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Utilization of Epoxidized Palm Oil as A Plasticizer in Synthetic Leather

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Abstract. Substituting petroleum-based plasticizers in the synthetic leather industry with renewable resources, such as epoxidized palm oil (EPO), will mitigate environmental pollution. This study aims to produce synthetic leather using EPO and compare its characteristics to those of synthetic leather using commercial plasticizers. Raw materials were compounded, coated on embossed paper, covered with reinforcing fabric, and cured in an oven at 180 °C for 1 minute. Control leather (CoL) was produced using diisononyl phthalate (DINP) and epoxidized soybean oil (ESBO), A leather (AL) was produced using DINP and EPO, B leather (BL) was produced only using EPO, and C leather (CL) was produced using EPO and ESBO. Synthetic leather was characterized for mechanical properties (SNI 1294:2009), glossiness (gloss meter), surface morphology (digital microscope), thermal properties (DSC), and functional groups (FTIR). The synthetic leather characteristics show that the thickness of AL and CL was close to CoL. The longitudinal tensile strength of BL (186.52 N) was not significantly different from CoL (191.41 N) and AL (190.06 N), whereas its transversal tensile strength (44.13 N) was not significantly different from CL (42.37 N). Both the longitudinal and transversal tear strengths of BL were not significantly different from those of AL. Furthermore, the glossiness of BL (4.37 GU) was not significantly different from CoL (4.22 GU) and AL (4.32 GU). Unlike CoL, EPO produces a smooth surface morphology (in AL, BL, CL) and eliminates pinhole defects. CoL, AL, BL, and CL exhibit similar functional groups. DSC analysis shows that BL exhibits thermal stability identical to CoL, AL, and CL. Our findings indicate that EPO could be a promising plasticizer for producing synthetic leather.

Keywords: Epoxydized Palm Oil, Plasticizer, Synthetic Leather

INTRODUCTION

Phthalic acid and its esters, derived from petroleum, are another widely used industrial plasticizer (Harrison and Ounaies, 2002). However, due to the lack of covalent bonding between phthalates and the polymer matrix, their industrial application poses significant environmental risks. Phthalates can leach out the polymer matrix during manufacturing

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processes, including synthetic leather production, releasing them into the environment. Consequently, phthalates have been detected as pollutants across various environmental media, including the atmosphere, water, soil, and sediments (Chen *et al*., 2019; Das *et al*., 2021).

Plasticizers play a crucial role in synthetic leather production by enhancing the flexibility, durability, and processability of polyvinyl chloride (PVC)-based materials. The presence of plasticizers reduces the brittleness of PVC, allowing it to mimic the texture and mechanical properties of natural leather. However, the choice of plasticizer significantly influences synthetic leather’s environmental footprint and performance.

Epoxidized soybean oil (ESBO) is currently the most commonly used renewable vegetable-based plasticizer due to its non-toxic, biodegradable nature and stability under heat and light conditions, making it environmentally friendly (Afrillia, 2024). However, as a major producer of palm oil rather than soybean oil, Indonesia has significant potential to develop palm oil-derived epoxy oil as an alternative plasticizer. Indonesia produced more than half of the world's palm oil in 2022, with exports consisting of 2% crude palm oil (CPO) and 4% crude palm kernel oil (CPKO), while downstream products, including refinery outputs, saw increases of 73% and 21%, respectively (InfoSAWIT, 2024). Given this context, developingpalm oil-based epoxy oil as a plasticizer represents a promising opportunity for Indonesia's export market (Lim, et al., 2015; Jalil *et al*., 2019; Ramli *et al*. 2021).

Epoxidized palm oil (EPO) is produced through the epoxidation of unsaturated fatty acids in palm oil. This process involves the reaction of palm olein ester with an oxygen donor, such as peracetic acid or performic acid, leading to the formation of oxirane rings within the fatty acid chains. These oxirane rings improve the plasticizing efficiency of EPO, allowing it to serve as an effective substitute for conventional plasticizers, including ESBO (Utami *et al*., 2024). The presence of epoxide groups enhances the compatibility of EPO with PVC, contributing to improved thermal stability and mechanical properties.

Palm oil fatty acids contain approximately 45% monounsaturated fatty acids (oleic acid) and about 10% polyunsaturated fatty acids (linoleic acid). Research has been conducted on plasticizers derived from oleic acid for application in polyvinyl chloride (PVC) (Triwulandari and Haryono, 2007; Waskitoaji *et al*., 2012). These studies primarily focused on optimizing synthesis process parameters (such as temperature, time, and catalyst percentage) and evaluating the characteristics of palm ester plasticizers and their impact on synthetic leather's mechanical properties, including modulus of elasticity and elongation at break. Additionally, research has explored using palm oil-based alkyd as a plasticizer in synthetic leather (PVC) (Jalil *et al*., 2019). The influence of EPO as a plasticizer on the thermal and mechanical properties of poly (hydroxybutyrate-co-valerate)/ poly (caprolactone) blends has also been investigated (Ramli *et al*., 2021). However, studies specifically investigating the use of EPO as a plasticizer in synthetic leather production and comparative studies on the performance of EPO and other commercial plasticizers on synthetic leather characteristics have not yet been conducted.

This study aims to investigate the influence of EPO on the characteristics of synthetic leather, particularly when combined with a commercial petroleum-based plasticizer (diisononyl phthalate, DINP), a commercial vegetable-based plasticizer (ESBO), and in the absence of both DINP and ESBO. The characteristics examined in this study include physical properties (glossiness), mechanical properties (thickness, tensile strength, and tear strength), functional group composition, thermal properties, and surface morphology.

METHODOLOGY

Materials

Resin Polyvinyl chloride (PVC) K-value 74 and K-value 67 were obtained from Kaneka Paste Polymers Sdn. Bhd. Trimethyl Pentanyl Diisobutyrate (TXIB), DINP, ESBO, and Stabilizer LBZ were obtained from PT Sempurnaindah Multinusantara. Calcium carbonate ($CaCO_3$) was obtained from PT. Graha Jaya Pratama Kinerja. EPO was obtained from lab-scale self-production following the method of Utami *et al.*, 2024.

Synthetic Leather Production

Four types of synthetic leather were produced in this study: control leather (CoL), leather A (AL), leather B (BL), and leather C (CL). The CoL was formulated according to the synthetic leather standard formula using diisononyl phthalate (DINP) and epoxidized soybean oil (ESBO). AL, BL, and CL were formulated using the same amount of plasticizer as in CoL. Still, a portion was replaced with EPO, considering the viscosity of the plasticizer and plastisol mixtures. The AL was formulated using DINP and EPO, while the BL utilized only EPO. The CL was formulated using a combination of EPO and ESBO.

The coating technique used in synthetic leather production. The raw materials were weighed according to the design formulas in Table 1 for the top coat and Table 2 for the base coat. The base coat was the primary layer that provides adhesion to the substrate and contributes to the material's mechanical strength, flexibility, and overall durability. Meanwhile, the top coat was a protective layer that enhances surface properties such as abrasion resistance, weatherability, and aesthetic appeal.

Table 1. Synthetic leather top coat formula

Materials	**Quantity (phr*)**			
	CoL	**AL**	**BL**	**CL**
Resin Polyvinyl chloride (PVC) K-value 74	30	30	30	30
Resin PVC K-value 67	70	70	70	70
DINP	55	27.5	0	0
ESBO	10	0	0	10
EPO	0	37.5	65	55
Trimethyl Pentanyl Diisobutyrate (TXIB)	1.5	1.5	1.5	1.5
Stabilizer LBZ	2.5	2.5	2.5	2.5
Calcium carbonate ($CaCO_3$)	25	25	25	25

*phr (per hundred resin)

Table 2. Synthetic leather base coat formula

Materials	**Quantity (phr*)**
Resin PVC K-value 74	50
Resin PVC K-value 67	50
DINP	50
$CaCO_3$	25

*phr (per hundred resin)

The top and base coat raw materials were mixed separately using a stirrer at 500 rpm for 30 minutes until a homogeneous compound was obtained. The embossed paper was coated with a 0.5 mm thick top coat compound and then cured at 180 °C for 1 minute. The top coat compound was coated with a 0.1 mm thick base coat compound and covered with a reinforcing fabric (Spandex-type). It was then cured at 180 °C for 1 minute. Finally, the embossed paper is removed from the synthetic leather. The sample preparation scheme and coating process are presented in Fig. **1**: Sample preparation illustration and Fig. **2**, respectively.

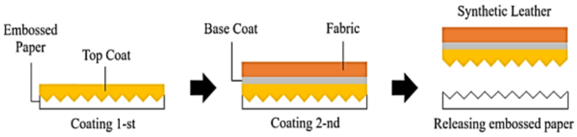


Fig. 1: Sample preparation illustration

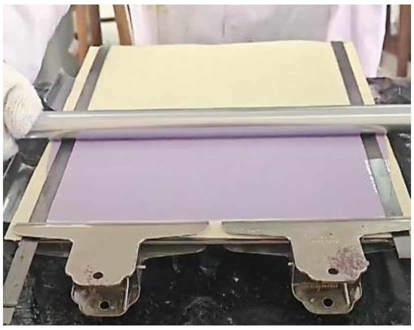


Fig. 2: Coating process

Synthetic Leather Characterization

The measurement of synthetic leather's thickness, tensile strength, and tear resistance refers to the Indonesian National Standard (SNI) 1294-2009 (SNI 1294:2009). The thickness of the synthetic leather was measured using a digital thickness gauge Syntek TGH25 (Taiwan). Synthetic leather's tensile strength and tear resistance were measured using a universal testing machine (UTM) ETM-D (China). The glossiness level of the synthetic leather surface was measured using a portable gloss meter Landtek GM-6 (China), and the surface morphology of the synthetic leather was observed using a digital microscope CMOS 1600X (China). Thermal characteristics and functional groups of synthetic leather were identified using Differential scanning calorimetry (DSC) 4000 PerkinElmer (United States of America) and Fourier-transform infrared (FTIR) spectroscopy Frontier C96600 PerkinElmer (United States of America), respectively.

RESULTS AND DISCUSSION

The formulation was adapted from the reference manufacturer's established formulation. The study investigated the influence of EPO on synthetic leather characteristics, specifically when combined with DINP (primary plasticizer)-AL, ESBO (secondary plasticizer)-CL, and without the addition of DINP and ESBO-BL. The subsequent sub-chapters describe the attributes exhibited by synthetic leather when made using different plasticizer formulas.

Functional Groups

The functional group analysis of synthetic leather is displayed as FTIR spectra in Figure 3, with each peak's identification listed in Table 3. The FTIR method is typically used to examine synthetic leather's carbon-chlorine bonds in PVC resin and the plasticizer groups (such as carbonyl for esters and epoxy for oxidized oils).

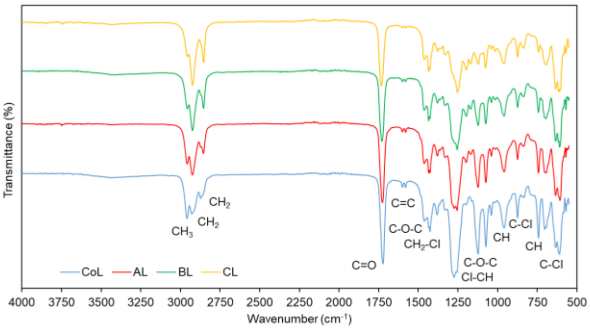


Fig. 3: FTIR spektra of synthetic leather

The FTIR spectra of PVC plastisol in synthetic leather revealed distinct peaks corresponding to various molecular bonds. These include stretching of C-H (CH_2 and CH_3) bonds at 2855 cm^{-1}, 2930 cm^{-1}, and 2960 cm^{-1}; angular deformation of CH_2-Cl bonds at 1426 cm^{-1}; deformation of CH_3 and CH_2 groups at 1380 cm^{-1} and 1335 cm^{-1}; out-of-plane angular deformation of Cl-CH bonds at

1255 cm^{-1}; out-of-plane trans deformation of C-H bonds at 960 cm^{-1}; and stretching of C-Cl bonds at 835 cm^{-1}, 694 cm^{-1}, and 635 cm^{-1}. The observed peaks align with the data reported in the literature for PVC films, specifically the findings of Coltro (2013).

Table 3. Identification of functional group of synthetic leather

Wavenumber (cm^{-1})	Types of vibration	Functional group (Coltro, 2013)
2960	Asymmetric stretching	CH_3
2930	Asymmetric stretching	CH_2
2855	Symmetric stretching	CH_2
1730	Stretching	C=O
1600	Stretching	C=C aromatic
1580	Stretching	C=C aromatic
1462	Stretching	C–O–C
1426	Angular deformation	CH_2–Cl
1380	Symmetric angular deformation	CH_3
1335	Angular deformation	CH_2
1255	Out of plane angular deformation	Cl–CH
1195	Stretching	C–O–C
1118	In plane angular deformation	C–H aromatic
1074	In plane angular deformation	C–H or C–C aromatic
960	Out of plane trans deformation	C–H
835	Stretching	C–Cl
732	Out of plane angular deformation	C–H aromatic 1,4 bi-substituted
694	Stretching	C–Cl
635	Stretching	C–Cl

The FTIR spectra of the synthetic leather exhibited distinctive peaks associated with plasticizers, such as C-H out-of-plane angular deformation at 732 cm^{-1}, C-H aromatic vibrations at 1074 cm^{-1}, and 1118 cm^{-1}, C-O-C stretching at 1462 cm^{-1} and 1195 cm^{-1}, and C=O stretching at 1730 cm^{-1}, 1600 cm^{-1}, and 1580 cm^{-1}. These peaks correspond to the results given by Coltro (2013). The FTIR spectra of CoL, AL, BL, and CL exhibited identical peaks while the absorption intensities varied. The absorption intensity at 1195 cm^{-1} in AL, BL, and CL is greater than in CoL. This is because AL, BL, and CL contain more epoxy rings (C-O-C stretching) due to using additional epoxy plasticizers, as shown in Table 1. In contrast, the absorption intensities at 732 cm^{-1}, 1074 cm^{-1}, and 1118 cm^{-1} are reduced in AL, BL, and CL compared to CoL, possibly because there is a higher rate of plasticizer evaporation from AL, BL, and CL. Different types of epoxy oil, like ESBO, have lower boiling points than DINP (440 °C). The boiling point of ESBO and EPO is 150 °C (Ataman Kimya, n.d.) and 204-207 °C (Abd Maurad *et al.*, 2017), respectively. This means epoxy oil evaporates more quickly when the leather plastisol cures at 180 °C. In addition, the CoL sample contains a larger concentration of DINP, which has not yet evaporated at the curing temperature. This increases absorption intensities at wavelengths of 732 cm^{-1}, 1074 cm^{-1}, and 1118 cm^{-1}.

Thermal Characteristics

The thermal properties of synthetic leather are crucial for predicting its durability under heat exposure. This research uses differential scanning calorimetry (DSC) to study these properties, focusing on the glass transition point (*Tg*) and the decomposition point. As shown in Fig. **4**, the DSC curve indicates the decomposition point.

Two endothermal peaks show processes that need heat in the DSC curves from 230 °C to 350 °C. Usually, decomposition processes happen at the same time. These curves correspond to the plasticizer's decomposition and the resin's dehydrochlorination reaction (Dalagnol *et al.*, 2013). The first peak corresponds to the plasticizer, while the second and third peaks, if present, are related to the PVC resin (Marcilla and Beltrán, 1996). The polymer's molecular weight impacts the broad endothermic peak of the dehydrochlorination process. Polymers with a lower molecular weight break down at lower temperatures, whereas those with a higher molecular weight break down at higher temperatures.

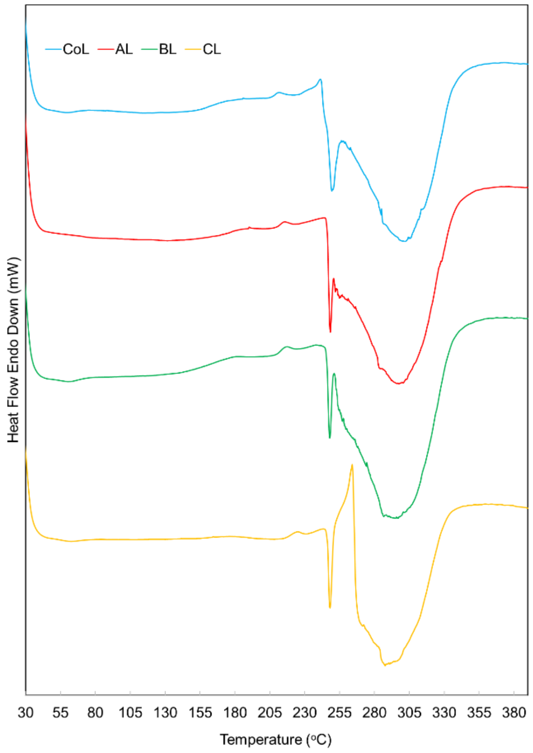


Fig. 4: DSC curve of synthetic leather

For CoL using commercial plasticizers, two peak degradation points are observed at 248 °C and 298 °C. Interestingly, AL, BL, and CL show similar values, suggesting that EPO provides equally excellent thermal stability. However, the CL curve displays a gap between the first and second peaks, potentially due to compatibility issues between the resin and plasticizer.

The primary goal of adding plasticizers is to lower the glass transition temperature (*Tg*) of rigid PVC resins (Alobad and Alraheem, 2018), allowing them to be used as flexible materials like synthetic leather (Dalagnol *et al.*, 2022). The DSC results for *Tg* are presented in Fig. **5**.

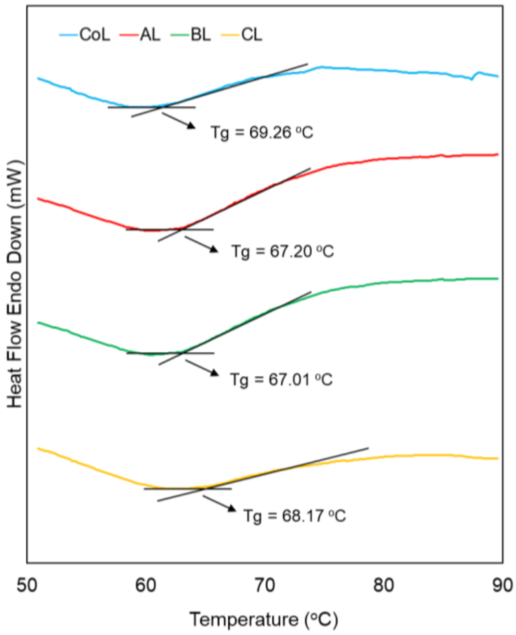


Fig. 5: Transition glass temperature of synthetic leather

Fig. **5** shows that the *Tg* value of EPO is slightly lower than that of CoL, with BL showing the lowest temperature. Plasticizers act as spacers between polymer chains, reducing molecular interaction and increasing molecular mobility (Dalagnol *et al.*, 2013). The Tg value can be used to estimate a plasticizer's effectiveness: A greater decrease in *Tg* indicates better performance.

Physical and Mechanical Characteristics

The characteristics of the synthetic leather, as shown in Table 4, indicate that the thickness of AL and CL did not differ significantly from the reference leather or the control (CoL). In the CL formulation (Table 1), EPO was used to replace DINP in the control leather formulation. The thickness test results for CL demonstrate that EPO effectively substitutes DINP as a plasticizer in CoL, which aligns with previous studies showing that epoxidized oils contribute to film formation and stability in polymeric materials (Utami *et al.*, 2024). This is also in line with the study by Al-Mulla *et al.* (2010), who found that EPO

Table 4. Physical and mechanical characteristics of synthetic leather

Characteristics	CoL	AL	BL	CL
Thickness (mm)	0.6984 ± 0.0276^{a}	0.6615 ± 0.0047^{a,b}	0.6323 ± 0.0251^{b}	0.6857 ± 0.0439^{a,b}
Longitudinal tensile strength (N/mm^2)	191.4100 ± 0.0172^{a}	190,0567 ± 2.1517^{a}	186.5179 ± 1.7639^{a}	164.3587 ± 5.8903^{b}
Transversal tensile strength (N/mm^2)	57.2790 ± 0.0182^{b}	104.3600 ± 8.1650^{a}	44.1310 ± 1.3663^{c}	42.3691 ± 0.022^{c}
Longitudinal tear strength (N/mm)	11.3800 ± 0.0166^{a}	7.4566 ± 0.3934^{b}	7.0592 ± 0.3927^{b}	7.2565 ± 0.5826^{b}
Transversal tear strength (N/mm)	6.6766 ± 0.0202^{c}	9.5551 ± 0.5629^{b}	10.0690 ± 0.4529^{b}	16.4320 ± 1.7623^{a}
Glossiness (GU)	4.2166 ± 0.1258^{a}	4.3166 ± 0.1528^{a}	4.3733 ± 0.1419^{a}	3.8900 ± 0.1153^{b}

Note: Different statistical notations indicate significant differences

improved the flexibility and dimensional stability of the plasticized polymer. In the AL formulation, the combination of EPO and DINP results in a leather thickness that remains statistically similar to CoL, suggesting that partial substitution of DINP with EPO does not compromise the matrix integrity of the PVC resin.

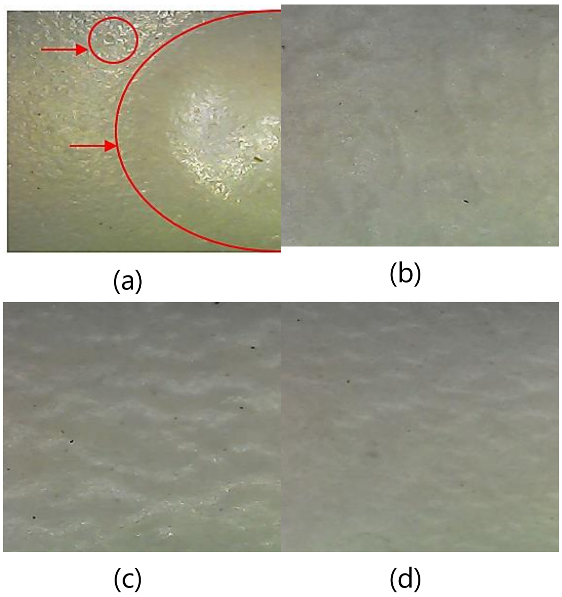
The AL and BL's longitudinal tensile strength values were not significantly different from CoL, indicating that EPO can maintain the tensile integrity of synthetic leather even when completely replacing DINP and ESBO. However, the transversal tensile strength of BL and CL was lower than CoL, possibly due to differences in intermolecular interactions between EPO and PVC compared to conventional plasticizers. EPO molecules, with their epoxide functional groups, introduce polarity that may influence the plasticizing efficiency and stress distribution under tensile load. This phenomenon is consistent with findings reported by other researchers, who noted that epoxidized oils, while effective plasticizers, may slightly alter polymer network flexibility depending on concentration and blending conditions (Chieng *et al.*, 2016).

The longitudinal tear strength of AL, BL, and CL exhibited no significant differences; however, all three displayed markedly lower values compared to CoL. This suggests that while EPO provides effective plasticization, its molecular structure and interaction with PVC might result in different stress distribution patterns that affect tear resistance. Conversely, the transversal tear strength of AL, BL, and CL was higher than CoL, implying that EPO contributes to improved resistance to tear propagation in the transversal direction. This may be due to the ability of EPO to enhance the energy dissipation mechanism during the tearing process, as Sarwono *et al.*, 2012 reported in their study on epoxy blends modified with EPO.

According to the glossiness evaluation, the leather samples AL (4.32 GU) and BL (4.37 GU) exhibited glossiness values statistically comparable to CoL (4.22 GU). EPO appears to produce a smoother surface morphology, eliminating pinhole defects commonly observed in CoL. This observation aligns with previous reports that bio-based plasticizers can enhance surface uniformity in polymer coatings (Utami *et al.,* 2024). The improved smoothness in AL, BL, and CL suggests that EPO may contribute to better plasticizer-PVC compatibility, leading to a more homogeneous surface finish.

Surface Morphology

The surface morphology of synthetic leather samples was examined using a digital microscope at 1600x magnification, as depicted in Fig. **6**. The control leather (CoL), which utilized diisononyl phthalate (DINP) as a plasticizer, exhibited noticeable pinhole defects on its surface. In contrast, samples incorporating EPO, specifically AL (DINP and EPO), BL (EPO only), and CL (EPO and epoxidized soybean oil, ESBO), demonstrated smoother surfaces devoid of such defects.



(a) (b)

(c) (d)

Fig. 6: Surface morphology of synthetic leather (a) CoL, (b) AL, (c) BL, and (d) CL

The absence of pinhole defects in EPO-containing samples suggests that EPO contributes to a more uniform and defect-free surface morphology in synthetic leather. This improvement can be attributed to the molecular interactions facilitated by the oxirane rings in EPO, which enhance compatibility with polymer matrices. Sarwono *et al.* (2012) reported that blending EPO with epoxy resin resulted in a miscible blend, improving morphological properties due to effective molecular interactions between EPO and the resin matrix.

Furthermore, the smoother surface morphology observed in EPO-plasticized synthetic leather may be linked to the plasticizing effect of EPO, which enhances the flexibility and processability of the polymer matrix. Al-Mulla *et al. (*2010) found that incorporating EPO as a plasticizer in polylactic acid (PLA) improved mechanical properties and resulted in a more homogeneous morphology, indicating better dispersion and interaction within the polymer matrix.

Additionally, Hasnan *et al.* (2020) demonstrated that coatings formulated with EPO exhibited superior adhesion performance and smoother surfaces compared to those using epoxidized cooking oil. This finding underscores the significance of oxirane content in achieving desirable surface characteristics, as the higher oxirane content in EPO enhances crosslinking density and uniformity in the coating matrix.

In summary, incorporating EPO as a plasticizer in synthetic leather formulations leads to a smoother and more uniform surface morphology, likely due to enhanced molecular interactions and plasticizing effects. These findings align with existing literature, highlighting the potential of EPO to improve the quality and performance of synthetic leather products.

CONCLUSION

The utilization of EPO-DINP and EPO-ESBO as combination plasticizers produces comparable thicknesses in synthetic leather, with tensile and tear strengths of single-use EPO showing no significant differences compared to controls and DINP combinations. At the same time, glossiness remains statistically similar across all variants. EPO enhances synthetic leather by achieving a smooth surface morphology and eliminating pinhole defects while exhibiting similar functional groups with varied intensities in the plasticizer peaks; additionally, single-use EPO demonstrates thermal stability comparable to control synthetic leather and combination plasticizers, albeit with a marginally lower Tg suggesting improved performance.

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