained by dividing the shear stress (τ) over the shear rate (γ). K is the viscosity index, while n is the non-Newtonian index, which refers to the fluidity of a polymer in a viscous flow state.

$$\eta(\gamma) = K\gamma^{n-1} \quad (2)$$

3. RESULTS AND DISCUSSION

The melt flow index (MFI) data presented in Figure 1 illustrates the changes in the flow properties of PP/PLA blends and PP/PLA/Kenaf composites for over 3 months of soil burial period. MFI is a key indicator of polymer degradation, as it reflects molecular weight changes due to chain scission or crosslinking. Initially, the 70/30 blend and its composites generally have higher MFI values compared to the 90/10 blend and composites before the soil burial testing. This is attributed to the higher content of PLA low melt strength inside the blend or composite compared to PP. Thus, the higher the amount of PLA content will directly contributes to an increased in the compound flowability. Meanwhile, the incorporation of kenaf particle of 5 and 10 phr in the PP/PLA blends slightly reduce the MFI value with respect to the same amount of 70/30 blend. This is due to the kenaf–matrix interactions, which create hindrance within the polymer matrix and consequently reduce the composite MFI. Thus, the higher the kenaf loading, the lower the MFI value of the composites [13].

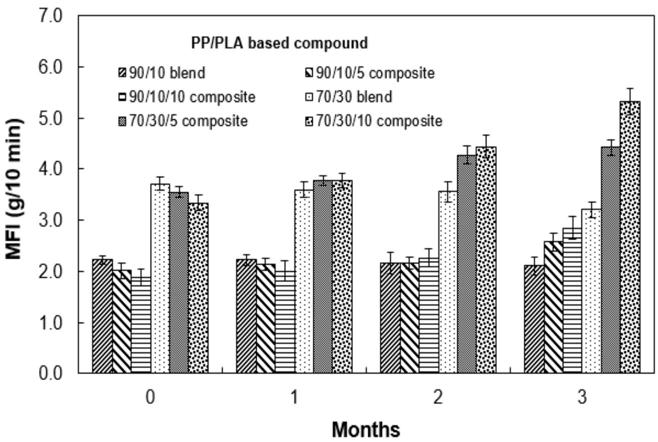

Figure 1. MFI for PP/PLA blends and PP/PLA/kenaf composites before and after 3 months of soil burial testing.

After the 3 months of soil burial degradation, the 90/10 blend only indicate a steadily decrease from 2.23 to 2.01 g/10min after 1 month which were later express a subtle increase in MFI value after 3 months of soil burial test. The reason is primarily due to the crosslinking phenomenon at the early stage of the soil burial test where the attached ethylene component or copolymer on PP chains, as discussed in our previous findings [14,15,8]. As the burial duration progresses, the crosslinking was tune to chain scission mode. As for the 70/30 blend, it is noticed the blend exhibit a prompt increase in MFI value after the soil burial testing. This is caused by PLA's is more susceptible to hydrolysis in the soil environment, causing it to degrade more rapidly under soil burial conditions. According to Pang et al., outdoor soil burial results in a greater degradation effect compared to indoor conditions for their thermoplastic starch (TPS)/PP blends [16].

As the soil burial duration progresses, the 70/30 composites with 10 phr kenaf particle content shows a rapid increase in MFI value from 3.3 g/10 min to 5.11 g/10 min indicating faster degradation rate with respect to 5 phr kenaf loading composite. This signify that the presence of kenaf particles accelerates the degradation rate of the composites. The effect is more pronounced because at higher kenaf content, more moisture absorption site will be embedded in the PP/PLA blend simultaneously promotes microbial interaction especially for the PLA component leading to its chain breakdown [14,17]. Hence, the MFI results suggest that the addition of a higher amount of kenaf particles accelerates PLA chain scission, thereby reducing the molecular weight of the entire composite system. This trend aligns with the observations of Chee et al. in their study on soil-buried kenaf/bamboo/epoxy hybrid composites, where the increased in kenaf content give rise to a decline in tensile properties due to epoxy chain scission primarily caused by hydrolysis [13].

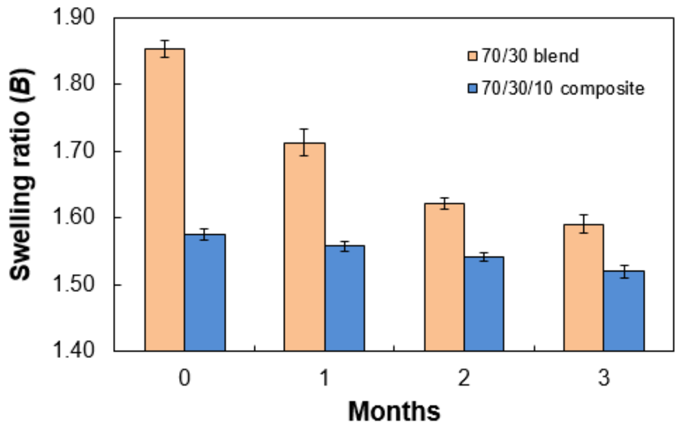

Figure 2. MFI extrudate swelling ratio for PP/PLA blends and PP/PLA/kenaf composites before and after 3 months of soil burial testing.

Figure 2 presents the die swelling ratio of the MFI extrudate, while Figure 3 shows the appearance of the swollen extrudate for the 70/30 blend and 70/30/10 composite over a period of three months. It can be seen that the swelling was more extreme for the 70/30 blend compared to the 70/30/10 composite. In the absence of fillers, the polymer matrix chains experience a chain "memory effect" upon exiting the narrow MFI die. The 70/30 blend initially exhibited the highest swelling ratio of 1.87, reflecting the viscoelastic behavior of the blended polymer melt [19,20]. As the burial duration increased, a clear decreasing trend in the swelling ratio of the extrudate was observed. This reduction suggests that hydrolytic degradation of PLA, possibly coupled with microbial activity, led to chain scission, thereby reducing polymer elasticity and die swell. This implies that the shortening of the PLA chains over time slows down the elastic recovery of the polymer, consistent with the reduction in molecular weight due to the hydrolysis-prone nature of PLA.

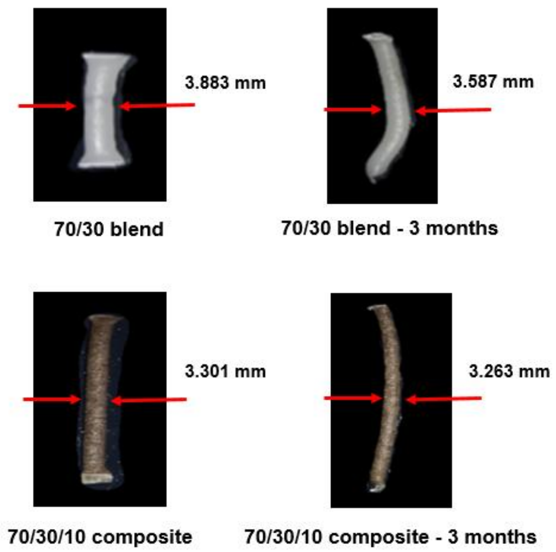

Figure 3. MFI extrudate swell appearance for PP/PLA blends and PP/PLA/kenaf composites before and after 3 months of soil burial testing.

Unlike the 70/30 blend, the 70/30/10 composite showed a lower die swell ratio of 1.58 with the diameter of 3.301 mm prior to soil burial. This reduction is likely due to the presence of kenaf particles, which restrict the system polymer chain mobility consequently limit further excessive swelling during extrusion [18]. After the soil burial test, the diameter of the composite extrudate only shows a slight decrease in value from 3.301 mm to 3.184 mm, indicating a smaller degradation effect with respect to the 70/30 blend. The kenaf particles has acted as a reinforcing phase, helping to retain structural integrity despite ongoing degradation [7,19].

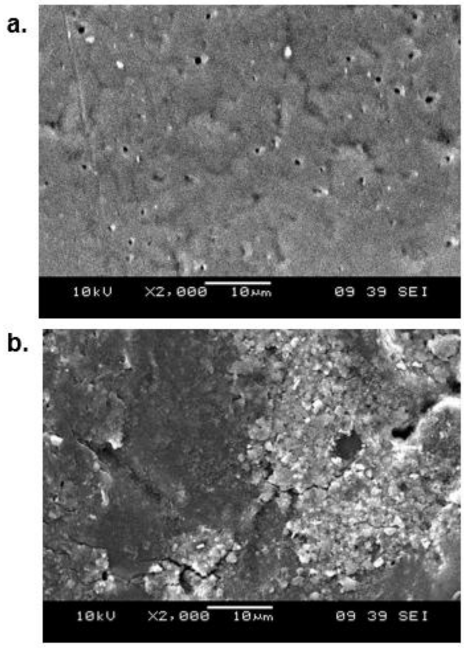

Figure 4. SEM images for MFI extrudate surface for a) 70/30 PP/PLA composite and b) 70/30 PP/PLA/kenaf composites after 3 months of soil burial testing.

Meanwhile, Figure 4 shows the SEM images of the MFI extrudate surface of the 70/30/10 composite before and after soil burial testing. Although the extrudate surface appeared visually unchanged at the macro level after 3 months of soil burial degradation, microscopic analysis revealed significant changes in the composite surface roughness and morphology. Evidence of delamination and poor interfacial adhesion between PP and PLA also became apparent. This is due to the biodegradable and hydrolytically unstable nature of PLA, especially in the presence of moisture, heat, or microorganisms. In contrast, PP is more chemically resistant and degrades at a much slower rate [17]. After 3 months, this mismatch in degradation behavior leads to increasing incompatibility, resulting in the loss of surface contact and weaker interfacial adhesion between the PP and PLA phases even in the melted stage.

Next, Figure 5 presents the apparent shear stress (τ_{app}) vs. apparent shear rate ($\dot{\gamma}_{app}$) curves for the 70/30 blend and 70/30/10 composite, both before and after 3 months of soil burial degradation. The term "apparent" is used because no corrections, such as entrance pressure loss or non-Newtonian flow adjustments, were applied to the rheological data collected from the capillary rheometer for the blend and composite system. From the graph, it can be clearly observed that all samples display a shear-thinning behaviour which indicates that the melts behave similar to thermoplastic non-Newtonian flow properties [14,20]. Further observation, the 70/30 blend exhibits a slight reduction in τ_{app} following the 3 months soil burial period. This finding supports the earlier discussed MFI results, indicating that the hydrolysis of PLA has causes an obvious chain scission phenomenon, leading to a decrease in the PLA molecular weight and consequently, a reduction in the blended system τ_{app} required to initiate the flow. Previously, similar behavior was reported by Kara and Molnar for their moisture-degraded stereoregular PLA systems [21].

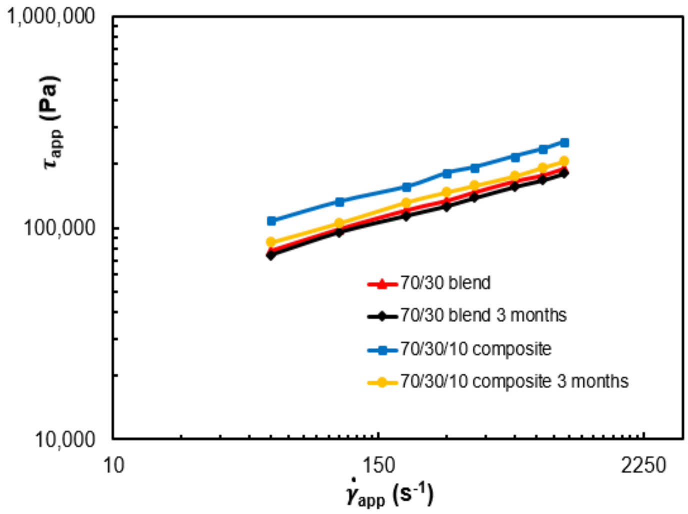

Figure 5. τ_{app} vs. $\dot{\gamma}_{app}$ of 70/30 blend and 70/30/10 composite before and after 3 months of soil burial testing.

As for the 70/30/10 composites, the incorporation of kenaf particles into the PP/PLA blend has resulted in an increase in the τ_{app}, indicating enhanced resistance to flow and deformation in the molten state. This τ_{app} improvement suggests that kenaf particles act as reinforcing fillers, effectively restricting the mobility of the polymer chains and increasing the melt viscosity due to kenaf particle retard the polymer chain to slide among each other within the composite. However, after 3 months of soil burial testing, a significant reduction in τ_{app} is observed in the 70/30/10 composite. Similarly, this decline is primarily attributed to the substantial degradation of the PLA component, which undergoes hydrolytic chain scission over time. The degradation effect becomes increasingly pronounced as the interfacial contact area between the kenaf fibers and the PLA/PP matrices expands, accelerating the breakdown process [2,3].

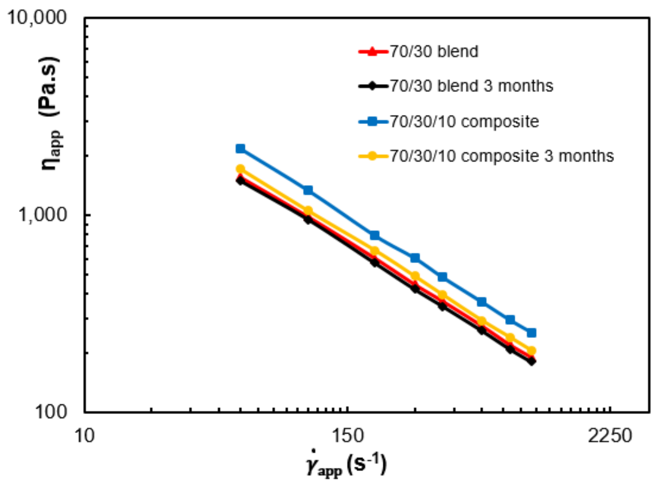

Figure 6. η_{app} vs. $\dot{\gamma}_{app}$ of 70/30 blend and 70/30/10 composite before and after 3 months of soil burial testing.

Figure 6 illustrates that both the blend and composite system exhibit a shear-thinning behavior, where the apparent viscosity (η_{app}) continuously decreases as the $\dot{\gamma}_{app}$ increases. This behavior refers to the characteristic of non-Newtonian fluids, in which the melt molecular alignment at higher $\dot{\gamma}_{app}$ substantially reduces both melted polymer chains internal resistance to flow. Among the two compound, the 70/30/10 composite shows a significantly higher η_{app} compared to the 70/30 blend, suggesting that the incorporation of kenaf particles enhances the melt's resistance to flow. This is attributed to stronger polymer-filler interactions and restricted chain mobility caused by the dispersed kenaf fibers as discussed earlier [9]. A parallel trend was reported by Xian et al., who observed an increase in η_{app} in a polypropylene-based composite system reinforced with kenaf and sago particles [8]. After 3 months of soil burial degradation, the 70/30/10 composite exhibits a slight reduction in ηapp. This decrease is primarily due to the hydrolytic degradation of the PLA, leading to partial detachment of filler particles. These changes weaken the interfacial bonding between the matrix and the filler, reduce molecular entanglement, and ultimately improve the flowability of the degraded composite melt [19,22].

4. CONCLUSION

The melt behavior of soil-buried PP/PLA blends reinforced with kenaf particles reveals critical insights into their rheological degradation under soil burial conditions. The soil burial degradation test confirms that PP/PLA blends and PP/PLA/kenaf composites degrade over time, with the rate of degradation influenced by the PLA content and the presence and amount of kenaf particles. Higher PLA content and kenaf particle loading accelerate the degradation rate for both blends and composites, as reflected in the rising MFI values over the 3 months of soil degradation testing. The reduction in MFI extrudate diameter and the appearance of surface irregularities after soil burial further confirm the degradation of both the 70/30 blend and the 70/30/10 composite. The 70/30 blend experiences extreme dimensional changes, while the 70/30/10 composite retains better structural integrity due to the reinforcing effect of kenaf particles. The shear-thinning behavior observed in all samples indicates their non-Newtonian flow properties. Additionally, the decline in η_{app} after soil burial degradation confirms that kenaf particles incorporation accelerates the aging process in the 70/30/10 composites. These rheological findings are essential for understanding processability, recyclability, and structural stability of biodegradable composites. The insights gained support the rational design of bio-filled thermoplastics with predictable melt behavior tailored for sustainable, application-specific uses.

ACKNOWLEDGMENTS

We would like to thank Mr Mohd Faisal Abd Rahim and Mr. Mohd Aizat Abdul Rahman for their technical assistance while conducting the research work

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