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Investigation of Electrical and Mechanical Properties of PLA/Exuviae Biocomposites for High-Voltage Insulation

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Abstract

This paper discusses the development of a new biocomposite made of polylactic acid (PLA) reinforced with black soldier fly (BSF) exuviae to improve the electrical insulation performance of high-voltage (HV) applications. Fourier transform infrared (FTIR) analysis was carried out to establish the type of chemical interaction between the PLA and the BSF exuviae. The electrical behaviour of the composite was measured in terms ofdirect current (DC) breakdown voltage, and mechanical strength was measured in terms of tensile strength. FTIR analysis confirmed the successful loading of exuviae into the PLA matrix, and new absorption bands were observed at amide I and II vibrations and the lipid C=O stretching, which is indicative of a high level of chemical interactions. Electrical testing showed that DC breakdown strength decreased greatly with exuviae content, especially at high filler loadings, and this was attributed to the effects of agglomeration. Mechanical testing revealed a distinct reduction in tensile strength and Young's modulus with increasing exuviae content from 0% to 3%, which means the exuviae content reduced the load-bearing capacity with increased filler content. The results are useful in understanding the electro-mechanical behaviour of PLA/BSF exuviae composites and their potential and drawbacks as a sustainable material to be used in HV insulation.

1. Introduction

Polylactic acid (PLA) has become one of the most promising bio-based and biodegradable polymers in the search for sustainable alternatives to insulation materials. Made from renewable resources such as corn starch or sugarcane [1], PLA has a much more favorable environmental profile than fossil-based plastics. PLA also makes a surprisingly good electrical insulator. It has been demonstrated that its volume resistivity and dielectric constant are similar to those of cross-linked polyethylene (XLPE), which is a commonly used power cable material [2]. In addition, it has been reported to have a mean impulse breakdown strength about 1.3 times that of XLPE, and it

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does not generate as much space charge under direct current (DC) stress. PLA is frequently used for 3D printing of medical devices and composite scaffolds. Orthopedic implants, tissue engineering, wound healing, drug delivery, and filtration are some of the applications of PLA-based nanocomposites, which have superior stability, structure, and mechanical properties in biomedical applications [3, 4].

Nevertheless, these promising electrical characteristics are marred by a number of important drawbacks that, at present, prevent their application in long-service-life high-voltage (HV) applications. The worst of these is that it is not durable, especially with regard to its degradability. As noted in [1], damage to PLA coating, which was manifested by a progressive loss of integrity after 12 days in Ringer solution at 37 °C, was probably due to interaction with the corroding magnesium substrate. The process of degradation is highly enhanced by high temperature, humidity, and pH, and results in a quick loss of molecular weight and mechanical integrity. This may result in a weakened protective effect within a comparatively short time in environments that are pertinent to the operation of HV equipment.

Besides its unfavorable hydrolytic stability, PLA has a number of disadvantages. It has a low heat deflection temperature and a low melting point, which means that it cannot be used in applications that produce a lot of heat [3]. Mechanically, it is a brittle material that has low elongation at break and, therefore, its toughness and impact resistance are limited. Although some of these properties can be enhanced by the addition of additives and fillers, these additions usually have their own trade-offs, and the inherent problem of hydrolytic instability is a significant impediment [1].

This critical evaluation shows that there is a core performance gap in the area of insulating polymers. Although the most promising bio-based polymer, PLA, is sustainable, it does not have the durability needed to be used in long-term HV applications [3]. The gap promotes the development of a novel composite material, which combines PLA with bio-based fillers such as exuviae, the naturally shed exoskeletons of insects such as the black soldier fly (BSF). Exuviae, as a functional filler with high sustainability, can improve the mechanical and electrical properties of PLA and can address the weaknesses of the material itself, thus improving its overall performance as an insulating material. The analysis indicates, with great likelihood, that none of the neat polymers are sufficient, and a composite solution is the way to go, with PLA as a sustainable matrix and exuviae as the reinforcement and electrical property improvement, filling the gap between environmental friendliness and durability [5, 6].

BSF is an insect species that has attracted a lot of attention due to its outstanding capacity to transform low-value organic waste into high-value biomass, specifically protein and fat [7]. This renders BSF farming one of the pillars of new circular economy systems, which offers a sustainable supply of animal feed and other products such as biodiesel, bioplastics, and biofertilizers [8]. The BSF life cycle involves the pupa shedding its outer shell, which is known as pupal exuviae, to become an adult fly. These exuviae are among the main by-products of BSF farming, and puparium (pupal shells) have been reported to have high extraction yields [7, 9].

Instead of being discarded, this research proposes to valorize this waste stream as a high-performance filler, given that BSF exuviae are rich in chitin, a structural biopolymer [9]. It was reported in [10] that chitin obtained from BSF was 9% in the pupae exuviae and 23% in the imago. This makes BSF a highly sustainable and ecologically sound alternative to conventional chitin sources, which are primarily derived from marine crustaceans like shrimp and crab shells, the harvesting of which carries its own environmental concerns. Thus, the use of BSF exuviae shown in Figure 1 aligns perfectly with the goal of developing advanced materials from renewable, waste-derived resources.



Fig. 1 *BSF exuviae*

Int. Journal of Integrated Engineering Vol. 18 No. 7 (2026) p. 123-134

The potential of BSF exuviae as a functional filler extends beyond its sustainability. Its chemical composition, refined by evolution to serve as a protective exoskeleton, endows the material with unique structural, mechanical, and electrical properties that make it an attractive reinforcement material. The primary structural component of BSF exuviae is chitin, the second-most-abundant natural polysaccharide on Earth after cellulose [7]. Chitin is a linear polymer composed of repeating β-(1,4)-linked N-acetyl-D-glucosamine units [9]. In insects and crustaceans, the substance predominantly exists as α-chitin, a semi-crystalline form characterized by polymer chains arranged in an antiparallel fashion. This arrangement promotes strong intermolecular hydrogen bonding, resulting in a rigid and stable structure. This inherent rigidity and strength, which provide mechanical protection for the developing insect, suggest that chitin can serve as an effective mechanical reinforcement when incorporated into a polymer matrix, potentially improving the hardness and tensile properties of a brittle matrix like PLA.

From an electrical standpoint, chitin is far from being an inert filler. Since chitin is a natural polysaccharide [7], it is chemically similar to other lignocellulosic bio-fillers like cellulose and lignin, which contain abundant free hydroxyl groups that impart polarity and readily absorb moisture [11]. The material has been shown to be a proton conductor, particularly under humidified conditions [12]. This proton conductivity mechanism in chitosan is likely controlled by a Grotthuss-type water wire mechanism, mitigated by the acid-base equilibrium between amine groups and water present in the fibers or films [13]. It has been postulated that this property could influence charge transport at the filler-matrix interface. Chitin, when combined with other materials, has been proven to display enhanced dielectric energy storage capabilities. This evidence indicates the suitability of the material as an active dielectric component within a composite, not merely as a passive structural support. Figure 2 shows the schematic of chitin nanocrystal isolation from the chitin polymer.

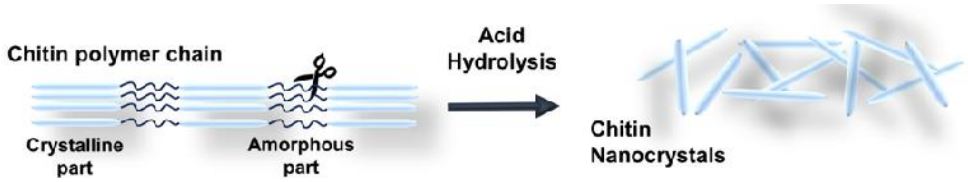


Fig. 2 *Chitin nanocrystals isolation from chitin polymer [7]*

A second key component identified in BSF pupal exuviae is melanin, the pigment responsible for its prominent dark color [7]. Melanin has long been recognized for its unique electrical properties, though its conduction mechanism has been the subject of ongoing debate [14]. Initially, melanin was modeled as a conventional amorphous organic semiconductor; however, recent advanced spectroscopic studies have revealed a more complex behavior. Notably, melanin's electrical conductivity is highly dependent on its hydration state. In its dry form, melanin is a poor conductor, but upon absorbing water, a chemical process known as comproportionating equilibrium is activated [15]. This reaction facilitates the transfer of protons and electrons among hydroxyquinone, semiquinone, and quinone groups within the melanin macromolecule, effectively "self-doping" the material with mobile charge carriers: electrons derived from semiquinone free radicals and protons present as hydronium ions [15]. As a result, hydrated melanin behaves as an electronic-ionic hybrid conductor, exhibiting enhanced conductivity due to the synergistic movement of electrons and ions.

This unique conduction mechanism has profound implications for its potential role in an HV insulating composite. The accumulation of localized static space charge is a key precursor to insulation failure via streamer and treeing mechanisms [16]. The hybrid conductivity of melanin could provide a localized pathway for the benign dissipation of this space charge before it can build up to critical levels, thereby suppressing the initiation of breakdown. This function would be highly advantageous, as it can directly address a primary failure mode of conventional insulators.

Furthermore, the electrical properties of melanin are tunable. It was reported in [17] that high-vacuum annealing has been shown to increase the material's conductivity by several orders of magnitude through the creation of more ordered molecular structures, such as transforming granules into a more sheet-like superstructure or reorganizing bonds to enhance electron density overlap and delocalization. This tunability, combined with its inherent biocompatibility [18] and good film stability [19], makes melanin a uniquely functional component within the BSF exuviae filler. Table 1 is a summary of the properties of BSF exuviae chitin and melanin.

A critical assessment of state-of-the-art polymeric insulators reveals a significant performance gap. Conventional petroleum-based epoxy resins offer excellent durability and resistance to electro-mechanical breakdown but are fundamentally unsustainable [20]. Conversely, the leading bio-based alternative, PLA, offers an excellent environmental profile but suffers from poor thermal stability and, most critically, rapid hydrolytic degradation that makes it unsuitable for long-service-life applications [3].

Overall, polymer composites present a proven strategy for overcoming the limitations of neat polymers, and the use of bio-fillers is a promising route toward sustainable, high-performance materials. This review has

identified BSF exuviae as a uniquely promising bio-filler. It is a sustainable by-product of a circular-economy industry [9] and, more importantly, is a multi-functional material. Its primary constituents, chitin and melanin, possess a combination of structural [15, 21], dielectric, and conductive properties that are theoretically well-suited to reinforce the PLA matrix while simultaneously protecting it from degradation [14, 21] and enhancing its ability to withstand high-voltage stress by managing space charge [22].

Table 1 *Comparative properties of key high-voltage insulation materials*

Property	Chitin	Melanin
Chemical Structure	Polysaccharide of N-acetyl-D-glucosamine [7]	Calcium carbonate ($CaCO^3$), particularly calcite [23]
Typical Content in Exuviae	16.2% (extraction yield), though another study reported 9% of dry body weight [7, 10]	5.1% (crude ash) [7]
Key Mechanical Property	Acts as a scaffold, can enhance the tensile properties of composite films [21]	Act as a protective layer against external stresses [7]; contributes to the hardness and elasticity of the cuticle [50]
Key Electrical Property	Dielectric; proton conductor under humidity [19]	Hydration-dependent electronic-ionic hybrid conductor [17]

Although the theoretical potential of PLA/BSF exuviae composites is significant, current scholarly research has yet to investigate this area thoroughly. As such, there is a notable knowledge gap concerning critical data such as Fourier transform infrared (FTIR) spectra, DC breakdown voltage, tensile strength, and the overall performance of these composites for HV insulation applications. This paper aims to fill that gap by developing PLA/BSF exuviae composites and experimentally evaluating their DC breakdown voltage alongside other important electro-mechanical properties. The findings are intended to advance the fields of materials science and high-voltage engineering.

2. Research Methodology

2.1 Sample Preparation and Production of PLA/Exuviae Composite

This study employed an extrusion process to prepare the PLA/exuviae composite. A twin-screw extruder, which consisted of two parallel screws rotating inside a barrel and could be used as co-rotating or counter-rotating, was utilized. This configuration provides strong shear and mixing action, enabling uniform dispersion of fillers such as exuviae particles into the PLA matrix [24]. Before extrusion, PLA pellets were dried in a vacuum oven at 85 °C for 24 hours to remove moisture that could cause the air to be trapped during processing, resulting in inhomogeneity of composites. Four composite formulations were prepared, with a pure PLA sample used as a control. The compositions of each sample, including the mixing of PLA, exuviae, glycerol, maleic anhydride (MA), and magnesium hydroxide ($Mg(OH)_2$) in wt%, are summarized in Table 2. The materials were manually premixed for 5 minutes to ensure initial homogeneity before extrusion.

Table 2 *Composite material distribution in each sample*

Sample	Weight of composite components wt%				
	Exuviae	Glycerol	Maleic Anhydride (MA), C2H4O3	Magnesium Hydroxide (Mg) (OH_2)	PLA
Pure PLA	0	0	0	0	600
PLA + 1% Exuviae	6	6	12	6	570
PLA + 2% Exuviae	12	6	12	6	564
PLA + 3% Exuviae	18	6	12	6	558

Figure 3 shows the sample preparation of PLA/Exuviae composite, as referred to in Table 2. In the extrusion process, the temperatures of the system were manipulated within a range of 180 °C-245 °C, with speed consistency around 45-80 rpm to provide sufficient shear for dispersion of exuviae particles without causing polymer degradation.

Int. Journal of Integrated Engineering Vol. 18 No. 7 (2026) p. 123-134

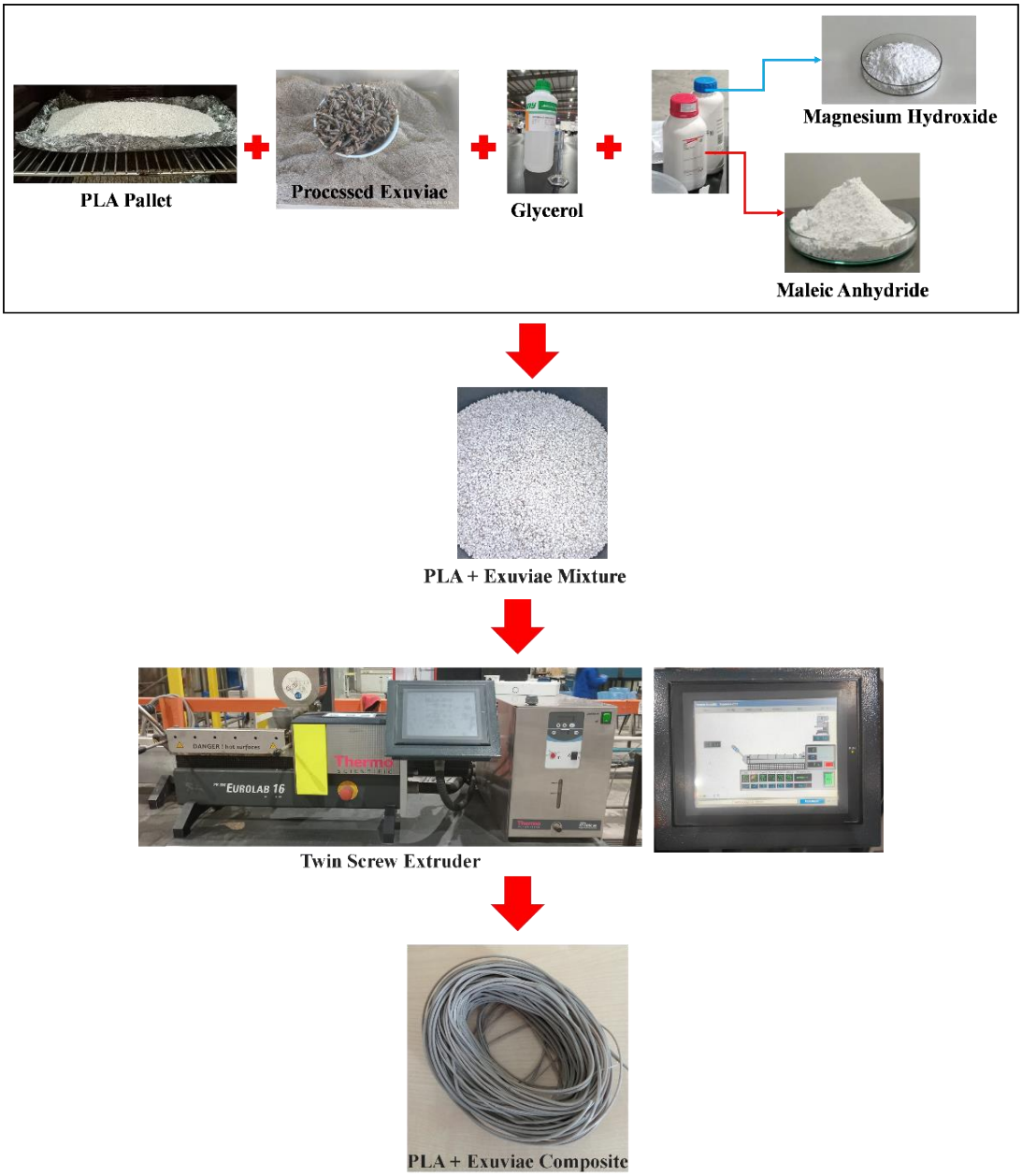


Fig. 3 *Sample preparation and production of PLA/Exuviae composite*

2.2 FTIR Testing

The characteristics of functional groups of composites were analyzed using FTIR spectroscopy (Shimadzu IRTracer-100), with a range of wavelengths from 500 cm-1 to 4000 cm-1. This instrument is a widely accepted and powerful technique for chemical characterization because it provides detailed information about molecular vibrations and functional groups present in materials. In this work, the analysis of spectroscopy of composites equipped with a mid-infrared triglycine sulfate (MIRTGS) detector is shown in Figure 4. This arrangement gives good sensitivity and stability, as well as increases the detection of fine spectral features.



Fig. 4 *Shimadzu IRTracer-100 FTIR*

2.3 DC Breakdown Testing

The DC breakdown strength of the PLA/exuviae composites was evaluated using a dielectric strength tester in accordance with ASTM D3755 standard. The test specimens were prepared with a nominal thickness of 100±5 µm (95 -105 µm). Each specimen was positioned between two stainless-steel spherical electrodes with a diameter of 6.3 mm, as illustrated in Figure 5. The electrode were immersed in transformer-grade mineral oil to prevent surface discharges during the test. All measurements were conducted at room temperature. Since the specimen was placed in direct contact with both electrodes, the electrode spacing was equivalent to the specimen thickness.

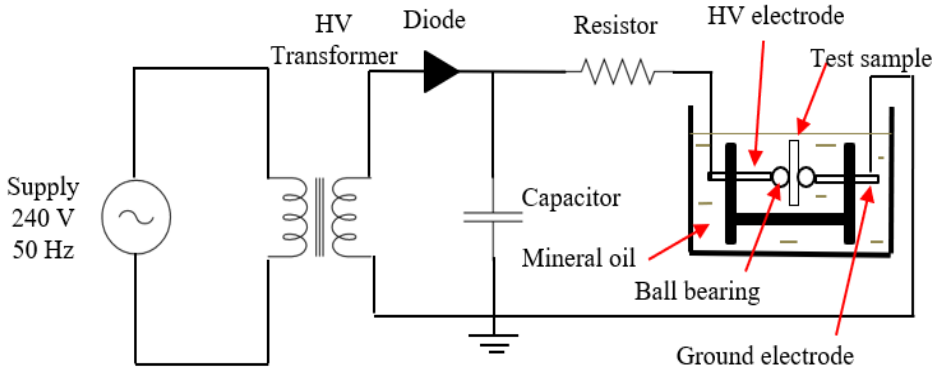


Fig. 5 *Illustration of the experimental setup employed for measuring the direct-current (DC) breakdown strength of the samples*

A DC voltage was applied at a controlled ramp rate of 2 kV every 20 s (equivalent to 100 V/s) until dielectric breakdown occurred. For each composite composition, three specimens were tested. To improve the statistical reliability of the results, five breakdown measurements were conducted at different locations on each specimen, resulting in a total of 15 breakdown measurements for each composition. The breakdown data were analysed using a two-parameter Weibull distribution to determine the characteristic breakdown strength and Weibull shape parameter.

2.4 Tensile Strength Testing

The tensile strength test was conducted using a Universal Testing Machine (UTM), which was calibrated prior to its use to ensure accuracy and minimize measurement errors. Figure 6 shows the UTM setup in which the tensile strength test was conducted. This machine is servo-driven and applies tensile loads through the displacement of the crosshead at a constant speed. The appropriate load cell should be selected to measure the forces involved, such as the failure load of the specimen. Mechanical wedge grips or pneumatic grips securely clamped the shoulder sections of the dog-bone specimens, preventing slippage without crushing the ends or causing premature failure.

Tensile testing was performed using a Universal Testing Machine (UTM) in accordance with ASTM D638 (Type I) at room temperature. Prior to testing, the width and thickness of the gauge section of each specimen were measured using a digital caliper to determine the initial cross-sectional area. The specimens were securely

Int. Journal of Integrated Engineering Vol. 18 No. 7 (2026) p. 123-134

clamped using mechanical grips with an initial grip separation of 40 mm, and an extensometer was attached to the gauge section to accurately measure strain. A constant crosshead speed of 10 mm/min was applied throughout the test. The test was continued until specimen failure, and five specimens were tested for each composite composition to ensure the reliability of the results.



Fig. 6 *Universal Testing Machine (UTM) setup for tensile test*

3. Results and Discussion

3.1 Chemical Content of Pure PLA and PLA Composites

The FTIR spectra of nanocomposites containing pure PLA and PLA composites are shown in Figure 7.

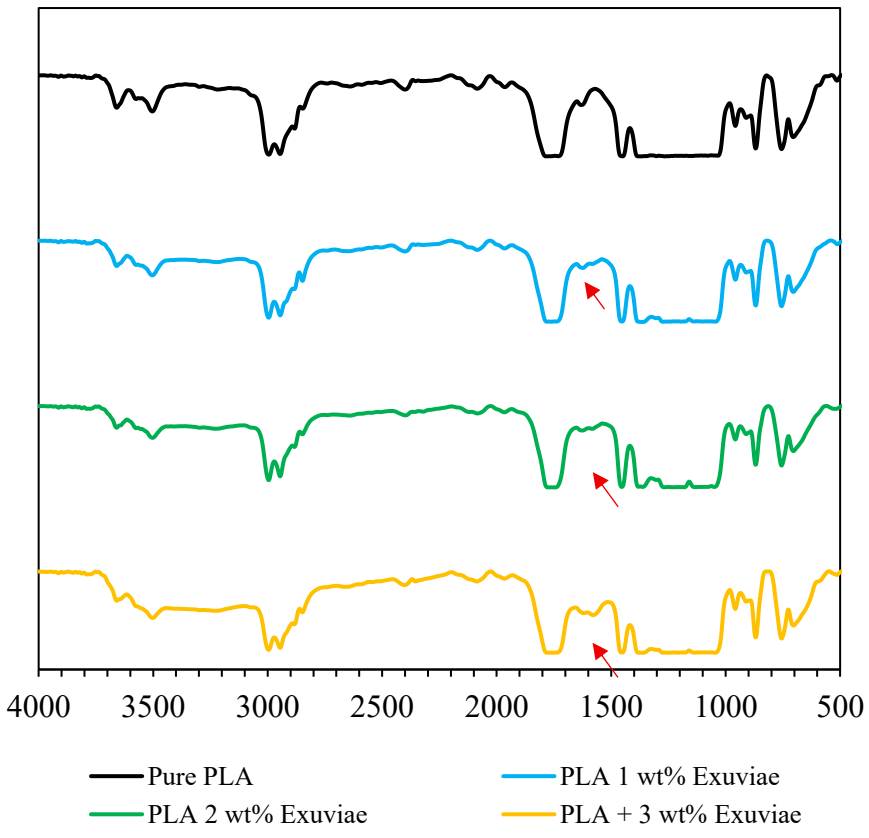


Fig. 7 *Comparison of pure PLA and PLA composites from FTIR spectra*

According to Figure 7, pure PLA exhibited stretching vibrations at 1746 cm^{-1} (C=O), 2995 cm^{-1} (asymmetric $-CH_3$), 2946 cm^{-1} (symmetric $-CH_3$), and 1080 cm^{-1} (C–O). Additionally, bending vibrations, corresponding to the asymmetric and symmetric $-CH_3$ groups, were observed at 1452 cm^{-1} and 1361 cm^{-1}, respectively. These findings are consistent with previously reported spectral features of PLA, as described in [25]. The incorporation of exuviae into pure PLA resulted in the appearance of new absorption bands at 1625 cm^{-1} and 1508 cm^{-1} (as indicated by the arrow), corresponding to amide I and amide II vibrations, respectively. Additional peaks observed at 2955 cm^{-1}, 2917 cm^{-1}, and 2848 cm^{-1} are attributed to the asymmetric stretching of $-CH_3$ and the asymmetric and symmetric stretching vibrations of $-CH_2$ groups, respectively. The emergence of a peak at 1733 cm^{-1}, associated with lipid C=O stretching, further confirms the successful integration of exuviae into the PLA matrix. This finding is in line with the research conducted in [26].

3.2 Dielectric Breakdown Strength of Pure PLA and PLA Composites

Figure 8 compares the DC breakdown strength of pure PLA and PLA composites, and their Weibull parameters are listed in Table 3. For pure PLA (as a reference), the measured DC breakdown strength was 319 ± 22 kV mm-1. After adding exuviae to PLA, the DC breakdown strength was reduced significantly to 102 ± 19 kV mm-1, 82 ± 15 kV mm-1, and 55 ± 16 kV mm-1 for 1 wt%, 2 wt%, and 3 wt% loading levels, respectively. The noticeable reduction in DC breakdown strength observed in the PLA composites, particularly at higher filler loadings, can be attributed to agglomeration effects. This observation aligns with the findings reported in [27].

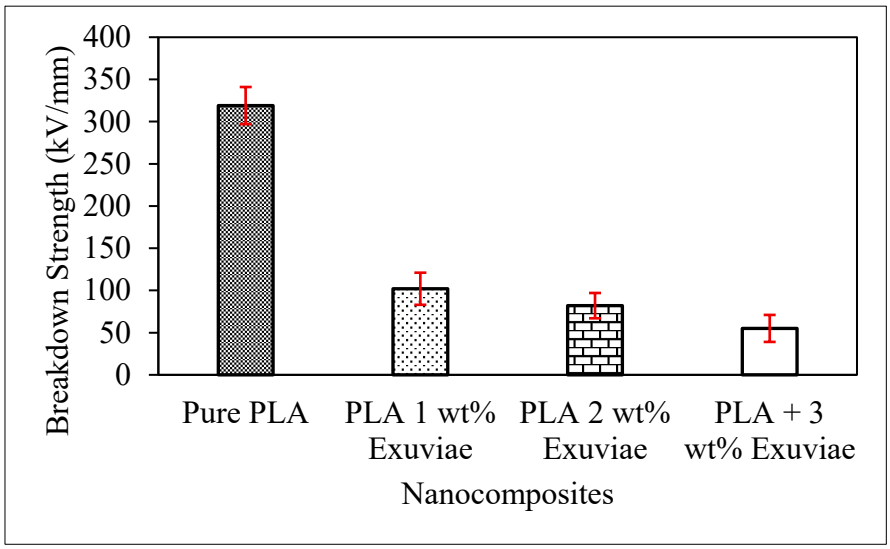


Fig. 8 *Comparison of DC breakdown strength of pure PLA and PLA composites*

Table 3 *DC breakdown parameters comparing pure PLA and PLA composites*

Sample	α (kV mm^{-1})	β
Pure PLA	319 ± 22	7 ± 2
PLA 1 wt% Exuviae	102 ± 19	3 ± 1
PLA 2 wt% Exuviae	82 ± 15	3 ± 1
PLA + 3 wt% Exuviae	55 ± 16	2 ± 1

Figure 9 presents an optical photograph of the PLA/exuviae composite, in which regions of non-uniform filler distribution can be qualitatively observed. Although the photograph suggests the presence of particle clustering, it does not provide definitive microstructural evidence of agglomeration. Nevertheless, the significant reduction in DC breakdown strength and tensile properties with increasing exuviae loading is consistent with the behaviour of polymer composites containing poorly dispersed fillers, as widely reported in previous studies [28]. It is therefore reasonable to infer that filler agglomeration and weak interfacial adhesion contributed to the deterioration of the electro-mechanical performance. Future work will employ scanning electron microscopy (SEM) to further examine the filler dispersion, interfacial bonding, and fracture morphology of the composites.

Int. Journal of Integrated Engineering Vol. 18 No. 7 (2026) p. 123-134

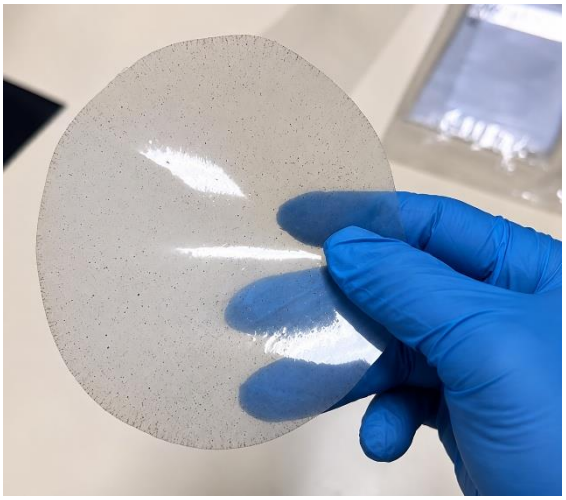


Fig. 9 *Agglomeration observed on the PLA/Exuviae sample*

3.3 Tensile Strength Results

Table 3 tabulates the tensile strength test results for pure PLA and PLA/exuviae composite. The results showed a clear trend: as the exuviae content increased from 0% (pure PLA) to 3%, both the maximum load and Young's modulus decreased. Pure PLA demonstrated the highest mechanical performance, with a maximum load of 120.77 N and a Young's modulus of 1721.66 MPa. The addition of exuviae, even at 1%, caused a significant drop in both properties (maximum load: 46.60 N, Young's modulus: 1460.58 MPa), and further increments in exuviae content to 2% and 3% continued this downward trend.

Table3 *Tensile strength test results for pure PLA and PLA/Exuviae composite*

Sample	Exuviae (%)	Maximum Load (N-avg)	Young's Modulus (MPa-avg)
Pure PLA	0	120.772	1721.66
PLA/Exuviae	1	46.5996	1460.575
	2	65.097	1440.478
	3	96.8122	1378.46

This reduction in mechanical properties with increasing exuviae content can be primarily attributed to poor interfacial adhesion and agglomeration of the exuviae particles within the PLA matrix. As the filler content increases, the likelihood of particle agglomeration rises, leading to the formation of stress concentration sites and defects that act as points of mechanical weakness during tensile loading. Agglomerated regions disrupt the continuity of the polymer matrix, impeding effective stress transfer from the matrix to the filler, which is crucial for reinforcement.

Similar findings have been reported in the literature [3, 7, 21, 29 – 31]. For example, excessive natural fiber content in PLA composites led to fiber association and network formation, which drastically reduced strength due to poor dispersion, and increased defect sites were observed in [29]. Agglomeration of nanoparticles in polymer matrices results in a decrease in mechanical properties because of the formation of weak zones, and inefficient load transfer was also highlighted in [30]. Furthermore, studies on PLA/natural filler composites consistently reported that without proper surface modification or compatibilization, the mechanical properties deteriorate with increasing filler loading due to these same mechanisms [31]. Thus, it can be concluded that increasing exuviae content in PLA composites leads to agglomeration and poor interfacial bonding, which in turn significantly reduces both the tensile strength and modulus of the material.

4. Conclusion

This study successfully developed PLA-based biocomposites reinforced with BSF exuviae and provided valuable insights into their chemical, electrical, and mechanical properties for high-voltage insulation applications. FTIR spectroscopy verified strong chemical interactions and effective incorporation of the exuviae within the PLA matrix. However, incorporation of exuviae resulted in significant reductions in DC breakdown strength and

mechanical properties, primarily due to filler agglomeration and poor dispersion, with values decreasing progressively at 1–3 wt% loadings. These results highlight both the potential and the challenges of using BSF exuviae as a sustainable filler in PLA composites. Future work should focus on improving filler dispersion to enhance the composite's electro-mechanical performance and fully realize its suitability for high-voltage insulation materials.

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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:*** *M. S. A. M. Sani, N. A. Othman;* ***data collection:*** *N. A. Othman, S. S Suradi, A. Azmi;* ***analysis and interpretation of results:*** *M. S. A. M. Sani, N. A. Othman, S. S Suradi, A. Azmi;* ***draft manuscript preparation:*** *M. S. A. M. Sani, N. A. Othman, S. S Suradi, A. Azmi, M.A.N.A.M. Nadzir. All authors reviewed the results and approved the final version of the manuscript.*

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