

PEDOT:PSS Conductive Polymer Ink for Functionalizing Glass Fiber Fabric Reinforcements Prior to Forming

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1. Abstract

With the rapid development and increasing adoption of synthetic- and natural-fiber-reinforced polymer composites across a wide range of industries, there is a growing need to introduce additional functionalities into next-generation composite products, particularly for smart process and product/service health monitoring. One promising approach is the integration of electrically conductive reinforcement fabrics into common composite manufacturing processes, enabling sensing capabilities to be embedded within the laminate, with minimal impact on structural performance. Because most conventional reinforcement fabrics are electrically insulating, imparting electrical conductivity is a critical first step. This work presents early results from our study aimed at developing and evaluating conductive inks suitable for coating composite fabrics while preserving essential reinforcement properties, such as flexibility, required during forming. To this end, conductive polymer inks based on poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) are investigated due to their known high electrical conductivity, chemical stability, and established use in other application areas, such as smart garments. Three scalable coating approaches, dip coating, drop casting, and spray coating, are examined for the deposition of PEDOT:PSS onto a typical glass fiber twill weave. The influence of the fabric surface chemistry on coating efficiency is investigated. Furthermore, processing with appropriate chemical dopants is performed to improve ink processability and adhesion to the substrate. The effectiveness of the selected coating techniques is evaluated and statistically compared in terms of coating uniformity, durability, and the functionality of the ensuing functional reinforcement.

2. Introduction

Advanced composite materials, particularly long fiber-reinforced polymers, are widely used in aerospace structures due to their high strength-to-weight ratio and design flexibility [1]. However, their laminated, anisotropic architecture makes them highly sensitive to defects that arise during manufacturing. In manufacturing processes such as forming and liquid composite molding, variations in fiber deformation, compaction, and resin flow can introduce defects, including fiber misalignment, wrinkling, voids, and dry regions. These process-induced defects act as stress concentrators and can considerably reduce the mechanical performance and reliability of the final structure [2]. As a result, there is a strong need for monitoring strategies that can identify the

defects at their origin, rather than relying solely on post-manufacturing inspection or in-service evaluation.

Structural health monitoring (SHM) has traditionally focused on detecting damage during the service life of products. However, its extension toward in-situ process monitoring is increasingly becoming important for ensuring manufacturing quality [2]. Conventional sensing approaches, such as embedded fiber optics or discrete sensors, are often intrusive, difficult to scale, and may disrupt the structural integrity of the composite during processing [3]. This has led to growing interest in self-sensing composites, where the reinforcement itself is functionalized to provide sensing capability. In this approach, fiber reinforcements can be engineered to respond electrically to deformation, enabling real-time tracking of forming behavior, resin infiltration, and defect formation directly within the material.

To enable such functionality, typical insulating reinforcements such as glass fiber (GF), as the focus of this case study, must be modified to become electrically active. GF is inherently non-conductive and chemically inert due to its silica-based network structure, which limits both electrical response and interfacial interactions with coatings [4]. This necessitates modifying the fabric's surface to enhance electrical conductivity or enable on-demand functionality. A range of approaches has been reported in the open literature to increase surface energy and introduce reactive functional groups, including plasma treatment, chemical grafting, acid-base etching, fluorination, and coating-based strategies [5]. Among these approaches, *silane* treatment as part of the coating strategies has been particularly effective. Silane coupling agents react with hydroxyl groups present on the GF surface, forming covalent Si-O-Si bonds and creating a functional interfacial layer. This modification improves wettability and provides chemical compatibility with organic coatings, enabling stronger adhesion and more uniform coverage.

Traditionally, conductivity has been introduced using metallic coatings or carbon-based nanomaterials such as carbon nanotubes and graphene [6], [7]. While these materials offer high conductivity, they often present challenges related to stiffness, dispersion, and compatibility with textile processing routes. Conductive polymers are considered a more compatible alternative for textile-based systems. For instance, poly (3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) is widely used due to its solution-processability, flexibility, and ability to form conformal coatings on fibrous substrates [8]. As shown in Figure 1, PEDOT:PSS consists of conductive (and hydrophobic) PEDOT domains stabilized by the insulating (and hydrophilic) PSS, where the eventual charge transport depends on the connectivity between PEDOT domains [9]. The electrical performance of this system can be considerably enhanced through secondary doping [10]. To this end, the addition of polar solvents induces structural rearrangement of the polymer chains, reducing inter-chain spacing and promoting more ordered conductive pathways, thereby improving electrical conductivity.

In previous investigations, PEDOT:PSS has been deposited onto textile substrates using a wide range of techniques, including solution-based methods such as dip coating and printing, as well as more advanced approaches such as vapor-phase polymerization and in situ polymerization [11]. While these techniques can achieve high-quality coatings, solution-based methods are more widely adopted due to their scalability and compatibility with large-area and flexible substrates. PEDOT:PSS has been extensively used in wearable and flexible electronics, where it can be readily applied to soft, polymer-based textiles, such as polyester, to impart electrical conductivity for sensing and energy storage [12]. Despite the widespread use of PEDOT:PSS in wearable systems,

its application to other fibrous textiles, particularly for structural and process-monitoring functionalities in the composites field, remains largely unexplored.

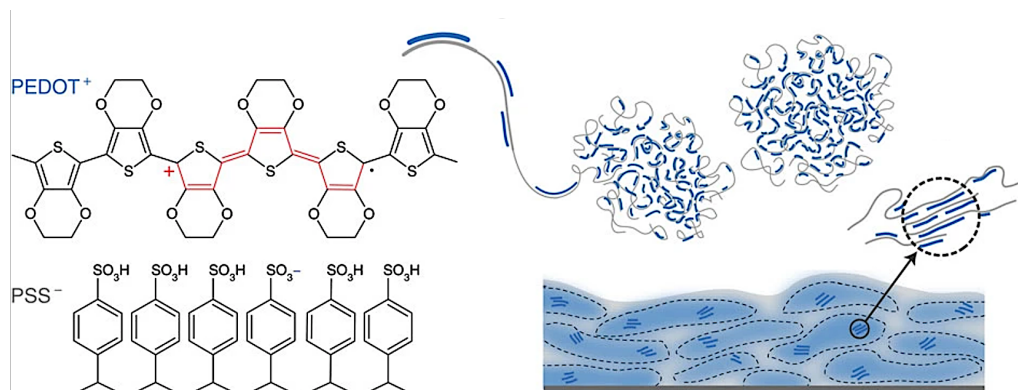


Figure 1. Schematic representation of the PEDOT:PSS system, showing the chemical structure, formation of the colloidal gel particles in aqueous dispersion, and the resulting film microstructure consisting of PSS-rich regions (gray) and PEDOT-rich conductive domains (blue). Adopted with permission from [13].

In addition to enabling sensing, conductive modification of textile reinforcements can allow the integration of multifunctional properties such as electromagnetic interference (EMI) shielding [14]. Herein, we aim to investigate the deposition of PEDOT:PSS onto a typical glass fiber twill weave and to provide early-stage insights into the fabrication of the ensuing conductive reinforcements for composite process-monitoring applications. Different solution-based deposition approaches, namely dip coating, drop casting and spray coating are deployed and compared in terms of coat quality, including uniformity and adhesion to the fibrous surface. The resulting coatings are also further assessed for their ability to impart electrical conductivity and EMI shielding to the glass fiber fabric.

3. Materials and Methods

3.1. Materials

A 280 g/m² 2×2 twill woven glass fiber (GF) fabric (Easy Composites, UK) was used as the reinforcement substrate. Aqueous dispersion of PEDOT:PSS was supplied by Heraeus (Clevios™ PH1000). (3-Aminopropyl)trimethoxysilane (APTMS) as the silane coupling agent and dimethyl sulfoxide (DMSO) as the PEDOT:PSS dopant were supplied by Sigma-Aldrich. All materials were used as received.

3.2. Surface Modification of Glass Fiber

Glass fiber fabrics were cut into 40×40 mm specimens and mounted in a custom-made polytetrafluoroethylene (PTFE) holder to maintain dimensional stability and ensure uniform exposure during subsequent treatment (Figure 2a). Surface functionalization with APTMS was performed to improve wettability and interfacial compatibility with the conductive coating. As such, the fibers were immersed in an 80 vol% ethanolic solution containing 2 vol% APTMS (homogenized via 30 min stirring). The samples were kept stationary for 24 h, followed by extraction and thermal treatment for 1 h at 120 °C to promote silane coating and remove residual solvent. The silane molecules react with surface hydroxyl (Si-OH) groups on the glass fiber, forming covalent Si-O-Si bonds and introducing amine-functional groups (-NH₂), which enhance coating adhesion and surface reactivity [15].

3.3. Preparation of Doped PEDOT:PSS Solution

To enhance the electrical conductivity of PEDOT:PSS, dimethyl sulfoxide (DMSO) was added as a secondary dopant at 5 vol%, based on reported optimal concentrations [10], [16]. The mixture was stirred for 30 min at room temperature and subsequently sonicated in a water bath for 15 min to ensure homogeneous dispersion. The addition of DMSO induces a screening effect, weakening Coulombic interactions between PEDOT and PSS chains, as shown in Figure 2b. This leads to conformational changes from a coiled to a more linear or expanded structure, reduces the π - π stacking distance, and promotes phase separation and ordering of PEDOT-rich domains, resulting in improved charge transport and electrical conductivity [17].

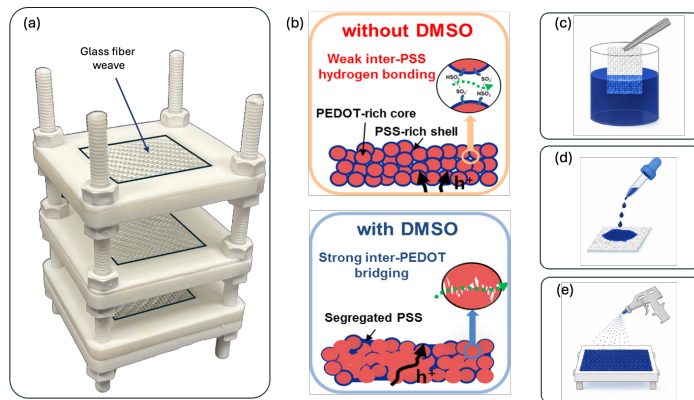


Figure 2. (a) Custom-made fabric sample holder made of PTFE. (b) Schematic of PEDOT:PSS before and after DMSO doping. DMSO induces PSS segregation and promotes inter-PEDOT chain bridging, leading to enhanced charge transport and electrical conductivity. Adopted with permission from [17]. Schematic of (c) dip coating, (d) drop casting, and (e) spray coating techniques.

3.4. Deposition Techniques

Surface-modified GF samples were coated with DMSO-doped PEDOT:PSS using dip coating, drop casting, and spray coating, as schematically illustrated in Figure 2c-e. Dip coating involved immersing the samples in the solution, while drop casting involved dispensing the solution onto the fabric surface and allowing excess liquid to drain. Spray coating was carried out using a spray gun at a distance of ~10 cm. For spray deposition, one, three, and five successive layers were applied to assess the effect of coating buildup. All coated samples were thermally treated at 130°C for 30 min to remove residual solvent and promote structural rearrangement of PEDOT:PSS, enhancing film cohesion and electrical conductivity.

3.5. Characterization

Fourier transform infrared spectroscopy (FTIR) was performed using a Thermo Scientific Nicolet iS20 instrument to evaluate chemical interactions and functional group changes. Surface morphology and elemental distribution were analyzed using field-emission scanning electron microscopy (FESEM) equipped with energy-dispersive spectroscopy (EDS) (Tescan Mira 3 XMU) at an accelerating voltage of 15 kV and working distance of 15 mm. Samples were sputter-coated with a 15 nm platinum layer prior to imaging.

Electrical conductivity was measured using a Loresta GX four-point probe system. Coating durability was evaluated using a tape peel test in accordance with ISO 2409. EMI shielding effectiveness (SE) was measured in the X-band frequency range (8.2-12.4 GHz) using a vector network analyzer (VNA; Keysight model P9374A) equipped with a WR90 waveguide fixture. For EMI SE measurement, the samples from 1 to 3 layers were placed between waveguide adaptors,

and complex scattering parameters (S_{11} , S_{21} , S_{22} , and S_{12}) were collected to calculate the shielding coefficients, including reflectance (R), transmittance (T), and absorbance (A):

$$R = |S_{11}|^2 \quad (1)$$

$$T = |S_{21}|^2 \quad (2)$$

$$A = 1 - R - T \quad (3)$$

The shielding coefficients were then used to calculate total shielding effectiveness (SE_T) from the sum of reflection loss (SE_R) and absorption loss (SE_A) using the following formula:

$$SE_R = 10 \log \left(\frac{1}{1 - R} \right) \quad (4)$$

$$SE_A = 10 \log \left(\frac{1 - R}{T} \right) \quad (5)$$

$$SE_T = SE_A + SE_R \quad (6)$$

Based on the above formulation, EMI shielding of a material system may be aimed at attenuating the incident electromagnetic radiation through reflection, absorption, or multiple internal reflections [18].

4. Results and Discussion

The evaluation of the physicochemical and functional properties of PEDOT:PSS-coated GF shows a strong correlation between coating uniformity and conductive network formation. FTIR spectra (Figure 3) confirmed the successful presence of PEDOT:PSS on the coated fabric samples (under different coating techniques) relative to the pristine GF. While pristine GF was dominated by the characteristic glass-related absorption band around 1029 cm^{-1} , assigned to Si-O-Si vibrations, the coated samples showed the emergence or strengthening of PEDOT:PSS-related features, particularly around 1197 cm^{-1} for S=O stretching in PSS sulfonate group and DMSO, and 1624 cm^{-1} indicating C=C stretching in thiophene ring of PEDOT. The differences in band intensity among the coated samples further suggested variations in deposited amount and coating continuity. In particular, the spray-coated fabrics exhibited more pronounced PEDOT:PSS-associated spectral features than the dip and drop cast samples.

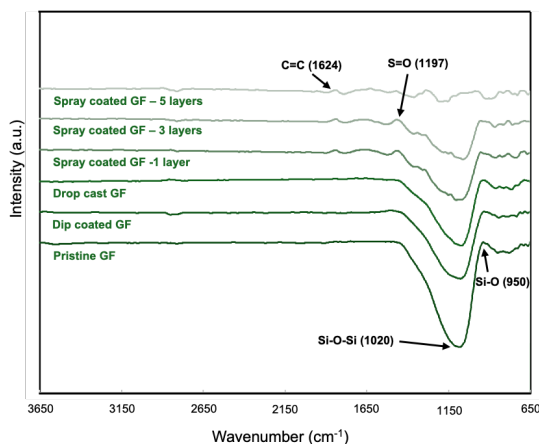


Figure 3. FTIR spectra of pristine and PEDOT:PSS-coated GFs prepared via different coating techniques.

FESEM imaging and sulfur EDS mapping further verified these differences, as demonstrated in Figure 4. Since sulfur originates from the components in the thiophene ring (C-S-C in PEDOT's backbone), the sulfonate group in PSS, and S=O in DMSO, its presence directly reflects the doped PEDOT:PSS coverage on the GFs. The spray-coated samples, especially the three-layer spray-coated samples, showed the strongest sulfur signal, with the highest sulfur content reaching 11.7%, indicating the most effective coating. By contrast, dip-coated and drop-cast samples exhibited lower sulfur content and less homogeneous distribution, with sulfur elemental ratios of 2.9% and 3.7%, respectively.

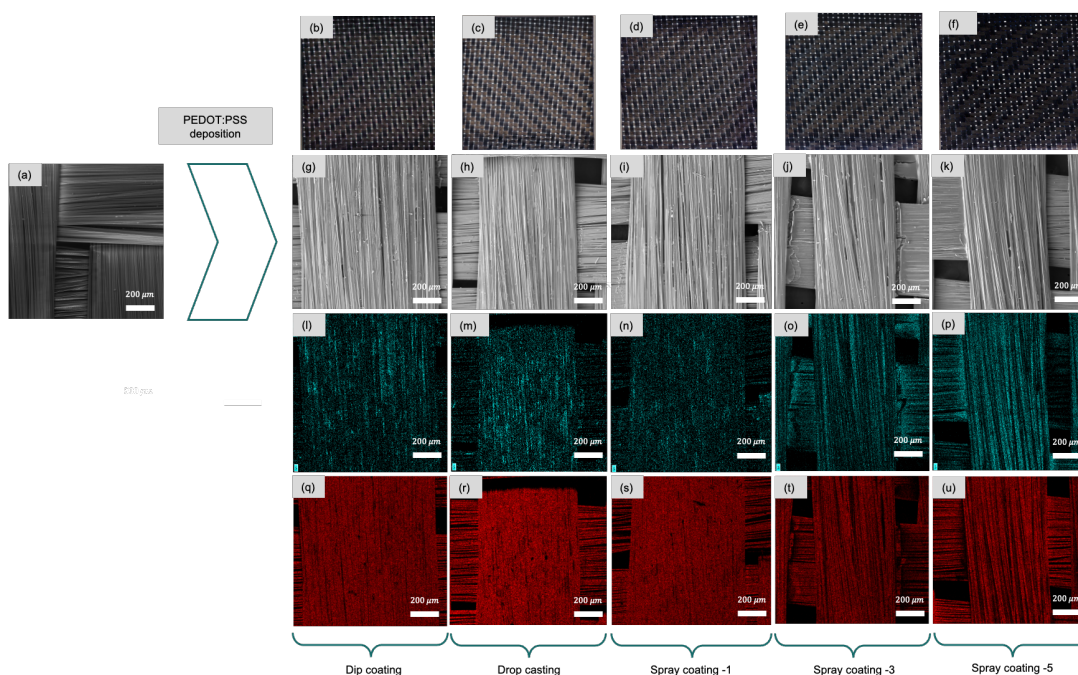


Figure 4. Morphology of pristine and PEDOT:PSS-coated GF fabrics across various coating techniques. (a) Pristine GF. (b-f) Optical images of 40 × 40 mm coated samples. (g-k) SEM images showing coating morphology and distribution. (l-p) Sulfur (S) EDS maps confirming PEDOT:PSS presence on glass fiber presented as (q-u) Silica (Si) EDS maps

When surface modification is present, amine groups can enhance interfacial interactions of the fabric surface through electrostatic attraction and hydrogen bonding with the sulfonate groups of PSS. The droplet-based deposition in spray coating increases the effective contact area between the polymer and these active sites, facilitating more uniform adsorption and anchoring of the conductive phase. In contrast, dip coating and drop casting involve bulk liquid immersion or deposition, where slower solvent evaporation and polymer redistribution often lead to accumulation in inter-fiber regions, the formation of thicker, localized films, and non-uniform coverage. As Figure 4.f shows, in the five-layer spray-coated samples, PEDOT:PSS is also present in the inter-yarn regions between warp and weft, suggesting that increased deposition promotes infiltration into the fabric architecture and partial pore filling, which may contribute to higher conductivity but reduced coating selectivity on individual filaments.

The conductivity results showed a strong dependence on both the coating method and the deposition level, as shown in Figure 5a. The mean conductivity of the fabrics with an average thickness of 0.31 mm ranged from approximately 0.14-0.19 S/cm for one-layer spray-coated and dip-coated samples to 0.99 S/cm for drop-cast fabrics and increased considerably to 2.70 S/cm and 8.00 S/cm for three-layer and five-layer spray-coated samples, respectively. This progression

indicates that increasing the number of spray passes promotes the formation of more continuous and effective conductive pathways than dip coating and drop casting, consistent with the FTIR, FESEM, and EDS results. For self-sensing materials, the literature generally reports that useful piezoresistive behavior is often achieved in moderately conductive networks near the percolation threshold rather than in excessively conductive systems, typically within a broad range of 0.0001 to 10 S/cm, depending on the sensing mode [19], [20]. Overall, the present conductivity results indicate that the developed coatings meet, and in the case of spray-deposited samples, exceed the typical requirements for conductive and self-sensing textile systems.

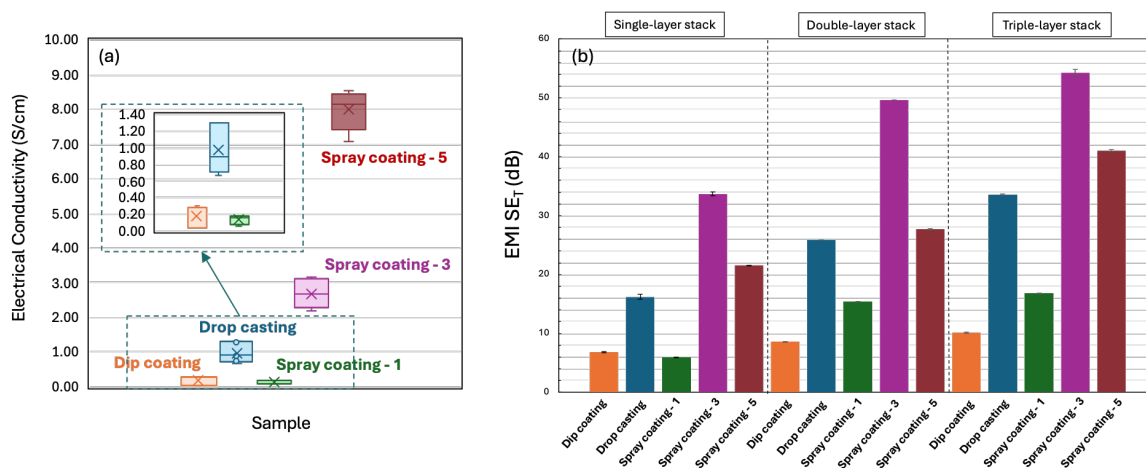


Figure 5. (a) Electrical conductivity of PEDOT:PSS-coated glass fiber fabrics for different coating methods. (b) Total EMI shielding effectiveness (SE_T) for single-layer, double-layer, and triple-layer stacked fabric configurations.

A one-way ANOVA conducted on the conductivity data confirmed that the coating method had a statistically significant effect on electrical performance ($F = 436.58$, $p < 0.0001$). This indicates that the observed differences among dip coating, drop casting, and layered spray coating are not due to random variation. The largest separation occurred between the lower-performing groups (dip coating and one-layer spray coating) and the higher-performing multilayer spray coating group, consistent with the trends observed in FTIR and EDS.

In order to evaluate the shielding performance, stacked/laminate configurations were also considered, where one, two, and three coated fabric layers were placed on top of each other (Figure 5b). Notably, the highest shielding performance in single-layer stacking was achieved with the three-layer spray-coating condition (33 dB), despite the five-layer spray-coated samples exhibiting the highest electrical conductivity. Hypothetically, this suggests that EMI shielding is not solely governed by electrical conductivity but also by coating uniformity, thickness, and microstructural continuity. Successive deposition in five sprays might lead to polymer aggregation in inter-yarn regions and localized thickening, likely reducing shielding efficiency despite improved conductivity, a notion that needs to be further assessed in a future study.

As expected, SE increased with the number of stacked layers across all coating methods, with three stacks of three-layered spray-coated GF reaching an average shielding of 54 dB. From an application perspective, ~20 dB SE corresponds to ~99% attenuation and ~30 dB to ~99.9%, and values in the range of 40-50 dB are considered suitable for advanced electronics and lightweight aerospace applications [21].

Adhesion behavior was qualitatively assessed through a tape-peeling approach inspired by the classification scheme of ISO 2409 [22], and illustrated in Figure 6. In this method, adhesive tape was applied to the coated fabric surface and subsequently removed to evaluate coating retention. The dip-coated, drop-cast, and single-layer spray-coated samples showed no visible PEDOT:PSS removal (as shown in Figure 6.d for the single-layer spray-coated sample), corresponding to a classification of 0 (no flaking). In contrast, the three-layer and five-layer spray-coated samples exhibited only minor traces of coating on the tape, consistent with a classification of 1, indicating minimal detachment. It is also notable that all coated fabrics retained their ability to bend and shear, indicating that the coating did not significantly compromise the textile structure's inherent flexibility. However, quantitative mechanical testing is required in future work to fully assess the effect of coating on fabric deformability.

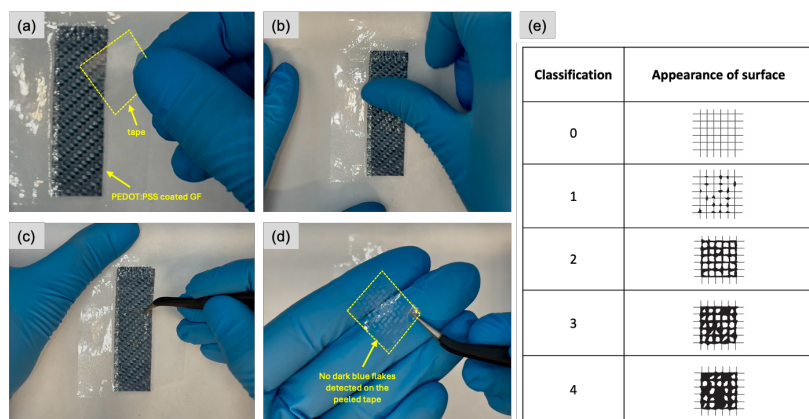


Figure 6. Tape-peeling adhesion assessment of PEDOT:PSS-coated glass fiber. (a-d) Sequential steps for a spray-coated sample, as an example: adhesive tape applied to the coated surface, pressed to ensure contact, and peeled off to evaluate coating retention. (e) Reference classification scale adopted from ISO 2409 used for qualitative comparison of coating removal [22].

5. Conclusion

This study demonstrated a scalable approach for functionalizing glass fiber woven fabrics with PEDOT:PSS to introduce electrical conductivity prior to composite forming, with the deposition method identified as a key parameter governing performance. Among different coating techniques tested, spray coating produced the most conformal coatings, with conductivities up to 2.97 S/cm and an EMI shielding effectiveness of ~33 dB at the optimal three-layer condition. All samples exhibited conductivity within the functional range for conductive and self-sensing textiles. It was observed that while the five-layer spray coating produced the highest conductivity, three-layer spray-coated samples delivered the best overall performance by combining strong PEDOT:PSS presence, good adhesion, and high conductivity, along with the maximum EMI shielding effectiveness. This observation suggested that excessive coating deposition in multi-layer configurations might lead to polymer aggregation in inter-yarn regions, cause localized thickening and incident-wave internal reflections, and reduce shielding efficiency despite the thicker laminate, an aspect that warrants further investigation. Future work should also evaluate the effectiveness of the top-ranked spray deposition method in terms of subsequent composite process compatibility; e.g., sufficient flexural behavior during fabric forming, while reporting wrinkles.

6. References

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