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Development and Characterization of Recycled PET-Based Adhesive Films for Supporting Insulin Injection Site Rotation

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Abstract

Insulin injections are vital for diabetes management, but repeated injections at the same site may cause lipohypertrophy (LH), leading to poor glycemic control and inconsistent insulin absorption. Since proper site rotation is often difficult for patients to maintain, this study proposes a recycled poly (ethylene terephthalate) (PET)-based adhesive film, fabricated from plastic bottles using the Thermal Induced Phase Separation (TIPS) method, to support injection site rotation. Poly (vinyl alcohol) (PVA) was incorporated to enhance the film's adhesive properties. The film's performance was evaluated through mechanical testing, thermogravimetric analysis (TGA), and pH measurement, showing favorable results. TIPS was employed for film fabrication. The film demonstrated appropriate pH (5.4 ± 0.1), tensile strength (3.78 MPa), and good thermal stability (220°C), retaining structural integrity after UV sterilization. TGA showed weight loss of 86% at 480°C, suggesting significant mass loss associated with plasticizer evaporation and polymer degradation. Adhesion testing showed sufficient tackiness, with balls stopping at 1.5–2.0 cm on the 4 cm film, showing good adhesion. The film also remained stable after UV sterilization, supporting its safety and durability. Characterization of pH, tensile strength, and tackiness was performed in triplicate (n = 3). Repurposing PET helps reduce plastic waste while offering a cost-effective material for healthcare use. The adhesive film may serve as a visual aid for insulin injection site rotation, with potential to reduce the risk of lipohypertrophy and improve insulin absorption. These findings are preliminary and provide a basis for more comprehensive future study.

1. Introduction

Individuals with diabetes will inject insulin subcutaneously to lower blood sugar levels. Repeatedly injecting insulin at the same sites can lead to the development of a palpable, tumor-like lump of adipose tissue known as lipohypertrophy (LH) (Arda Sürücü & Okur Arslan, 2018). Lipohypertrophy is a common complication in people with diabetes who use subcutaneous insulin injections. Its aetiology is thought to be multifactorial, with contributing factors including repeated injections at the same site, improper injection techniques, needle reuse, poor rotation of injection sites, and inconsistent depth and angle of injections (Huang et al., 2023; Hirsch et al., 2014). Studies have reported that the prevalence of lipohypertrophy in individuals with diabetes who use insulin injections ranges from 15% to 50% (Blanco et al., 2013; Deng et al., 2017). This condition can have significant clinical implications, including poor glycemic control, increased insulin dosage requirements, unpredictable insulin absorption, hypoglycemic episodes, and a reduced quality of life for affected individuals (Mader et al., 2024). Current management of insulin injection–related lipohypertrophy relies primarily on avoidance of affected areas, single-use needles, and regular site inspection rather than a dedicated therapeutic device. Preventing lipohypertrophy involves educating patients and healthcare professionals on proper injection techniques, consistent rotation of injection sites, and regular monitoring of glycemic control. Patient education programs, individualized treatment plans, and close follow-up with healthcare providers have shown promise in reducing the risk of lipohypertrophy (Huang et al., 2023; Kalra et al., 2016; Cunningham & Mckenna, 2013).

Research conducted by Deeb et al. (2019) highlighted the importance of proper insulin injection technique in maximising the absorption of insulin and its subsequent impact on glucose levels in the body. Adhering to the recommended injection guidelines can significantly improve the effectiveness of insulin therapy and contribute to better management of blood sugar. To mitigate the risk of tissue stress, Hirsch and Strauss (2019) propose maintaining a minimum distance of 1 cm, approximately the size of an adult finger, between injection sites. Alternatively, when injecting in the buttocks or thighs, dividing the site into quadrants or halves and systematically rotating through these areas in a consistent direction, such as clockwise, can help prevent localised tissue damage. By following these recommended practices, individuals with diabetes can optimise the absorption of insulin, minimise the likelihood of lipohypertrophy, and enhance their overall diabetes management.

Ensuring proper insulin injection technique is essential for optimal diabetes management. However, some diabetic patients face challenges remembering their last injection site, leading to the development of unfavorable habits and behaviors. Injection site rotation is a crucial aspect of insulin therapy for individuals with diabetes. According to Bahendeka et al. (2019), this practice involves changing the location of insulin injections to different sites on the body to prevent the development of lipohypertrophy, minimize tissue damage, and optimize insulin absorption.

In this study, an adhesive film was developed to facilitate insulin injection site rotation. The film was fabricated via the Thermal Induced Phase Separation (TIPS) method using recycled poly (ethylene terephthalate) (PET) derived from waste plastic bottles. Poly (vinyl alcohol) was incorporated as both a plasticizer and adhesive due to its excellent film-forming properties and adhesion to skin and soft tissues (Vineeth et al., 2021; Kahvand & Fasihi, 2019; Khan et al., 2016). The prepared film was characterized and evaluated to determine its performance and functional properties. The development of such a material may contribute to a more user-friendly and supportive approach for diabetes management. This work also demonstrates the potential of converting discarded plastic bottles into functional films, thereby addressing environmental concerns while promoting sustainable resource utilization. Furthermore, the adhesive polymeric film may serve as a visual reference for insulin injection site rotation, helping to minimize the risk of repeated injections at the same site. Since this manuscript reports preliminary findings, further detailed studies, particularly on biocompatibility and dermatological safety, are necessary before clinical implementation.

2. Materials and Methods

2.1 Materials

PET was collected from recycled material, dimethyl sulfoxide (DMSO) was sourced from Merck USA, polyvinyl alcohol (PVA)(85k mw, hydrolysis (97%) was obtained from Sigma-Aldrich USA, and glycerin.

2.2 Preparation of Adhesive Film Via Thermally Induced Phase Separation (TIPS)

The adhesive PET polymer film was fabricated through thermally induced phase separation (TIPS). Recycled PET bottles were first ground into small particles. The ground PET flakes were washed with deionized water to remove surface debris, followed by an acetone rinse to eliminate residual adhesives and organic contaminants. Specifically, 5 g of PET was mixed with 40 mL of DMSO and heated at 195°C for 5 hours to create a homogeneous solution. Then, 2 g of PVA was dissolved in 20 mL of DMSO and added to the PET solution. The mixture was further heated at 195°C for 20 minutes. After achieving homogeneity, 2 mL of glycerin was added to the mixture. It was

J. of Science and Technology Vol. 18 No. 1 (2026) p. 58-68

then cooled in an ice bath for 30 minutes. The film was dried in an oven overnight at 40°C and subsequently rolled into a thin adhesive film. The thickness of the produced films was measured using a digital micrometer.

2.3 Thermal Analysis

The thermal stability analysis was conducted using a Shimadzu TGA 50 thermogravimetric analyzer (TGA) on the prepared adhesive film sample, ranging from 30°C to 800°C, with a temperature rate of 15°C/min and a flow rate of 50 mL/min.

2.4 Fourier Transform Infrared Spectroscopy (FTIR)

The film sample was characterized by using FTIR Spectrometer Frontier from Perkin Elmer for compound identification to identify the functional group of the film. The film cut into pieces and compressed into a thin pellet with potassium bromide (KBr) powder. The spectrums were recorded between 4000 cm^{-1} to 400 cm^{-1}.

2.5 Tensile test

Tensile tests of the developed film were performed using a Universal Testing Machine (UTM) with a gauge length of 50.00 mm at a tension speed of 10 mm/min. The tests were repeated (n = 3) to ensure reproducible results.

2.6 pH Test

pH tests were performed to assess the film under two distinct conditions: (1) in a melted state, (2) in contact with simulated wet skin, and (3) in contact with simulated sweaty skin. The pH was determined using a Mettler Toledo digital pH meter. For the melted condition, the pH electrode was submerged directly into the sample. For the wet and sweaty hand conditions, the measurement procedure was adapted from Oh et al. (2018). Briefly, the film was applied to a wet and sweaty hand for 5 minutes. A flat-surface pH probe was then placed in direct, firm contact with the wetted film surface. All measurements for each condition were performed in independent triplicates (n = 3), and the results were recorded as average values.

2.7 Rolling Ball Tack Test

The films were subjected to the rolling ball tack test modified from Bhomick (2014) to study their adhesive properties. Steel balls of varying weights were permitted to roll down from a specific starting point. These balls were inclined at an angle of 10°, which was selected for this study in relation to the base of the test platform. The speed of the moving balls was maintained at a constant rate. The adhesive film was cut into squares measuring 4 cm × 4 cm and placed on the base platform. The distance traveled by the balls before adhering to the film was recorded. The sketch of the method was shown in Figure 1 The tests were repeated (n = 3) to ensure reproducible results.

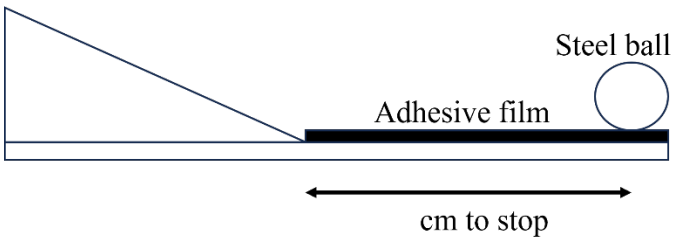

Fig. 1 *The rolling ball tack test*

2.8 UV-Sterilization

The developed film underwent UV sterilization, with the sample being exposed to UV (wavelength: 340nm, intensity: 1.55W/m^2) light for 30 minutes. The physical properties of the film were assessed both immediately after the UV exposure and again after one month to evaluate the effects of UV sterilization.

2.9 Water-Stripping Method

The developed film was applied to the dry and sweaty backs of the hand for one hour and then peeled off. The condition of the film was evaluated. This adhesive testing method was adapted from Hansen et al. (2020) with modifications.

3. Results and Discussion

3.1 Fabrication of Adhesive PET Film via TIPS

PET sources were obtained from waste plastic bottles, serving as the polymer base for this study. PET plastic bottle waste holds potential as a film material due to its capacity to generate thin films or layers, offering a viable solution for film production (Pulido et al., 2019). This approach also addresses environmental contamination caused by the increasing accumulation of non-degradable plastic bottle waste. Although PET contains hydrophilic functional groups, it shows promise for achieving high water flux in films (Ali et al., 2022). Mulyati et al. (2018) suggest that utilizing waste-derived polymers can reduce the reliance on commercial polymers and lower film manufacturing costs. This not only decreases production expenses but also promotes eco-friendly technology aimed at minimizing pollution. DMSO was selected as the solvent due to its ability to fully dissolve PET. It has strong affinity for both polar and nonpolar substances and its miscibility with a wide range of organic solvents including water (Reuter, 2017). This characteristic allows DMSO to penetrate and disrupt the hydrogen bonding and other intermolecular forces within the PET structure, leading to the dissolution of the polymer in the solvent. The solubility of PET in DMSO makes it a suitable option for various applications where PET needs to be dissolved or processed.

The initial pure PET film exhibited inadequate mechanical strength, brittleness, and poor adhesion, making it unsuitable for skin application (Pünnel & Lunter, 2021). To enhance its uniformity, flexibility, and adhesive properties, the PET was blended with polyvinyl alcohol (PVA) to create a composite membrane (Ferreira et al., 2020; Kusumawati et al., 2018). PVA was selected because it is a water-soluble, highly flexible, biocompatible, and completely biodegradable polymer (Abral et al., 2020). During formulation, glycerol was added to the heated PET-PVA blend. Consistent with the thermally induced phase separation (TIPS) technique, glycerol functioned as an antisolvent to induce film precipitation (Riewe et al., 2020) while further improving the film's adhesive properties. The mixture was subsequently solidified in an ice-water bath, resulting in the final adhesive film illustrated in Figure 2.

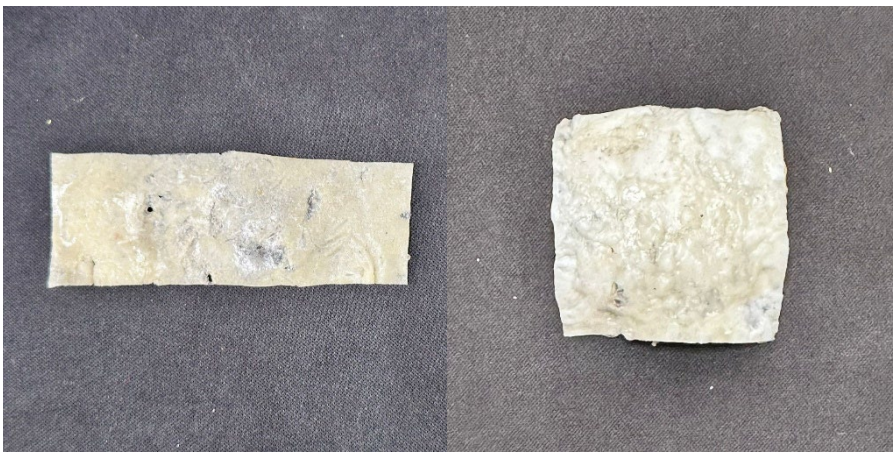

Fig. 2 *The adhesive film after solidified*

3.2 Characterization of the Newly Developed Adhesive PET Film

Figure 3 illustrates the FTIR spectra of pure PET, PVA, glycerin, and the formulated adhesive PET film, with the characteristic absorption peaks and their respective shifts summarized in Table 1. A strong and broad band was observed in the film at 3280 cm^{-1}, corresponding to the O–H stretching vibration. This indicates the presence of abundant hydroxyl groups from both PVA originally at 3283 cm^{-1} and glycerin originally at 3280 cm^{-1} (Kharazmi et al., 2015). The composite film also exhibits a distinct merged band at 2941 cm^{-1}, attributed to the C–H stretching vibrations contributed by all the incorporated organic components. Furthermore, the spectrum displays characteristic PET functional groups, including a prominent C=O ester stretch shifted to 1717 cm^{-1} from 1713 cm^{-1} in pure PET, C–O stretching for the aromatic ester shifted to 1260.62 cm^{-1} from 1240.75 cm^{-1}, and a band at 721 cm^{-1} corresponding to the aromatic C–H out-of-plane bending of the benzene ring (Pereira et al., 2017).

J. of Science and Technology Vol. 18 No. 1 (2026) p. 58-68

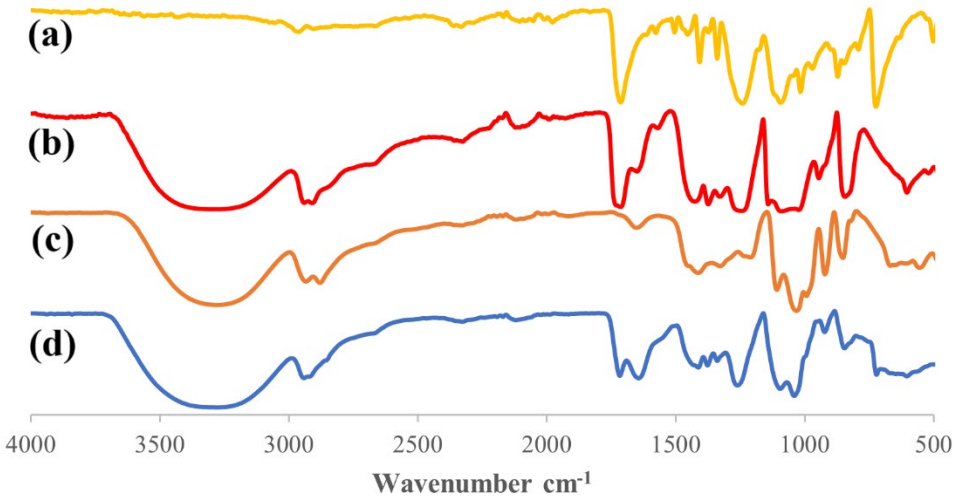

Fig. 3 *FTIR spectroscopy of (a) PET; (b) PVA; (c) Glycerine; and (d) Film*

Table 1 *Characteristic FTIR bands of PET, PVA, glycerin, and the prepared adhesive film*

Functional group	PET (cm^{-1})	PVA (cm^{-1})	Glycerin (cm^{-1})	Film (cm^{-1})
O–H stretching	-	3283	3280	3280
C–H stretching	2963	2909	2933	2941
C=O stretching	1713	1713	-	1717
C–O stretching	1241	1089	1031	1261
Aromatic C–H bending	724	-	-	721

According to Qian et al. (2015), when multiple substances are blended, the characteristic spectral peaks can indicate whether the mixture is purely physical or if chemical interactions have occurred. In this study, the characteristic peaks of PET, PVA, and glycerin were all observed in the composite film. In this study, peaks representing PVA and glycerin were observed, and slight changes in the band indicated interactions between the incorporated polymers, possibly due to non-covalent bonds. Thus, as noted by Mahmoud et al. (2014), this interaction of polymers serves as a crucial strategy for developing novel polymer films with uniquely tailored characteristics.

TGA was used to investigate the thermal decomposition of the polymer blend and determine its thermal stability. As shown in Figure 4, the film exhibited multi-stage weight loss with increasing temperature from the initial 30°C. In the first stage (a), the initial weight loss is attributed to the evaporation of absorbed moisture and the depletion of the glycerol plasticizer, which volatilizes as temperatures approach its boiling point of approximately 290°C. In this formulation, glycerol acts as the plasticizer, comprising roughly 26.5% of the film's initial mass. According to Klähn et al. (2019), plasticizers and polymers interact via external and internal plasticization. In external plasticization, the plasticizer does not form primary chemical bonds with the polymer chains but adheres through a dipole–dipole attraction. Consequently, these plasticizers are susceptible to loss through evaporation at elevated temperatures. As the plasticizer is depleted, the remaining polymer matrix typically becomes brittle, leading to a reduction in mechanical properties (Eslami et al., 2023).

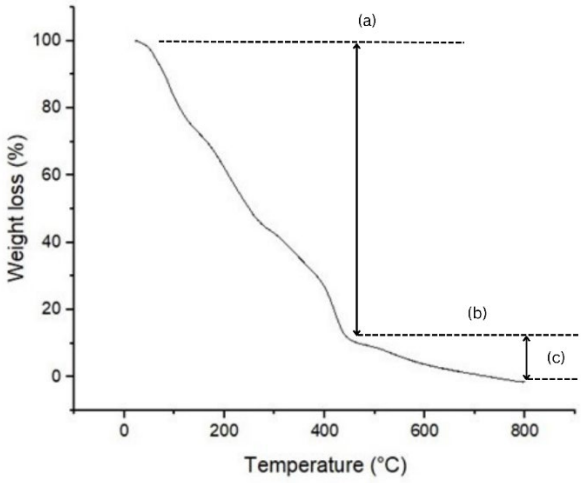

Fig. 4 *Thermogravimetric analysis (TGA) of the newly adhesive film*

The second stage (b) is characterized by major thermal decomposition of the polymer components, resulting in an 86% cumulative weight loss by 480°C. This significant mass reduction is not merely plasticizer loss but represents the primary chain scission and degradation of the polymer backbones. Within this temperature range, the PVA component decomposes, and the PET polymer undergoes its primary thermal degradation. As noted by Mert Kılınç et al. (2015), PET begins to thermally degrade at temperatures above 340°C, with Das and Tiwari (2019) demonstrating that major structural breakdown and weight loss of PET occur around 400°C. Finally, in the third stage (c), further weight reduction occurs due to the thermal breakdown and combustion of residual carbon char remaining from the polymer decomposition (Kim et al., 2021). Overall, the film exhibited major structural degradation after exposure to 480°C and lost complete thermal stability by 780°C. While the addition of PVA and glycerol modifies the film's properties, the TGA thermogram confirms that the blend follows the expected high-temperature degradation profile of its constituent polymers.

A tensile test was conducted to evaluate the tensile strength of the newly developed adhesive film using a UTM. The film fractured under stress at 3.78 MPa, with a maximum elongation or break displacement of 38 mm. The test results are presented in the Table 2 and Figure 5. For comparison, recent studies on biogels skin adhesive films report that tensile strengths ranging from approximately 1 to 3 MPa are highly comparable to the natural tensile strength of human skin, making them ideal for epidermal applications (Li et al., 2025). Therefore, the observed tensile strength of 3.78 MPa indicates that our preliminary formulation possesses an excellent level of mechanical properties which are strong enough to resist rupture during handling, yet sufficiently compliant to mimic and move with human skin.

Table 2 *The result of tensile test*

Thickness (mm)	Break Force (MPa)	Break Displacement (mm)	Maximum Force (N)
1.00	3.78	38	4.66

As reported by Wahyuningtyas and Dinata (2018), the addition of glycerine as a plasticizer increased the elongation at break and resulted in the highest tensile strength and percentage elongation. In this study, the adhesive film also contained glycerine as a plasticizer, which played a crucial role in enhancing the film's tensile strength. Consequently, the increase in elongation at the point of break is not solely attributed to the film's thickness but is also influenced by the chemical composition of the film.

J. of Science and Technology Vol. 18 No. 1 (2026) p. 58-68

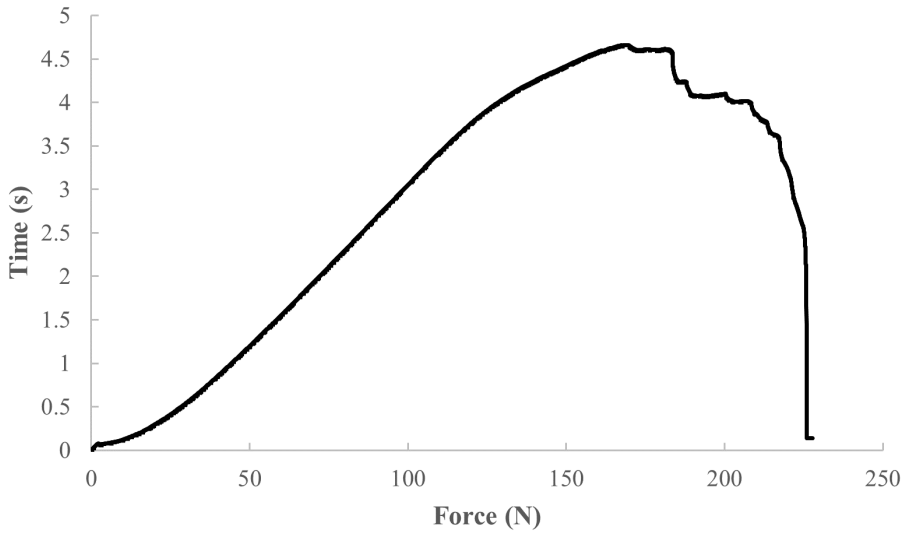

Fig. 5 *Time-force graph of the adhesive PET film*

Glycerine was instrumental in improving the film's ability to withstand tensile forces, with its interaction with other components contributing to the film's overall mechanical properties, including better flexibility and elongation at the point of break. However, under normal conditions, when the film was adhered to the skin and subsequently peeled off, neither film exhibited breakage, even after multiple applications. Both films demonstrated resilience without any signs of structural damage or deterioration. Therefore, the film exhibits adequate mechanical strength for preliminary skin application testing, making it well-suited for skin application.

It should be noted that the current study serves as a preliminary evaluation of the developed formulation's feasibility. Consequently, testing was conducted on a single, standardized film thickness to establish baseline adhesion, removal, and ultimate tensile properties. Comprehensive quantitative characterization, including the assessment of varying film thicknesses and detailed stress-strain profiling, will be the focus of future studies to fully optimize the film for commercial on-skin application

The skin's acidic pH acts as a protective barrier against harmful microorganisms and helps maintain a healthy skin environment (Baker et al., 2023). Therefore, it is important that the pH level of any product applied to the skin aligns with this range to prevent potential skin irritation and ensure skin efficacy and comfort. In this study, when the film was applied to the skin as outlined in the procedure, it exhibited a pH of 5.4 (± 0.1). According to Pünnel and Lunter (2021), human skin typically has a pH value of approximately 5. The adhesive film produced in this study was suitable for skin application, as its pH value did not cause harm. Blaak and Staib (2018) note that skin care products with a pH that deviates from this range can disrupt the skin's natural balance, potentially leading to irritation, dryness, or other undesirable reactions. Therefore, a formulation designed to be pH-compatible with the skin is more likely to be well-tolerated, enhancing overall effectiveness and promoting a positive user experience.

Furthermore, research by Zhang et al. (2023) on on-skin adhesive hydrogels demonstrated that a mildly acidic environment enhances adhesive performance, yielding higher interfacial shear and tensile strengths at pH 4.5 compared to pH 7.5. Because the adhesive film developed in this study exhibits a comparable pH, it is anticipated to possess similarly enhanced adhesive characteristics. This suggests that the film is not only chemically safe for skin contact but also holds strong potential for effective on-skin application.

The distance where the ball stopped at the adhesive film on the platform base was recorded after rolling down from the slope as shown in Table 3. This testing was conducted to measure the tackiness and adhesive strength of a material (States, 2017). The rolling ball tack test and probe tack test are widely employed as highly quantitative methods for adhesive tack measurement (Jucienė & Vaida Dobilaitė, 2021). According to Moriwaki et al. (2020), tack is a viscoelastic property that gauges the speed of bonding during the attachment and detachment process between a viscoelastic sample and a substrate. They stated the shorter the distance travelled by the ball, the higher was the tackiness of the adhesive coating.

Table 3 *The distance of metal ball stopped on the adhesive film*

Type of Ball (g)	Distance of metal ball stopped adhesive film (cm)
Havier (13.86)	1.5 (± 0.4)
Lighter (4.15)	2 (± 0.3)

In correlation with this study, although at different weight of metal balls; 13.86 g (heavier) and 4.15 g (lighter), the balls still can adhere on the film by stopping at 1.5 cm and 2 cm respectively of the whole 4 cm length of adhesive film respectively from slope or starting points. From the result, the produced adhesive film was capable of stopping the ball from moving. According to the previous study of Chu et al. (2020), the diffusion of small molecules such as tackifier from the adhesive into the film helped the adhesive to wet and quickly grip the substrate in this case was metal ball. Therefore, the adhesive film could give its tackiness properties which can be applied on diabetic patients' skin as a ruler injection site rotation.

The film was subjected to UV exposure for sterilization and to study the effect of UV sterilization on the physical properties of the adhesive film. The sterilizing effect of gamma irradiation has been extensively studied over the years. According to Yemmireddy et al. (2022), sterilization effectively eliminates bacterial growth on various surfaces. In this test, UV irradiation, part of the electromagnetic spectrum ranging from 200 to 400 nm, was used as the sterilization method. After one month of UV exposure, the results showed no changes in the physical properties of the film. The structure remained the same as before, with no visible physical degradation observed that could be harmful to skin application. Additionally, the adhesive film remained intact and did not rupture. This UV exposure is crucial for eliminating factors or environmental conditions, such as humidity, that can promote bacterial growth, particularly in the pharmaceutical industry (Tapia-Guerrero et al., 2020).

After one hour of wearing the film, it was observed that the film on dry skin retained its adhesive properties, allowing it to peel off easily even after one hour. In contrast, the film could not adhere to sweaty skin. The differences in peel forces were attributed to the interface between the substrate (skin) and the adhesive. A study conducted by Hansen et al. (2020) on the skin-adhesive interface found that the smooth spreading and effective bonding of the adhesive, intended to adhere securely to the skin, were limited by the presence of sweat. Similarly, Maciejewski et al. (2023) reported that the adhesive's ability to flow smoothly was reduced due to lower viscosity under sweaty conditions. Additionally, the adhesive film showed a decrease in adhesion after being applied to dry skin for five days, without deliberate peeling. This test was repeated three times under the respective skin conditions.

4. Conclusion

In conclusion, a recycled PET-based adhesive film was successfully developed as a potential material for supporting insulin injection site rotation. The film, prepared via the Thermal Induced Phase Separation (TIPS) method and modified with PVA and glycerol, showed acceptable preliminary physicochemical and adhesive properties, including skin-compatible pH, adequate adhesion, and stability after UV exposure. These findings indicate that the prepared film has potential as a practical material for further development. In addition, the use of recycled PET offers an environmentally sustainable approach for producing value-added healthcare materials from plastic waste. However, this study represents only a preliminary assessment of the proposed film. Since no in vivo evaluation, cytotoxicity analysis, or detailed dermatological safety testing was conducted, its clinical applicability cannot yet be established. Further studies are therefore required to optimize the formulation and to comprehensively evaluate its biocompatibility, safety, and functional performance before practical or clinical use can be considered.

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Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *Mohamad Wafiuddin Ismail, Azmir Ahmad, Author Mohd Zamani Zulkifli;* ***data collection:*** *Mohamad Wafiuddin Ismail, Siti Nor Akhirah Syahrial, Md. Uwaisulqarni Osman;* ***analysis and interpretation of results:*** *Mohamad Wafiuddin Ismail, Siti Nor Akhirah Syahrial, Azmir Ahmad;* ***draft manuscript preparation:*** *Mohamad Wafiuddin Ismail, Siti Nor Akhirah Syahrial, Md. Uwaisulqarni Osman. All authors reviewed the results and approved the final version of the manuscript.*

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