

Article

Preparation of MoS₂@PDA-Modified Polyimide Films with High Mechanical Performance and Improved Electrical Insulation

Xian Cheng ^{1,2}, Chenxi Wang ^{1,2}, Shuo Chen ^{1,2,*}, Leyuan Zhang ^{1,2}, Zihao Liu ^{1,2} and Wenhao Zhang ^{1,2}

¹ School of Electrical and Information Engineering, Zhengzhou University, Zhengzhou 450001, China; chengxian@zzu.edu.cn (X.C.); wangchenxi@gs.zzu.edu.cn (C.W.); zly19991124@163.com (L.Z.); 15139085273@163.com (Z.L.); zwh1823641629@163.com (W.Z.)

² He'nan Engineering Research Center of Power Transmission and Distribution Equipment and Electrical Insulation, Zhengzhou 450001, China

* Correspondence: chenshuo_zzu@gs.zzu.edu.cn

Abstract: Polyimide (PI) has been widely used in cable insulation, thermal insulation, wind power protection, and other fields due to its high chemical stability and excellent electrical insulation and mechanical properties. In this research, a modified PI composite film (MoS₂@PDA/PI) was obtained by using polydopamine (PDA)-coated molybdenum disulfide (MoS₂) as a filler. The low interlayer friction characteristics and high elastic modulus of MoS₂ provide a theoretical basis for enhancing the flexible mechanical properties of the PI matrix. The formation of a cross-linking structure between a large number of active sites on the surface of the PDA and the PI molecular chain can effectively enhance the breakdown field strength of the film. Consequently, the tensile strength of the final sample MoS₂@PDA/PI film increased by 44.7% in comparison with pure PI film, and the breakdown voltage strength reached 1.23 times that of the original film. It can be seen that the strategy of utilizing two-dimensional (2D) MoS₂@PDA nanosheets filled with PI provides a new modification idea to enhance the mechanical and electrical insulation properties of PI films.

Keywords: PI; MoS₂ nanosheets; composite; mechanical properties; aging life



Citation: Cheng, X.; Wang, C.; Chen, S.; Zhang, L.; Liu, Z.; Zhang, W. Preparation of MoS₂@PDA-Modified Polyimide Films with High Mechanical Performance and Improved Electrical Insulation. *Polymers* **2024**, *16*, 546. <https://doi.org/10.3390/polym16040546>

Academic Editors: Diego Antonioli and Raluca Nicoleta Darie-Nita

Received: 7 January 2024

Revised: 4 February 2024

Accepted: 5 February 2024

Published: 17 February 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Polyimide (PI) is widely used in the field of electronic devices, aerospace and high-performance coatings due to its superior thermal stability, mechanical strength, and chemical resistance [1–3]. In addition, PI materials are suitable for high-voltage insulation due to their superior electrical insulation properties, especially in the field of turn-to-turn insulation where they play a vital role [4,5]. As a key component of electrical equipment such as motors and transformers, inter-turn insulation needs strong electrical insulation properties to cope with high amplitude and frequency pulses of high voltage. More critically, the inter-turn insulation demands superior mechanical and frictional properties to reduce air gaps formed during the winding process due to the poor fit of insulating materials. These air gaps can lead to partial discharges under the action of strong electric fields. Moreover, strong leakage in electromagnetic fields may cause mechanical damage to the winding ends. The above factors are the main reasons for the deterioration of the insulation properties of PI materials [6–8].

Two-dimensional transition metal sulfides have received widespread attention in scientific research and industry due to their unique physical and chemical properties [9–11]. Molybdenum disulfide (MoS₂) is a two-dimensional material with a special layered structure and a high degree of surface activity, which makes it excellent in terms of mechanical and friction properties [12,13]. Yuan et al. [14] found that doping a small amount of MoS₂ (0.75 wt.%) can significantly enhance the mechanical properties of MoS₂/PI nanocomposite films. Liu et al. [15] found that MoS₂ can effectively reduce the deformation of the PI wear surface, thus reducing and stabilizing friction. Guo et al. [16] doped phenolic resin (CPF)

composites with 10 wt.% MoS₂, which resulted in a 56% decrease in wear rate compared to pure CPF. The above studies show that MoS₂-modified polymer composites have great strength and toughness, a low coefficient of friction, and high wear resistance. However, since MoS₂ is a semiconductor material with a narrow forbidden bandwidth, its application in the field of high-voltage insulation is limited. Pang et al. [17] showed that in terms of pressure cylinders in high-voltage switchgear, excessive accumulation of MoS₂ reduces the arc extinguishing and breaking capacity of the switchgear, which leads to a decrease in overall insulation performance. Meanwhile, the high surface energy of nanomaterials tends to cause agglomeration, so the interfacial interaction between MoS₂ and the PI matrix may also be affected by its dispersion [18–20].

Polydopamine (PDA) is a substance formed by the self-polymerization of dopamine in an alkaline environment, with a unique structure and functional groups. PDA has a great property boost effect on the mechanical properties of composites due to its ability to form various types of bonds with other materials, such as hydrogen and coordination bonds [21–23]. Dong et al. [24] found that PI composites doped with 5 wt.% of PDA-encapsulated graphitic carbon nitride had the highest breakdown field strength of 300 kV/mm, which was 67.6% higher than that of pure PI. Feng et al. [25] found that the interfacial interaction between filler and polyarylene ether nitrile was enhanced by a core-shell structural composite (MoS₂@PDA). Sanusi Hamat et al. [26] found that a coupling agent (PDA) could increase the tensile Young's modulus, flexural Young's modulus, and compressive stress of kenaf fibers and the PLA matrix by 13.4%, 15.3%, and 30%, respectively. Therefore, the use of MoS₂@PDA core-shell structure nanoparticles doped with modified PI materials is expected to further enhance the mechanical and friction properties of the composites, as well as effectively improve the electrical insulation properties of the composites. This comprehensive performance improvement is essential to safeguard the insulation life of the material during long-term use and is also significant in improving the operational reliability of electrical equipment.

In this research, a core-shell structure of MoS₂ encapsulated in PDA was strategically employed for the optimization and modification of PI composites. The PDA not only enhances the dispersion and compatibility of MoS₂ within the PI matrix but also significantly increases the flexibility of the composite film. This crucial enhancement in flexibility is essential for the minimization of the partial discharge phenomenon, particularly those discharges caused by air gaps between coil turns. Moreover, the PDA layer acts as an effective barrier, markedly reducing the electronic excitation and migration in the MoS₂ semiconductor. This substantial reduction is pivotal in significantly improving the overall electrical insulation properties of the composite material.

2. Materials and Methods

2.1. Materials

Thiourea (CH₄N₂S), ammonium molybdate tetrahydrate ((NH₄)₆Mo₇O₂₄·4H₂O), Tris(hydroxymethyl) aminomethane (Tris, C₄H₁₁NO₃), dopamine hydrochloride (C₈H₁₁NO₂·HCl), and N,N-Dimethylacetamide (CH₃CON(CH₃)₂) were purchased from Shanghai Macklin Biochemical Technology Co., Ltd. (Shanghai, China). The PI precursor (PAA) was purchased from Changzhou Runchuan Plastic Material Co., Ltd. (Changzhou, China) as well as the deionized water.

2.2. Synthesis of MoS₂ Nanosheets

In this procedure, 0.144 g of ammonium molybdate tetrahydrate and 1.010 g of thiourea were accurately measured and introduced into a beaker containing 60 mL of water as the chosen solvent. The mixture was then stirred continuously for 2 h to ensure the sample was completely dissolved. Once fully dissolved, the homogenous solution was carefully transferred into the reactor. The reactor was subsequently heated for a duration of 12 h at a stable temperature of 180 °C. After the heating process, MoS₂ was successfully obtained through a methodical process of filtration and subsequent drying under controlled conditions.

2.3. Synthesis of MoS₂@PDA Nanosheets

Approximately 0.5 g MoS₂ was dispersed in 250 mL of deionized water, ultrasonicated, and stirred for 0.5 h to obtain suspension A. Approximately 0.67 g of Tris was added to A, and we continued to stir it ultrasonically for 0.5 h to obtain suspension B. Finally, 0.1 g of dopamine hydrochloride was added to solution B, and it was placed on a magnetic stirrer at ambient temperature and stirred for 24 h. Then, it was filtered and dried in a vacuum drying oven for 12 h to obtain MoS₂@PDA nanosheets.

2.4. Synthesis of MoS₂/PI and MoS₂@PDA/PI Films

Figure 1 shows the synthesis process for MoS₂/PI and MoS₂@PDA/PI films. Approximately 150 mg MoS₂ was sonicated and dispersed in 5 mL N,N-dimethylacetamide and stirred for 30 min; then, 25 g of PAA was added and stirred for 24 h until the MoS₂ nanosheets were evenly dispersed in the above solution. After eliminating the air bubbles using a vacuum pump, the solution was poured into a glass plate and coated with the film evenly. Finally, MoS₂/PI film was obtained after heating the coating glass plate at different temperature gradients in an oven. The MoS₂@PDA/PI composite films could be obtained by means of the above steps using the same content of MoS₂@PDA nanosheets. In addition, the pure PI film was obtained by heating the PAA film.

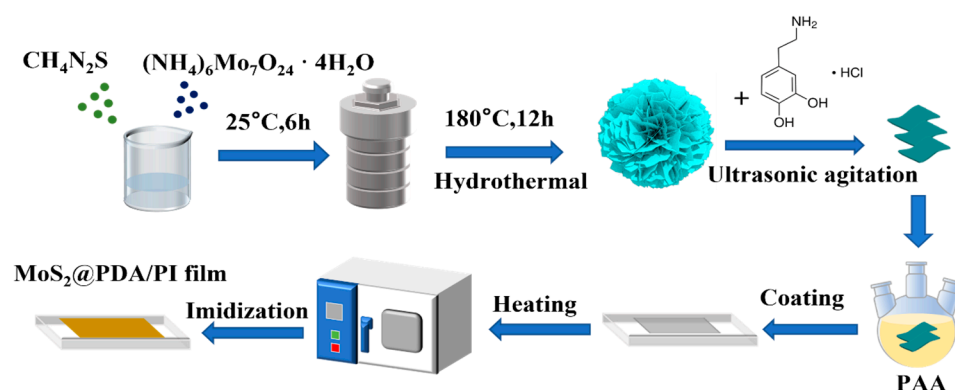


Figure 1. Schematic diagram of the synthesis process of MoS₂@PDA/PI composites.

2.5. Materials Characterization

The microstructure and morphology of the as-prepared samples were characterized by field emission scanning electron microscopy (F-SEM; JEOL JSM-7800F, JEOL, Tokyo, Japan). The sample was dripped onto a copper grid and dried before observation under transmission electron microscopy (TEM), with the test temperature set at 30 °C and the magnification range from 5000 to 50,000 times. The lattice structure of MoS₂ and MoS₂@PDA composites was further characterized by TEM (JEOL JEM-2100), with the test temperature maintained at 30 °C and the magnification range from 12,000 to 120,000 times. The surface functional groups of the samples were analyzed using Fourier-transform infrared spectroscopy (FT-IR, Nicolet iS50, Nicolet, Green Bay, WI, USA), with the test temperature held at 30 °C and the wavelength range tested between 600 and 4000 cm^{−1}. The crystalline structure of the samples was determined using powder X-ray diffraction (XRD, Rigaku Ultima IV with Cu-Kα radiation, λ = 0.15418 nm, Rigaku, Tokyo, Japan) over a range of 10–90° and X-ray photoelectron spectroscopy (XPS, AXIS Supra) over a range of 0–1200 eV, with the test temperature set at 30 °C. The friction and wear properties were measured using a friction wear test machine (MG2000). The experimental load was set to 120 N, the experimental duration was 5 min, and the speed of the friction wear testing machine was maintained at 200 r/min. The dielectric constant and loss tangent were obtained using a broadband dielectric spectrometer and impedance analyzer (Novocontrol Concept 80, Novocontrol Technologies, Frankfurt, Germany). The frequency range covered was from

10^2 to 10^6 Hz, and the test temperature was held constant at 30 °C. An electrode with a diameter of 30 mm was sprayed on both sides of the samples to facilitate the measurements.

3. Results

3.1. Morphology Analyses

The 2D nanostructure of MoS₂ can be observed through the SEM and TEM images. In Figure 2a, the MoS₂ nanosheets undergo a self-assembly process to form a nanoflower with a diameter of about 300 nm, and the size is relatively uniform. Figure 2b is the TEM image of a pure MoS₂ nanosheet, which confirms the stacked structure [27]. In the highly magnified TEM image (Figure 2c), the color of the MoS₂ single layer is lighter, indicating that the thickness of the nanosheet is thin [28]. The morphology and structure of the MoS₂@PDA (Figure 2d) do not have larger changes compared with MoS₂, and the thicker nanosheet layer (Figure 2e,f) illustrates the successful encapsulation of dopamine on the surface of MoS₂. The PI film displays the smooth surface morphology in Figure 2g and benefits from the ultrasonic stirring of the nanosheets before the experiment, and MoS₂ and MoS₂@PDA exhibit a flat two-dimensional structure on the surface of the PI film. Compared with MoS₂/PI film, the dispersion of MoS₂@PDA/PI is better and more uniform (Figure 2h,i). In Figure 2j, the distribution of MoS₂@PDA nanosheets can be clearly seen through the EDX-SEM mapping, and the concentrated area of Mo and S elements is consistent with the distribution of nanosheets in the SEM image.

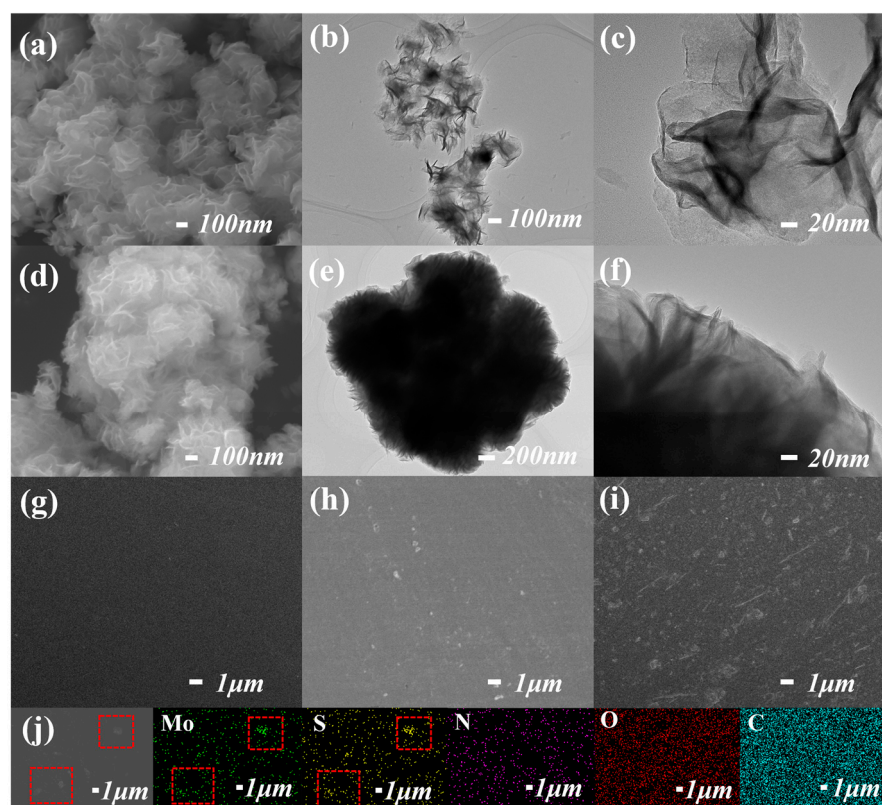


Figure 2. SEM (a) and TEM (b,c) images of MoS₂; SEM (d) and TEM (e,f) images of MoS₂@PDA; (g–i) are the SEM images of PI, MoS₂/PI, and MoS₂@PDA/PI; (j) is the SEM image and element mapping of MoS₂@PDA/PI. The red dotted frames highlight the distribution locations of MoS₂@PDA nanosheets.

3.2. Chemical Structure

XRD analysis can be used to obtain information about the structure of atoms or molecules and the composition of the sample. The XRD diffraction patterns of the five samples are shown in Figure 3a, MoS₂ and MoS₂@PDA had obvious diffraction peaks at

32.7°, 35.7°, 43.1°, and 57.7° which match with the (100), (102), (006), and (110) crystal planes of 2H-MoS₂ nanosheets, respectively [10,29]. The PDA is an amorphous structure with no obvious diffraction peaks. In addition, the pure PI, MoS₂/PI, and MoS₂@PDA/PI films have a broad peak at around 20°, indicating the presence of partial crystallization in the amorphous PI which is consistent with other literature sources [30]. The successful preparation of composite films was confirmed by the diffraction peaks of MoS₂/PI and MoS₂@PDA/PI near the (100) and (110) crystal planes of MoS₂. The faint diffraction peaks of MoS₂/PI and MoS₂@PDA/PI films are due to the low doping amount of the nanosheets. Figure 3b shows the FT-IR spectra of PI, MoS₂/PI, and MoS₂@PDA/PI. The peaks at 723 cm⁻¹ could be ascribed to the C=O bending in PI, while those absorption peaks at 1718 and 1776 cm⁻¹ could be assigned to the C=O of symmetrical stretching vibration and asymmetrical stretching vibration [31]. Due to the dehydration and cyclization of the PI precursor, there was no obvious characteristic absorption peak at about 1540 and 1645 cm⁻¹. Instead, there was a stretching vibration of -C-N-C at 1376 cm⁻¹, which indicated the amide to be replaced by the imide group [32,33]. From the above, it can be seen that imidization was almost complete, and this further proves the successful synthesis of the composite films in this research.

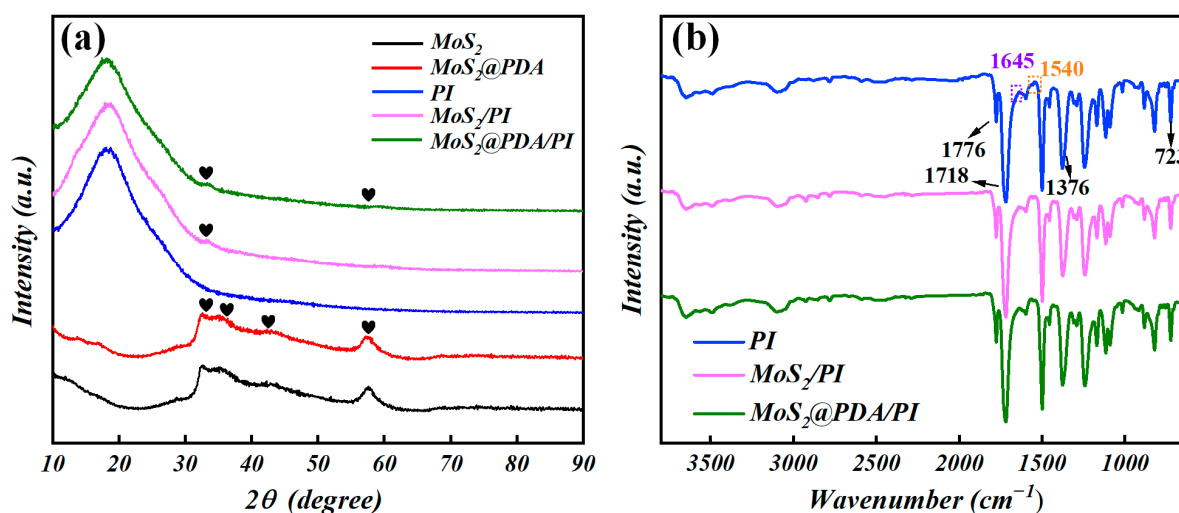


Figure 3. (a) The XRD pattern of MoS₂, MoS₂@PDA, PI, MoS₂/PI, and MoS₂@PDA/PI; (b) FT-IR spectrum of the PI, MoS₂/PI, and MoS₂@PDA/PI. The heart symbol represents the XRD diffraction peaks.

XPS characterization is a technique for analyzing the chemical valence states of elements on the surface of composite materials, and the survey spectra of MoS₂@PDA/PI composite films can be seen in Figure 4a. As Figure 4b shows, the C 1s spectrum can be deconvoluted into three diffraction peaks which are located at 284.6, 285.6, and 287.1 eV assigned to C-C/C=C, C-N, and C-O bonds, respectively [34,35]. The N 1s fine spectrum of MoS₂@PDA/PI exhibits an intense symmetrical peak centered at 399.6 eV, assigned to the imide (C-N) group. The N orbital shifts towards low energy compared to pure PI films in the literature, due to the non-covalent interaction between PI and MoS₂@PDA [14]. In Figure 4d, the O 1s spectrum is deconvoluted into two energy levels, located at the center of 531.5 and 533.1 eV, corresponding to C=O and C-O, respectively [34].

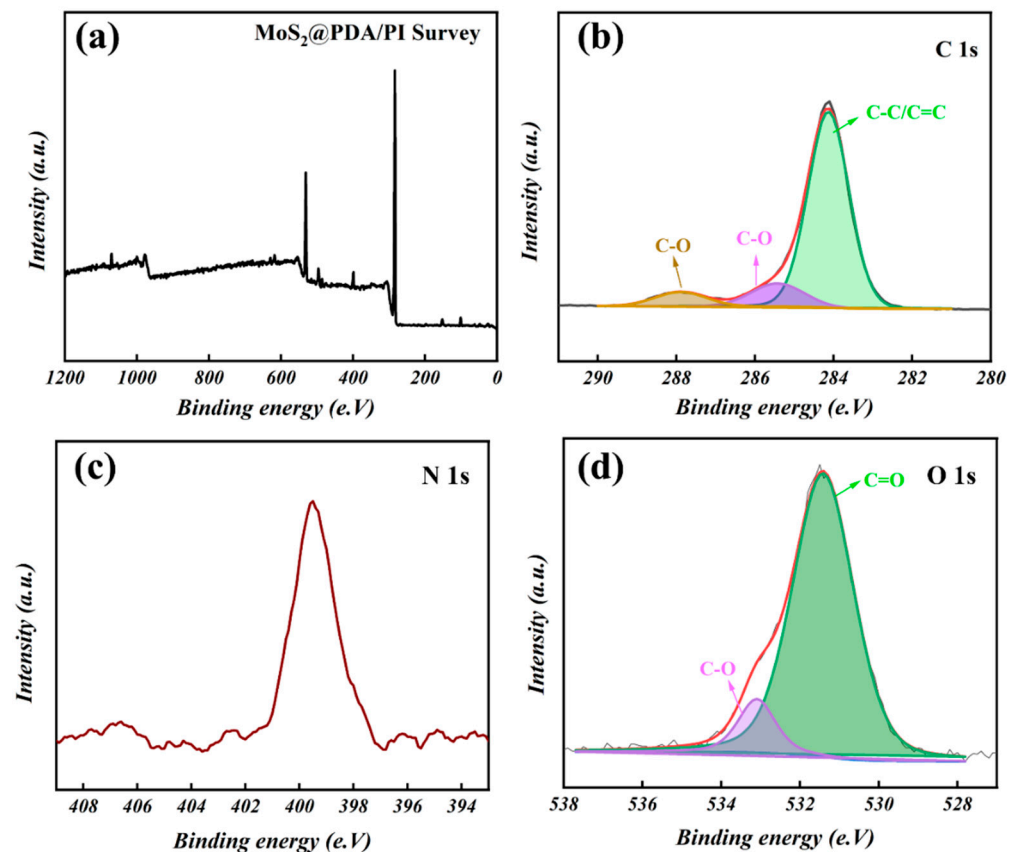


Figure 4. (a) The full spectra, (b) C 1s, (c) N 1s, and (d) O 1s of MoS₂@PDA/PI.

3.3. Mechanical Properties

The tribological properties of pure PI, MoS₂/PI, and MoS₂@PDA/PI composite films are presented in Figure 5. For the pure PI film, the friction coefficient is 0.39 and the wear rate is $1.8 \times 10^{-4} \text{ mm}^3/\text{Nm}$. When MoS₂ is incorporated, a decrease in both the friction coefficient and wear rate of the PI composite film is observed, and the values drop to 0.31 and $1.6 \times 10^{-4} \text{ mm}^3/\text{Nm}$, respectively. This reduction is attributed to the layered structure of MoS₂, which features layers interconnected by weak van der Waals forces. These layers can easily slide over each other under shear forces. In terms of wear, MoS₂ particles are found to migrate to the worn surface. They form a lubricating film, shifting wear from the composite material and its counterpart to the composite material. This shift effectively reduces the wear rate and friction coefficient of the composite. Furthermore, the excellent thermal conductivity of MoS₂ assists in dissipating heat generated during friction. This dissipation prevents heat accumulation that could cause local softening and deformation of the material, thereby enhancing tribological performance [36].

The frictional properties are further improved by modifying MoS₂ with PDA. The MoS₂@PDA/PI composites have a friction coefficient of 0.24 and a wear rate of $1.3 \times 10^{-4} \text{ mm}^3/\text{Nm}$, which are significantly reduced by 38.4% and 27.8%, respectively, compared with the pure films. The incorporation of PDA provides active sites on the MoS₂ nanosheets, improving their dispersion within the PI matrix and reducing aggregation. Therefore, the enhanced mechanical wear capacity of MoS₂@PDA/PI can be attributed to the uniform dispersion of MoS₂@PDA nanosheets in the PI matrix [37].

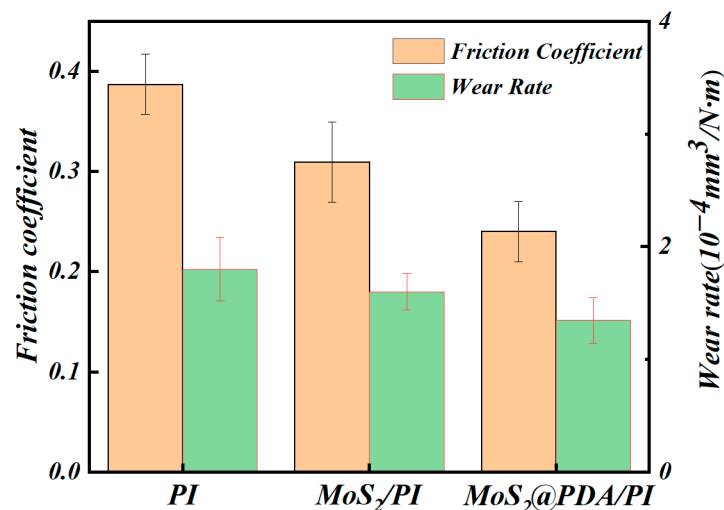


Figure 5. The friction coefficient and wear rate of PI, MoS₂/PI, and MoS₂@PDA/PI.

The tensile test results of pure PI, MoS₂/PI, and MoS₂@PDA/PI composite materials are shown in Figure 6. Among the three materials, MoS₂@PDA/PI exhibits the best mechanical properties, followed by MoS₂/PI in second place, and pure PI ranking as the least performant. Specifically, the tensile strength of the MoS₂/PI composite film increases from 101 MPa to 124.6 MPa, the elastic modulus increases from 2.0 GPa to 2.4 GPa, and the elongation at breaks increases from 21.1% to 30.2%. Notably, with the introduction of PDA-modified MoS₂, the tensile strength (146.2 MPa) of the composite material increases by 17.3% compared to MoS₂/PI. Meanwhile, the elastic modulus (2.5 GPa) of MoS₂@PDA/PI increases by 4.1%, and the elongation at break rises to 32.8% compared to MoS₂/PI. This phenomenon is primarily due to the high rigidity and layered structure of MoS₂ nanosheets, along with the interface optimization brought about by PDA modification. The increase in the rigidity and strength of the composite material is attributed to the high elastic modulus of MoS₂ and its stress dispersion and load-bearing role within the composite structure. However, the dispersion and interface compatibility of MoS₂ nanosheets in the PI matrix are limiting factors for their mechanical reinforcement effect. Building on this, the introduction of PDA not only improves the dispersion of the nanosheets but also forms cross-linked structures with active groups on their surface and the PI matrix [38]. This enhances the bonding between the nanosheets and the film, thereby improving the overall mechanical stability and durability of the composite material.

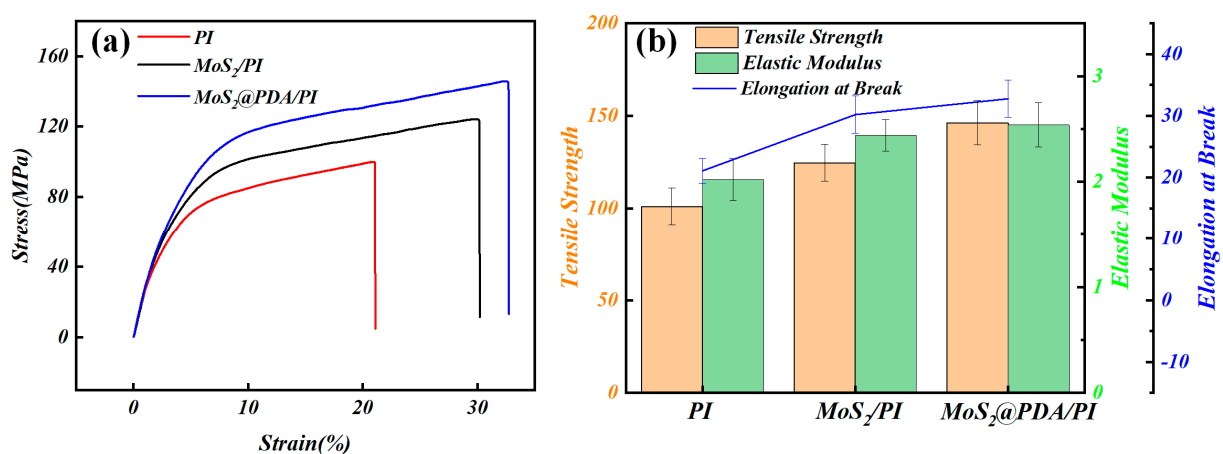


Figure 6. (a) The strain–stress curve of pure PI, MoS₂/PI, and MoS₂@PDA/PI composites; (b) tensile strength, elastic modulus, and break elongation of pure PI, MoS₂/PI, and MoS₂@PDA/PI composites.

3.4. Dielectric Properties

Figure 7 shows the variation in the dielectric constant as well as the dielectric loss angle tangent with frequency for pure PI, MoS₂/PI, and MoS₂@PDA/PI. From the figure, it can be seen that the dielectric constant of MoS₂/PI is between 4.5 and 4, while the dielectric constant of pure PI is between 3 and 3.5. This result indicates that the doping of nano-MoS₂ can significantly enhance the dielectric constant of PI films. The dielectric constant of MoS₂@PDA/PI, on the other hand, is between 3.5 and 4. It is speculated that the PDA parcel improves the interfacial bonding and compatibility between the nano-MoS₂ and the PI base material [39]. This facilitates the polarization of the dipole under the electric field and thus reduces the permittivity of the PI. Similar to the dielectric constant, the dielectric loss angle tangent is the largest for MoS₂/PI, followed by MoS₂@PDA/PI, and the smallest for pure PI. The incorporation of MoS₂ nanoparticles increases the polarization loss of the film, resulting in an increase in the angular value of the dielectric loss of MoS₂/PI [40], whereas the wrapping of PDA effectively reduces the polarization loss of the film. Thus, the angular value of dielectric loss of MoS₂@PDA/PI was reduced. This suggests that the dielectric constant and dielectric loss angle tangent of the composite film can be effectively reduced through PDA encapsulation and the composite films with superior electrical properties can be obtained. In conclusion, the overall dielectric properties of PDA-coated MoS₂-doped PI composites are better than those of MoS₂/PI.

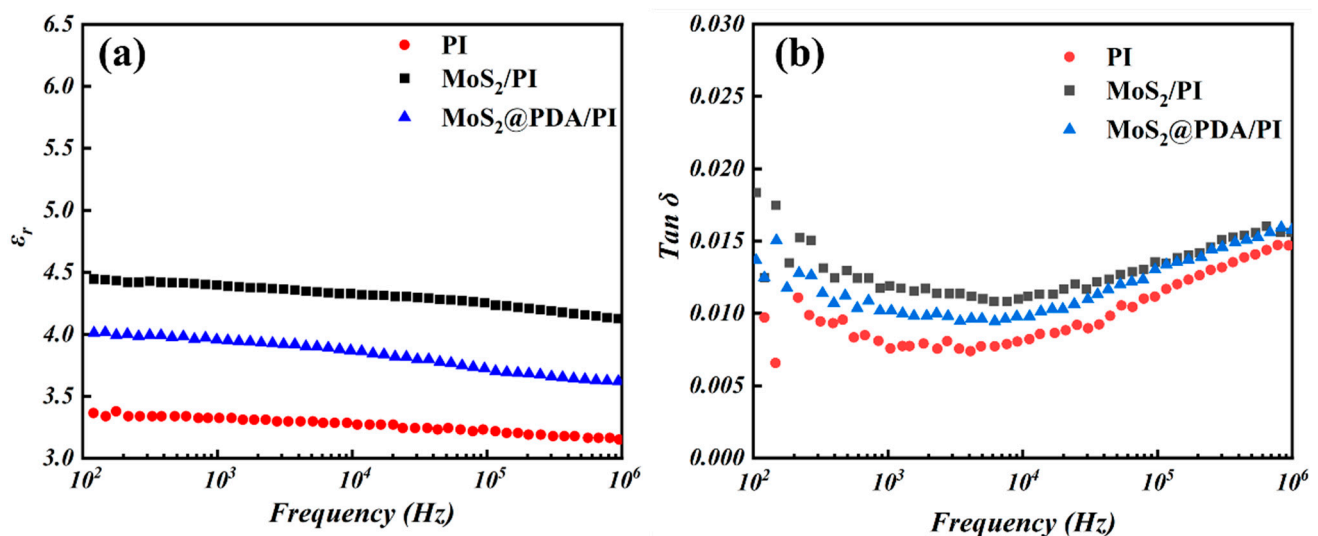


Figure 7. Dielectric properties of pure PI, MoS₂/PI, and MoS₂@PDA/PI: (a) dielectric constant and (b) dielectric loss tangent.

3.5. Electrical and Aging Characteristics

In order to investigate the breakdown and aging characteristics of composite films, we built a corresponding experimental test platform. As shown in Figure 8, the whole testbed is based on a permanent magnet direct-drive wind turbine with a capacity of 2.5 MW. During the experiment, the permanent magnet rotor of the generator is removed and only the stator part is retained. The stator part is connected to the PWM inverter via a cable of about 100 m in length. To facilitate measurement, the main insulation on the winding coils was removed before the experiment so that the first-turn conductors of the coils of each group in a generator stator winding branch in phase U were exposed. This was used as a measurement point for the voltage signals from the coils to the ground and between the coils. In addition, the insulation on the nose of the first coil near the motor terminals is removed so that the conductor of each turn of the coil is exposed as a measurement point for the inter-turn voltage signal. During measurement, the coils are numbered 1, 2, . . . , and 8 in order from terminal to neutral. The high-voltage probe used for the measurement is

the P6015A with a bandwidth of 75 MHz. During the experiment the gate voltage was set to 1.12 kV, the carrier frequency was 3 kHz, and the fundamental frequency was 35.7 Hz.

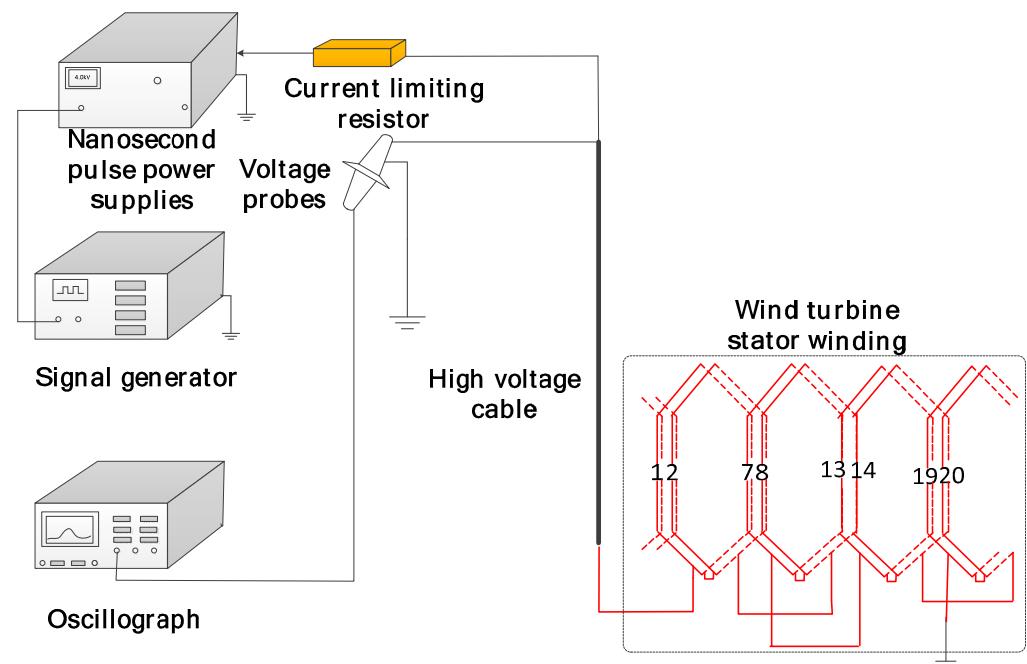


Figure 8. Structure of the pulse aging experimental device.

Figure 9 shows the Weibull distribution plots of breakdown voltage and partial discharge onset voltage for pure PI, MoS_2/PI , and $\text{MoS}_2@\text{PDA}/\text{PI}$. As can be seen from the figure, the Weibull distribution curve of the breakdown voltage of MoS_2/PI is located on the left side of pure PI and the position is very close. When the breakdown probability is 63.2%, the breakdown voltages of MoS_2/PI and pure PI films are basically the same: 17.92 kV and 19.56 kV, respectively.

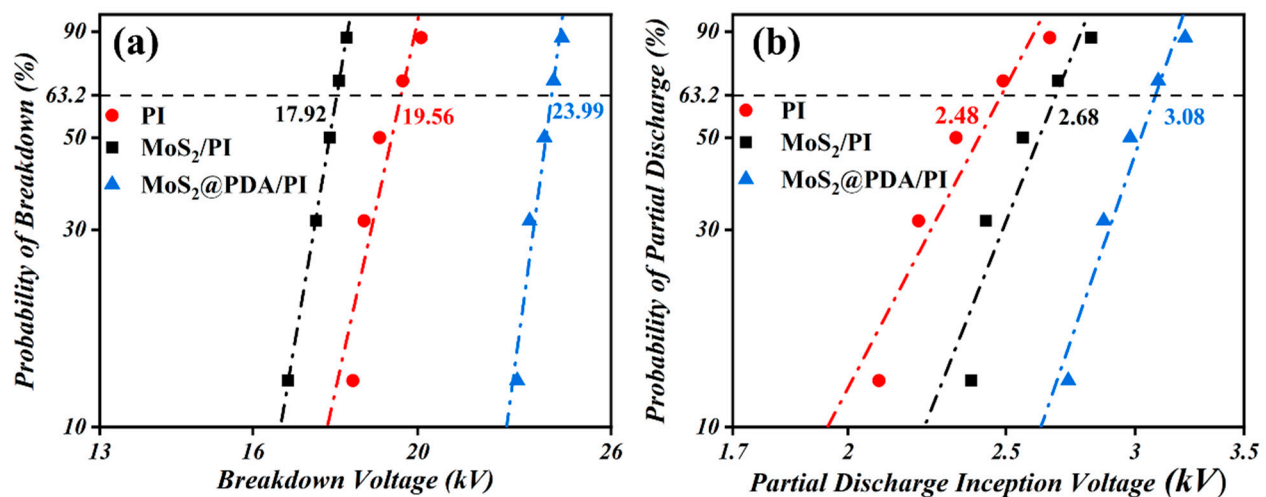


Figure 9. (a) Breakdown voltage and (b) partial discharge inception voltage of pure PI, MoS_2/PI , and $\text{MoS}_2@\text{PDA}/\text{PI}$.

The former is slightly weaker than the latter by about 9%. The Weibull distribution curve of the breakdown voltage of $\text{MoS}_2@\text{PDA}/\text{PI}$ is located on the right side of pure PI and they are farther away from each other. The breakdown voltage of PDA is 23.99 kV at 63.2% breakdown probability which is 1.23 times higher than pure PI film; this comes down

to the semiconductor characteristic of MoS₂ [41]. Under the action of a strong electric field, the electrons in the matrix are easily excited from the valence band to the conduction band and become free electrons, leading to a decrease in breakdown voltage. However, the PDA coating on the surface of MoS₂ changes the energy level of the MoS₂ surface [42]. It forms a kind of energy barrier that hinders the excitation and migration of electrons and reduces the number of free electrons, thus effectively enhancing the breakdown voltage of the composite film. Contrary to the internal breakdown voltage, the partial discharge initiation voltage of pure PI, MoS₂/PI, and MoS₂@PDA/PI increases sequentially. Specifically, for different types of nanocomposite films, the breakdown voltages of pure PI, MoS₂/PI, and MoS₂@PDA/PI with partial discharge probability of 63.2% are 2.48 kV, 2.68 kV, and 3.08 kV, respectively. MoS₂/PI and MoS₂@PDA/PI are enhanced by about 8% and 24.1%, respectively, compared with pure PI films. The reason is that MoS₂ can improve the mechanical flexibility of PI to reduce the air gap and thus enhance the corona voltage, while the semiconductor property of nano-MoS₂ will reduce the corona voltage [43]. The interaction between the two leads to a slightly stronger partial discharge onset voltage for MoS₂/PI than for pure PI. Meanwhile, the phenolic hydroxyl and amine groups in the PDA molecular structure can form stable chemical bonds and physical adsorption with the surfaces of many materials [44]. This structure is conducive to further enhancing the interfacial bonding between the composite material and the metal winding, and this tight bonding reduces the presence of air gaps and the risk of air gap discharge. Therefore, the starting discharge voltage of MoS₂@PDA/PI is enhanced due to the above reasons. In summary, MoS₂@PDA/PI has excellent electrical insulation properties compared to pure PI and MoS₂/PI, and the breakdown voltage of pure PI is higher than that of MoS₂/PI, while the partial discharge onset voltage is lower than that of MoS₂/PI.

The insulation life of pure PI, MoS₂/PI, and MoS₂@PDA/PI at different aging voltages is shown in Figure 10. From the figure, it can be seen that the insulation life of MoS₂/PI is weaker than that of pure PI at all voltage levels. Under the aging voltage of 9 kV, the insulation life of MoS₂/PI and pure PI are 224 h and 258 h, respectively. At 13 kV, the insulation life of MoS₂/PI and pure PI is 9.68 h and 11.13 h, respectively. Overall, the insulation life of pure PI is about 15% higher than that of MoS₂/PI. For MoS₂@PDA/PI, the insulation life is greater than that of pure PI and MoS₂/PI films. Its insulation life reaches 330 h at an aging voltage of 9 kV which is about 57.3% and 27.9% longer than MoS₂/PI and pure PI, respectively. The insulation life is 14.36 h at 13 kV and is about 48.3% and 29% higher than MoS₂/PI and pure PI. The main reason for this phenomenon is that pure MoS₂ and PDA-coated MoS₂ have different effects on the generation and movement of electrons in the polymer matrix. In the meantime, it has different effects on the destruction process of polymer molecular chains. In addition, the capture of electrons and holes in the trap may emit high-energy UV light [45–47]. High-energy UV light can also cause polymer chains to break. Polymer molecular chain breakage creates partial low-density regions that increase the inhomogeneity of the material; this is beneficial to the formation of discharge channels and makes the material less insulating. Thus, the lifetime of MoS₂/PI is usually shorter. When PDA is coated on the surface of MoS₂, it effectively changes the electronic properties of MoS₂ and reduces its electrical conductivity in PI materials [48,49]. This change can improve the life of the composite film. In summary, the aging insulation life of MoS₂@PDA/PI is significantly stronger than that of PI film, while PI film is stronger than MoS₂/PI.

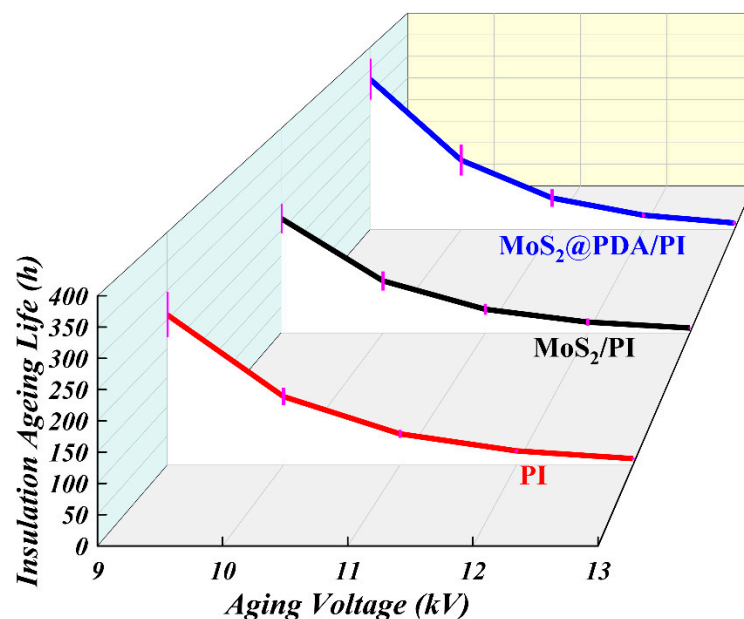


Figure 10. Insulation life of pure PI, MoS₂/PI, and MoS₂@PDA/PI under different aging voltages.

4. Conclusions

In this research, a MoS₂@PDA/PI composite film was created by adding MoS₂ nanoparticles coated with PDA into a PI matrix. In terms of mechanical properties, the friction coefficient and wear rate of the MoS₂@PDA/PI composite showed enhancements of 38.4% and 27.8%, respectively, compared to pure PI. In terms of tensile properties, there was an increase of 44.8% in tensile strength, 25.6% in elastic modulus, and 55.3% in elongation at break, relative to pure PI.

In terms of electrical insulation properties, the breakdown voltage and the partial discharge inception voltage for MoS₂@PDA/PI film increased by 22.6% and 24.1%, respectively, compared to pure PI. The long-term electrical aging tests revealed that compared to pure PI, a significant increase of 27.9% in electrical aging lifetime was exhibited by the MoS₂@PDA/PI film, attributable to its superior mechanical, tribological, and insulation properties. These results provide critical technical support for enhancing the reliability of associated electrical equipment.

Author Contributions: Conceptualization, X.C.; methodology, Z.L.; writing—original draft preparation, L.Z. and W.Z.; writing—review and editing, S.C. and C.W. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Special Support Program for High-Level Talents, grant number SO20220B06966, the National Nature Science Foundations of China, grant numbers 51977195 and 52107170, and the Major Projects of Henan Province, grant number 221100240500.

Institutional Review Board Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflicts of interest.

References

- Dayarian, S.; Yang, L.; Far, H.M. Development of polyimide aerogel stock shapes through polyimide aerogel particles. *J. Porous Mater.* **2023**, *30*, 2101–2112. [\[CrossRef\]](#)
- Zhang, D.Y.; Li, L.L.; Wang, Y.; Zhang, C.; Teng, C. Methods and strategies to decrease the dielectric properties of polyimide films: A review. *J. Sol-Gel Sci. Technol.* **2023**, *108*, 1–12. [\[CrossRef\]](#)
- Liu, Y.Y.; Wang, Y.K.; Wu, D.Y. Synthetic strategies for highly transparent and colorless polyimide film. *J. Appl. Polym. Sci.* **2022**, *139*, e52604. [\[CrossRef\]](#)

4. Li, Z.H.; Xie, Z.Q.; Han, Y.X.; Ren, H.W.; Li, Q.M.; Huang, X.W.; Wang, Z.D. Impact of nano-Al₂O₃ coating on the dielectric properties and High-Frequency surface electrical strength of the polyimide films. *Appl. Surf. Sci.* **2023**, *634*, 157666. [\[CrossRef\]](#)
5. Haq, S.U.; Jayaram, S.H.; Cherney, E.A. Evaluation of medium voltage enameled wire exposed to fast repetitive voltage pulses. *IEEE Trans. Dielectr. Electr. Insul.* **2007**, *14*, 194–203. [\[CrossRef\]](#)
6. Pauli, F.; Kilper, M.; Driendl, N.; Hameyer, K. Modeling of the Partial Discharge Process Between the Winding and the Stator of Low Voltage Machines for Traction Applications. *IEEE Trans. Energy Convers.* **2021**, *36*, 2310–2318. [\[CrossRef\]](#)
7. Li, H.G.; Yao, P.F.; Gao, Z.H.; Wang, F. Medium Voltage Converter Inductor Insulation Design Considering Grid Requirements. *IEEE J. Emerg. Sel. Top. Power Electron.* **2022**, *10*, 2339–2350. [\[CrossRef\]](#)
8. Ojaghi, M.; Sabouri, M.; Faiz, J. Diagnosis methods for stator winding faults in three-phase squirrel-cage induction motors. *Int. Trans. Electr. Energy Syst.* **2014**, *24*, 891–912. [\[CrossRef\]](#)
9. Li, J.Y.; Zhang, J.X.; Zhang, S.Z.; Ren, K.L. 2D MoS₂ Nanosheet-Based Polyimide Nanocomposite with High Energy Density for High Temperature Capacitor Applications. *Macromol. Mater. Eng.* **2021**, *306*, 2100079. [\[CrossRef\]](#)
10. Chang, Z.J.; Sun, X.L.; Liao, Z.Z.; Liu, Q.; Han, J. Design and Preparation of Polyimide/TiO₂@MoS₂ Nanofibers by Hydrothermal Synthesis and Their Photocatalytic Performance. *Polymers* **2022**, *14*, 3230. [\[CrossRef\]](#)
11. Wen, X.; He, C.; Hai, Y.Y.; Liu, X.F.; Ma, R.; Sun, J.Y.; Yang, X.; Qi, Y.L.; Chen, J.Y.; Wei, H. Fabrication of a hybrid ultrafiltration membrane based on MoS₂ modified with dopamine and polyethyleneimine. *RSC Adv.* **2021**, *11*, 26391–26402. [\[CrossRef\]](#)
12. Zhang, Y.B.; Yan, H.X.; Xu, P.L.; Guo, L.L.; Yang, K.M.; Liu, R.; Feng, W.X. A novel POSS-containing polyimide: Synthesis and its composite coating with graphene-like MoS₂ for outstanding tribological performance. *Prog. Org. Coat.* **2021**, *151*, 106013. [\[CrossRef\]](#)
13. Pi, X.H.; Sun, X.X.; Wang, R.Q.; Chen, C.L.; Wu, S.B.; Zhan, F.R.; Zhong, J.B.; Wang, Q.; Ostrikov, K.K. MoS₂ nanosheets on plasma-nitrogen-doped carbon cloth for high-performance flexible supercapacitors. *J. Colloid Interface Sci.* **2022**, *629*, 227–237. [\[CrossRef\]](#) [\[PubMed\]](#)
14. Yuan, H.; Liu, X.H.; Ma, L.M.; Yang, Z.G.; Wang, H.G.; Wang, J.Q.; Yang, S.R. Application of two-dimensional MoS₂ nanosheets in the property improvement of polyimide matrix: Mechanical and thermal aspects. *Compos. Part A Appl. Sci. Manuf.* **2017**, *95*, 220–228. [\[CrossRef\]](#)
15. Liu, S.T.; Dong, C.L.; Yuan, C.Q.; Bai, X.Q.; Tian, Y.; Zhang, G.L. A new polyimide matrix composite to improve friction-induced chatter performance through reducing fluctuation in friction force. *Compos. Part B Eng.* **2021**, *217*, 108887. [\[CrossRef\]](#)
16. Guo, Y.G.; Fang, C.Y.; Wang, T.M.; Wang, Q.H.; Song, F.Z.; Wang, C. Tribological Behavior of Cotton Fabric/Phenolic Resin Laminated Composites Reinforced with Two-Dimensional Materials. *Polymers* **2023**, *15*, 4454. [\[CrossRef\]](#)
17. Xianhai, P.; Boyan, J.; Zhiheng, S.; Yuyou, L.; Shixin, X.; Tao, L. Influence of Grease on Characteristics of Gas Decomposition Product After Breaking Current of SF₆ Circuit Breaker. In Proceedings of the 2019 5th International Conference on Electric Power Equipment—Switching Technology (ICEPE-ST), Kitakyushu, Japan, 13–16 October 2019.
18. Zhou, K.Q.; Liu, J.J.; Wen, P.Y.; Hu, Y.; Gui, Z. A noncovalent functionalization approach to improve the dispersibility and properties of polymer/MoS₂ composites. *Appl. Surf. Sci.* **2014**, *316*, 237–244. [\[CrossRef\]](#)
19. Zhang, A.T.; Li, A.H.; Tian, W.X.; Li, Z.C.; Wei, C.; Sun, Y.; Zhao, W.; Liu, M.L.; Liu, J.Q. A Target-Directed Chemo-Photothermal System Based on Transferrin and Copolymer-Modified MoS₂ Nanoplates with pH-Activated Drug Release. *Chem.-Eur. J.* **2017**, *23*, 11346–11356. [\[CrossRef\]](#)
20. Xu, B.B.; Su, Y.Y.; Li, L.; Liu, R.; Lv, Y. Thiol-functionalized single-layered MoS₂ nanosheet as a photoluminescence sensing platform via charge transfer for dopamine detection. *Sens. Actuators B Chem.* **2017**, *246*, 380–388. [\[CrossRef\]](#)
21. Gu, B.; Pu, G.H.; Ding, B.N.; Zhang, K.; He, R.; Fan, J.H.; Xing, T.; Wu, J.Y.; Yang, W.B. Improved interfacial bonding strength of silicone rubber/carbon fiber modified by dopamine. *Polym. Compos.* **2022**, *43*, 6975–6986. [\[CrossRef\]](#)
22. Chen, Y.W.; Zhan, Y.Q.; Dong, H.Y.; Li, Y.L.; Yang, X.L.; Sun, A.; Chen, X.M.; Zhu, F.; Jia, H.S. Two-dimensional lamellar MXene nanosheets/waterborne epoxy composite coating: Dopamine triggered surface modification and long-term anticorrosion performance. *Colloids Surf. A* **2023**, *674*, 131865. [\[CrossRef\]](#)
23. Zhu, W.Q.; Lin, Y.L.; Kang, W.W.; Quan, H.Y.; Zhang, Y.Y.; Chang, M.L.; Wang, K.; Zhang, M.; Zhang, W.B.; Li, Z.Q.; et al. An aerogel adsorbent with bio-inspired interfacial adhesion between graphene and MoS₂ sheets for water treatment. *Appl. Surf. Sci.* **2020**, *512*, 145717. [\[CrossRef\]](#)
24. Dong, Y.J.; Wang, Z.Y.; Huo, S.C.; Lin, J.; He, S.J. Improved Dielectric Breakdown Strength of Polyimide by Incorporating Polydopamine-Coated Graphitic Carbon Nitride. *Polymers* **2022**, *14*, 385. [\[CrossRef\]](#) [\[PubMed\]](#)
25. Feng, M.N.; Ma, D.Y.; Chen, X.R.; Wang, P.; Lin, J. Enhanced dielectric properties of poly (arylene ether nitrile) composite films employing MoS₂-based semiconductors for organic film capacitor applications. *Mater. Sci. Semicond. Process.* **2021**, *136*, 106127. [\[CrossRef\]](#)
26. Sanusi Hamat, M.R.I.; Salit, M.S.; Yidris, N.; Ali, S.A.S.; Hussin, M.S.; Manan, M.S.A.; Suffin, M.Q.Z.A.; Ibrahim, M.; Khalil, A.N.M. The Effects of Self-Polymerized Polydopamine Coating on Mechanical Properties of Polylactic Acid (PLA)-Kenaf Fiber (KF) in Fused Deposition Modeling (FDM). *Polymers* **2023**, *15*, 2525.
27. Liu, C.B.; Wang, L.L.; Tang, Y.H.; Luo, S.L.; Liu, Y.T.; Zhang, S.Q.; Zeng, X.Y.; Xu, Y.Z. Vertical single or few-layer MoS₂ nanosheets rooting into TiO₂ nanofibers for highly efficient photocatalytic hydrogen evolution. *Appl. Catal. B Environ.* **2015**, *164*, 1–9. [\[CrossRef\]](#)

28. Egerton, R.F.; Hayashida, M.; Malac, M. Transmission electron microscopy of thick polymer and biological specimens. *Micron* **2023**, *169*, 103449. [\[CrossRef\]](#)
29. He, J.Q.; Sun, J.L.; Meng, Y.N.; Pei, Y. Superior lubrication performance of MoS₂-Al₂O₃ composite nanofluid in strips hot rolling. *J. Manuf. Process.* **2020**, *57*, 312–323. [\[CrossRef\]](#)
30. Zhang, S.Z.; Wang, J.; Wang, Z.; Xu, G.Y.; Jiang, Y.G.; Xiao, Y.Y.; Ding, F. Reusable Nanoporous Al₂O₃-Containing Polyimide Aerogels for Thermal Insulation of Aircraft. *ACS Appl. Nano Mater.* **2023**, *6*, 15925–15936. [\[CrossRef\]](#)
31. Zhang, P.P.; Zhao, J.P.; Zhang, K.; Bai, R.; Wang, Y.M.; Hua, C.X.; Wu, Y.Y.; Liu, X.X.; Xu, H.B.; Li, Y. Fluorographene/polyimide composite films: Mechanical, electrical, hydrophobic, thermal and low dielectric properties. *Compos. Part A Appl. Sci. Manuf.* **2016**, *84*, 428–434. [\[CrossRef\]](#)
32. Wan, B.Q.; Zheng, M.H.; Yang, X.; Dong, X.D.; Li, Y.C.; Mai, Y.W.; Chen, G.; Zha, J.W. Recyclability and Self-Healing of Dynamic Cross-Linked Polyimide with Mechanical/Electrical Damage. *Energy Environ. Mater.* **2023**, *6*, e12427. [\[CrossRef\]](#)
33. Luo, J.W.; Wang, Y.; Qu, Z.J.; Wang, W.; Yu, D. Anisotropic, multifunctional and lightweight CNTs@CoFe₂O₄/polyimide aerogels for high efficient electromagnetic wave absorption and thermal insulation. *Chem. Eng. J.* **2022**, *442*, 136388. [\[CrossRef\]](#)
34. Wang, C.X.; Liu, Y.; Jia, Z.R.; Zhao, W.R.; Wu, G.L. Multicomponent Nanoparticles Synergistic One-Dimensional Nanofibers as Heterostructure Absorbers for Tunable and Efficient Microwave Absorption. *Nano-Micro Lett.* **2023**, *15*, 2311–6706. [\[CrossRef\]](#) [\[PubMed\]](#)
35. Hou, T.Q.; Jia, Z.R.; Dong, Y.H.; Liu, X.H.; Wu, G.L. Layered 3D structure derived from MXene/magnetic carbon nanotubes for ultra-broadband electromagnetic wave absorption. *Chem. Eng. J.* **2022**, *431*, 133919. [\[CrossRef\]](#)
36. Du, J.; Pu, C.Z.; Sun, X.Y.; Wang, Q.; Niu, H.Q.; Wu, D.Z. Preparation and Interfacial Properties of Hydroxyl-Containing Polyimide Fibers. *Polymers* **2023**, *15*, 1032. [\[CrossRef\]](#)
37. Yang, L.; Xu, W.; Shi, X.L.; Wu, M.L.; Yan, Z.Y.; Zheng, Q.; Feng, G.N.; Zhang, L.; Shao, R. Investigating the thermal conductivity and flame-retardant properties of BN/MoS₂/PCNF composite film containing low BN and MoS₂ nanosheets loading. *Carbohydr. Polym.* **2023**, *311*, 120621. [\[CrossRef\]](#)
38. Yang, X.; Zheng, X.H.; Zhang, T.; Chen, H.S.; Liu, M. Experimental study on thermal conductivity and rectification of monolayer and multilayer MoS₂. *Int. J. Heat Mass Transf.* **2021**, *170*, 121013. [\[CrossRef\]](#)
39. Lv, S.Q.; Xu, W.; Huang, W.; Lv, G.C.; Wang, G.S. Microwave absorption enhancement by adjusting reactant ratios and filler contents based on 1D K-MnO₂@PDA and poly(vinylidene fluoride) matrix. *RSC Adv.* **2019**, *9*, 13088–13095. [\[CrossRef\]](#)
40. Wang, X.Y.; Zhu, T.; Chang, S.C.; Lu, Y.K.; Mi, W.B.; Wang, W. 3D Nest-like Architecture of Core-Shell CoFe₂O₄@1T/2H-MoS₂ Composites with Tunable Microwave Absorption Performance. *ACS Appl. Mater. Interfaces* **2020**, *12*, 11252–11264. [\[CrossRef\]](#) [\[PubMed\]](#)
41. Patra, K.K.; Ghosal, M.K.; Bajpai, H.; Raj, S.; Gopinath, C.S. Oxidative Disproportionation of MoS₂/GO to MoS₂/MoO₃-x/RGO: Integrated and Plasmonic 2D-Multifunctional Nanocomposites for Solar Hydrogen Generation from Near-Infrared and Visible Regions. *J. Phys. Chem. C* **2019**, *123*, 21685–21693. [\[CrossRef\]](#)
42. Yuan, L.; Wen, T.P.; Jiang, L.Y.; Liu, Z.L.; Tian, C.; Yu, J.K. Modified superhydrophilic/underwater superoleophobic mullite fiber-based porous ceramic for oil-water separation. *Mater. Res. Bull.* **2021**, *143*, 111454. [\[CrossRef\]](#)
43. Yu, L.L.; Lee, Y.H.; Ling, X.; Santos, E.J.G.; Shin, Y.C.; Lin, Y.X.; Dubey, M.; Kaxiras, E.; Palacios, T. Graphene/MoS₂ Hybrid Technology for Large-Scale Two-Dimensional Electronics. *Nano Lett.* **2014**, *14*, 3055–3063. [\[CrossRef\]](#)
44. Ma, S.S.; Li, H.J.; Li, C.; Tian, H.C.; Tao, M.X.; Fei, J.; Qi, L.H. Metal-organic frameworks/polydopamine synergistic interface enhancement of carbon fiber/phenolic composites for promoting mechanical and tribological performances. *Nanoscale* **2021**, *13*, 2234–2247. [\[CrossRef\]](#)
45. Sun, S.X.; Jiao, T.F.; Xing, R.R.; Li, J.H.; Zhou, J.X.; Zhang, L.X.; Peng, Q.M. Preparation of MoS₂-based polydopamine-modified core-shell nanocomposites with elevated adsorption performances. *RSC Adv.* **2018**, *8*, 21644–21650. [\[CrossRef\]](#)
46. Sun, W.F.; Chern, W.K.; Chan, J.C.Y.; Chen, Z. A Reactive Molecular Dynamics Study on Crosslinked Epoxy Resin Decomposition under High Electric Field and Thermal Aging Conditions. *Polymers* **2023**, *15*, 765. [\[CrossRef\]](#)
47. Lv, Y.; Jin, Y.H.; Li, Z.Z.; Zhang, S.A.; Wu, H.Y.; Xiong, G.T.; Ju, G.F.; Chen, L.; Hu, Y.H. Reversible photoluminescence switching in photochromic material Sr₆Ca₄(PO₄)₆F₂:Eu²⁺ and the modified performance by trap engineering via Ln³⁺ (Ln = La, Y, Gd, Lu) co-doping for erasable optical data storage. *J. Phys. Chem. C* **2020**, *8*, 6403–6412. [\[CrossRef\]](#)
48. Luque, A.; González, M.; Gordillo-Vázquez, F.J. Streamer discharges as advancing imperfect conductors: Inhomogeneities in long ionized channels. *Plasma Sources Sci. Technol.* **2017**, *26*, 125006. [\[CrossRef\]](#)
49. Zhang, X.Y.; Yi, J.; Yin, Y.A.; Song, Y.H.; Xiong, C.X. Thermal conductivity and electrical insulation properties of h-BN@PDA/silicone rubber composites. *Diam. Relat. Mater.* **2021**, *117*, 108485. [\[CrossRef\]](#)

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.