

Electric-field-coupled rotary DBD plasma-modified mBN enables epoxy encapsulation with both high thermal conductivity and excellent electrical insulation: high material utilization index

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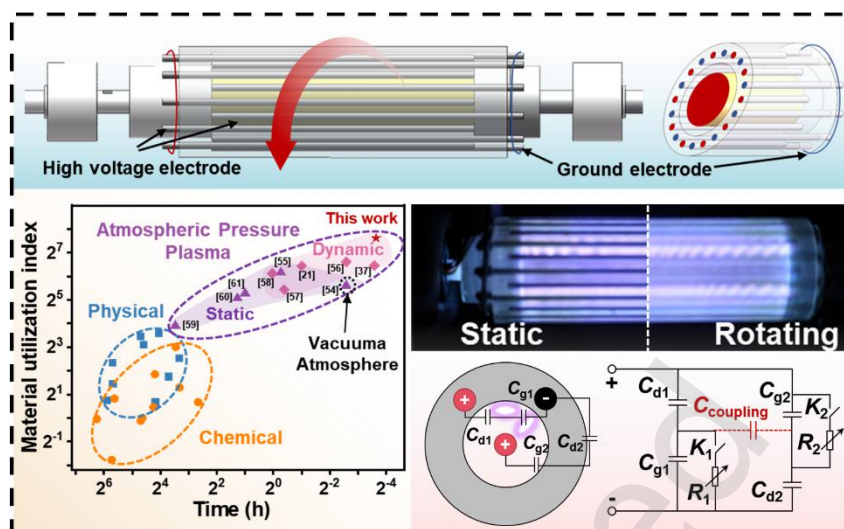
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An electric-field-coupled rotary DBD reactor is developed to achieve efficient and uniform plasma surface hydroxylation of boron nitride microplatelets (mBN). The modified mBN enables epoxy composites with enhanced thermal conductivity and recovered electrical insulation, while the proposed Material Utilization Index provides a comparative metric for evaluating modification efficiency.

Electric-field-coupled rotary DBD plasma-modified mBN enables epoxy encapsulation with both high thermal conductivity and excellent electrical insulation: high material utilization index

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ABSTRACT: The rapid development of artificial intelligence data centers has driven the demand for epoxy (EP)-based encapsulation composites with high thermal conductivity and electrical insulation for high-power-density power electronics. However, poor interfacial compatibility between boron nitride microplatelets (mBN) and EP generates microvoids and local electric-field distortion. Here, an electric-field-coupled rotary dielectric barrier discharge reactor is developed for uniform plasma hydroxylation of mBN using water as a green reaction medium. The coupling of surface and volume discharges with dynamic rotation suppresses powder agglomeration and enables homogeneous surface functionalization. The resulting mBN-OH/EP composite exhibits a thermal conductivity of 0.472 W/(m·K), representing a 105.2% enhancement over pure EP, and a breakdown strength of 120.02 kV/mm, 14.48% higher than that of untreated mBN/EP. Improved interfacial compatibility also alleviates local electric-field distortion and reduces the apparent partial-discharge charge by approximately 43%, achieving a favorable balance between thermal conductivity and electrical insulation. Furthermore, a Material Utilization Index (MUI) is introduced to quantify material and energy efficiency. The proposed method exhibits higher utilization efficiency than conventional physical, chemical, and static plasma modification approaches, providing a green and potentially scalable strategy for preparing high-performance thermally conductive and electrically insulating composites.

KEYWORDS: boron nitride microplatelets, rotary dielectric barrier discharge, plasma hydroxylation, epoxy composites, thermal conductivity, electrical insulation

1 Introduction

Driven by the rapid development of artificial intelligence (AI) data centers and the emerging 800 VDC power distribution architecture, high-power-density power electronic devices, represented by solid-state transformers (SSTs), impose increasingly stringent requirements on the heat dissipation and electrical insulation properties of epoxy (EP)-based encapsulation materials[1-4]. Owing to its superior intrinsic thermal conductivity and electrical insulation, hexagonal boron nitride (h-BN) is regarded as a highly promising thermally conductive filler. In particular, boron nitride microplatelets (mBN), exfoliated through processes such as ball milling, exhibit significant potential for constructing efficient thermally conductive networks due to their high aspect ratio. Furthermore, their advantages of high yield and low cost in manufacturing demonstrate broad prospects for industrial applications[5, 6]. However, mBN exhibits strong surface chemical inertness, resulting in poor interfacial compatibility with the organic EP matrix[7, 8]. Additionally, its large specific surface area makes the powder highly susceptible to agglomeration within the resin.

This agglomeration tends to introduce interfacial defects and microvoids, which lead to local electric-field distortion and a decline in breakdown strength, thereby making the composites fail to meet the requirements for high-reliability insulation[9, 10]. Therefore, simultaneously improving thermal conductivity while maintaining or even enhancing insulation reliability remains a critical challenge for the safe and efficient operation of next-generation energy infrastructures.

To prevent insulation deterioration induced by the polarity mismatch between inorganic fillers and the organic matrix, surface modification of the fillers is essential to repair interfacial defects. Conventional modification approaches primarily include physical methods (such as mechanical ball milling and vapor deposition) and chemical methods (such as wet chemical grafting and coupling agent treatment)[11-14]. While these methods exhibit certain effectiveness, they are hindered by inherent limitations: physical methods often involve complex procedures, whereas chemical approaches entail excessive solvent consumption, toxic byproducts, and intensive energy inputs, severely contradicting the principles of sustainable

manufacturing. As a dry, eco-friendly, and highly efficient surface activation technique, low-temperature plasma technology has attracted widespread attention in recent years[15, 16]. By utilizing high-energy reactive species to introduce polar functional groups onto the material surface, this technology enables the precise regulation of interfacial properties without compromising the intrinsic bulk properties of the material. Furthermore, it offers significant advantages, including high selectivity, rapid conversion rates, and energy-efficient, environmentally friendly characteristics[17-20]. Among various plasma generation methods, dielectric barrier discharge (DBD) represents the primary approach for generating large-area, uniform plasma at atmospheric pressure, making it particularly suitable for material surface treatments.

However, applying DBD technology to the surface modification of powder materials still poses unique challenges. Due to their small particle size and light weight, powders are highly susceptible to floating; furthermore, their high specific surface energy renders them prone to powder agglomeration. Consequently, achieving sufficient and uniform modification of the powder remains difficult. To enhance treatment uniformity, researchers initially attempted to combine static parallel-plate DBD setups with mechanical stirring[21]. For instance, Wu et al.[22]utilized a DBD reactor with a parallel-plate electrode structure, where nano- Al_2O_3 powders were spread evenly across the dielectric barrier. After plasma discharge, the powders were manually stirred and re-leveled, a process repeated multiple times to ensure treatment uniformity. Although this method effectively activates the nanoparticle surfaces to enhance their adsorption capacity for silane coupling agents—thereby improving particle dispersion within the polymer matrix—it is inherently limited by the restricted penetration depth of the plasma and the inevitable formation of "dead zones" during stirring. As a result, uniform and high-throughput treatment of bulk powders remains challenging. To achieve dynamic processing, researchers adopted fluidized-bed technology from the field of chemical engineering, using gas flow to fluidize the powder and thereby increase the contact area between the particles and the plasma. For example, García- Guerrero et al.[23] employed a coaxial DBD fluidized-bed reactor to treat TiO_2 powders, successfully grafting hydrophobic functional groups onto the powder surfaces to enhance hydrophobicity and accelerate their sedimentation rate in water. While feasible for small-scale applications, industrial scaling of this method demands an excessively high flow rate of inert gas to fluidize bulk powders, which incurs exorbitant operational costs and introduces operational risks such as mesh and pipe clogging. As an alternative improvement, rotary-drum DBD systems utilize surface discharges combined with mechanical rotation, which avoids massive gas consumption and effectively mitigates powder agglomeration. Akram et al. [24] employed a rotary-drum DBD apparatus for atmospheric-pressure plasma treatment of polyvinylidene fluoride (PVDF) powders. They successfully coated the powder surfaces uniformly with the conductive polymer PEDOT and established C-S-C chemical bonds, significantly enhancing the interfacial bonding after compounding with

epoxy. However, because the active region of surface discharge is heavily confined near the inner wall of the reactor, the powder is only treated when in direct contact with the wall. This limitation leads to insufficient overall treatment efficiency and modification depth, failing to satisfy the demands for high-throughput industrial production. Consequently, simultaneously enhancing the discharge intensity, energy utilization efficiency, and processing capacity while ensuring dynamic rolling agitation of the powders remains a critical challenge for the application of DBD technology in powder modification. In our previous work, we developed a static three-electrode coaxial DBD reactor that utilized a combined AC and pulsed power supply to achieve electric-field coupling at specific phases[25]. This configuration significantly intensified the plasma discharge and achieved an exceptionally high reduction efficiency for graphene oxide, demonstrating that electric-field coupling is a highly effective route for boosting plasma energy efficiency.

Motivated by the aforementioned analysis, this study innovatively designs an electric-field-coupled rotary DBD reactor by combining a highly efficient electric-field coupling mechanism with a dynamic rotary-drum structure. Through an optimized electrode array design, surface dielectric barrier discharge (SDBD) and volume dielectric barrier discharge (VDBD) are coupled to generate a composite surface-volume dielectric barrier discharge (S-VDBD), which enhances the discharge intensity and spatial uniformity. Meanwhile, the dynamic rotary-drum structure continuously renews the exposure of powder particles to the plasma, thereby improving the uniformity of powder surface treatment. In this work, mBN is selected as the target powder, and water is employed as a green reaction medium to achieve surface hydroxylation. The modification efficacy of this novel DBD reactor is systematically investigated. The results demonstrate that the plasma-activated water medium effectively facilitates the surface hydroxylation of mBN, introducing oxygen-containing functional groups and establishing B-O related chemical bonds while preserving the crystalline structural integrity of h-BN. Consequently, the thermal conductivity of the prepared mBN-OH/EP composite reaches 0.472 W/(m·K), representing a 105.2% enhancement compared with pure EP. Simultaneously, the breakdown strength reaches at 120.02 kV/mm, corresponding to a 14.48% improvement over the untreated mBN/EP composite. Phase-resolved partial discharge (PRPD) analyses further reveal that interfacial defects are mitigated after modification; this effectively suppresses local partial discharge activities, leading to an approximately 43% reduction in the apparent charge of partial discharge. Interestingly, by employing water—conventionally perceived as a conductive medium—as the reaction medium, we counterintuitively achieve an enhancement in the electrical insulation of the composite. Furthermore, to quantitatively assess the process efficiency, a Material Utilization Index (MUI) is introduced to comprehensively evaluate material consumption and energy utilization. The results indicate that the MUI of the proposed strategy is substantially higher than those of conventional physical and chemical methods, as well as traditional static plasma

treatments, thereby maximizing the overall treatment efficiency.

2 Experimental

2.1 Experimental materials

EP and its curing agent (D230) were purchased from Kunshan Jiulimei Electronic Materials Co., Ltd. h-BN powder, with a particle size of 5–10 μm and a purity of 99%, was obtained from Shanghai Macklin Biochemical Co., Ltd. Argon gas (Ar, 99.999% purity) was supplied by Nanjing Special Gas Factory Co., Ltd. Isopropanol (IPA, 99.9% purity) was purchased from Shanghai Titan Scientific Co., Ltd.

2.2 Preparation of mBN

The raw h-BN powder and ZrO_2 milling balls (with diameters of 1 mm, 2 mm, and 3 mm) were mixed at a ball-to-powder weight ratio of 100:1. IPA was added to occupy one-third of the milling jar's volume (approximately 20–30 mL), ensuring that both the powder and the balls were completely submerged. Mechanical exfoliation was achieved by milling the mixture in a planetary ball mill at a rotation speed of 400 rpm for 24 h. Subsequently, the milled mixture was centrifuged at 8000 rpm for 5 min. The resulting product was dried in an oven at 60 °C for 24 h to remove the residual solvent, yielding the final dried mBN product (as shown in **Figure S1a**). These systematic mechanical ball-milling procedures effectively facilitated the interlayer exfoliation of boron nitride, laying a solid foundation for subsequent material applications.

2.3 Experimental setup and procedure

The experimental platform consists of an electric-field-coupled rotary DBD reactor, a nanosecond pulsed power supply, an electrical characteristic detection system, and a gas flow control system (**Figure 1a**). A photograph of the actual platform is shown in **Figure 1b**. The main body of the reactor is a cylindrical quartz glass tube (length 100 mm, outer diameter 36 mm, inner diameter 22 mm, wall thickness 7 mm), with 20 tungsten rod electrodes (diameter 2 mm, length 100 mm) evenly embedded along the circumference of the tube wall. A copper rod (diameter 16 mm, length 90 mm) serving as the central electrode is fixed at the center of the cylinder via specially designed fixing plugs (**Figure 1c**, **Figure S2**). Ten tungsten rods on one side were connected in parallel with the central electrode to the nanosecond pulsed high-voltage source, while the remaining ten tungsten rods on the opposite side were grounded. Consequently, a SDBD was generated along the tube wall, and a VDBD was generated between the central electrode and the grounded tungsten rods, thereby realizing a composite S-VDBD. The reactor was driven by an electric motor, while a through-bore conductive slip ring was employed to maintain the electrical connection to the electrodes during continuous rotation. During treatment, the rotation caused the powder to continuously tumble and redistribute within the discharge region.

The nanosecond pulsed power supply utilized in the experimental platform (Model ADW-20K10A) featured an adjustable voltage amplitude range of 0–30 kV, a frequency range of 1–10 kHz, and adjustable rise and fall times ranging from 50 to 250 ns. Stable discharges could be achieved

within the voltage amplitude range of 3–9 kV and the frequency range of 3–6 kHz. For these experiments, the optimal parameters were set to a voltage of 7 kV, a frequency of 4 kHz, and rise and fall times of 50 ns.

The gas flow control system (Sevenstar D08-4F) consisted of a mass flow controller and a flow indicator, with a control precision reaching 1 mL/min. Argon was selected as the working gas and introduced into the gas mixing chamber through one channel of the flow controller. The gas flow rate was adjustable from 0 to 10 L/min, with an optimized rate of 0.5 L/min. Water (H_2O) was chosen as the reaction medium for powder surface modification. Using a bubbling method, a low-flow-rate argon stream carried the H_2O vapor through another channel of the flow controller into the mixing chamber. Subsequently, this mixture was introduced into the reactor's gas gap space alongside the working gas. The flow rate of the medium could be adjusted within the range of 0–200 mL/min, and an optimal flow rate of 125 mL/min was selected[21]. Prior to igniting the discharge, the gas mixture was allowed to flow into the reactor for 1 min to purge the residual air, thereby eliminating its interference with the discharge.

The voltage amplitude applied across the high-voltage and grounded electrodes was measured using a Tektronix P6015A high-voltage probe. The current flowing through the grounded end was monitored utilizing a Pearson current monitor. Experimental waveforms and data were recorded using a Tektronix MDO34 oscilloscope, and optical emission spectra were captured using an Ocean Optics HR4000CG spectrometer.

2.4 Preparation of the Composites

The modified mBN and EP were thoroughly mixed via mechanical stirring according to the designed filler loading, followed by vacuum degassing for 30 min. Subsequently, the curing agent D230 was added at a stoichiometric ratio, and the mixture was uniformly blended using a Thinky mixer. The resulting slurry was poured into molds and transferred to a drying oven for isothermal curing at 80 °C for 4 h. After cooling naturally to room temperature, the composite samples were demolded. The detailed preparation procedure is illustrated in **Figure S1b**.

2.5 Material Characterization

The microscopic surface morphology of the samples was characterized using a thermal field emission scanning electron microscope (FE-SEM, Ultra55, Zeiss) equipped with an energy-dispersive X-ray spectroscopy (EDS) system. Prior to observation, the samples were sputter-coated with gold. The surface chemical composition and crystal structure were analyzed via Fourier transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), and X-ray diffraction (XRD). FTIR spectra were recorded using a Nicolet iS50 FTIR spectrometer (Thermo Fisher Scientific) with a resolution of 4 cm^{-1} over a wavenumber range of 400–4000 cm^{-1} . XPS measurements were performed on an ESCALAB 250XI system (Thermo Scientific) equipped with a monochromatic Al $K\alpha$ X-ray source (1486.6 eV), acquiring detailed spectra at a pass energy of 100 eV and a resolution of 1 eV. The XPS results were deconvoluted and fitted using XPSpeak41 software to determine the elemental

composition ratios. XRD patterns were collected using a SmartLab SE diffractometer (Rigaku, Japan) within a scanning range of 20°–80°, utilizing a step size of 0.02° and a scanning rate of 10 °/min. Thermal stability was evaluated by thermogravimetric analysis (TGA) using a NETZSCH STA 449F5 analyzer. The samples were heated from room temperature to 800 °C at a heating rate of 10 °C/min in an air atmosphere.

2.6 Performance Testing

Thermal conductivity was measured using a DRL-V thermal conductivity tester (Xiangyi). The breakdown

strength was determined by conducting power-frequency alternating current breakdown tests on the specimens. The tests employed a sphere-sphere electrode configuration, with both the samples and electrodes immersed in transformer oil to prevent surface flashover. A voltage ramp rate of 2 kV/s was applied. For each group, 15 replicate measurements were performed, and the resulting data were analyzed using a Weibull distribution. The partial discharge characteristics were evaluated using an HZJF-124 partial discharge detector to measure the apparent charge of partial discharge. Furthermore, the surface temperature rise was recorded utilizing a Fluke TiX580 infrared thermal imager.

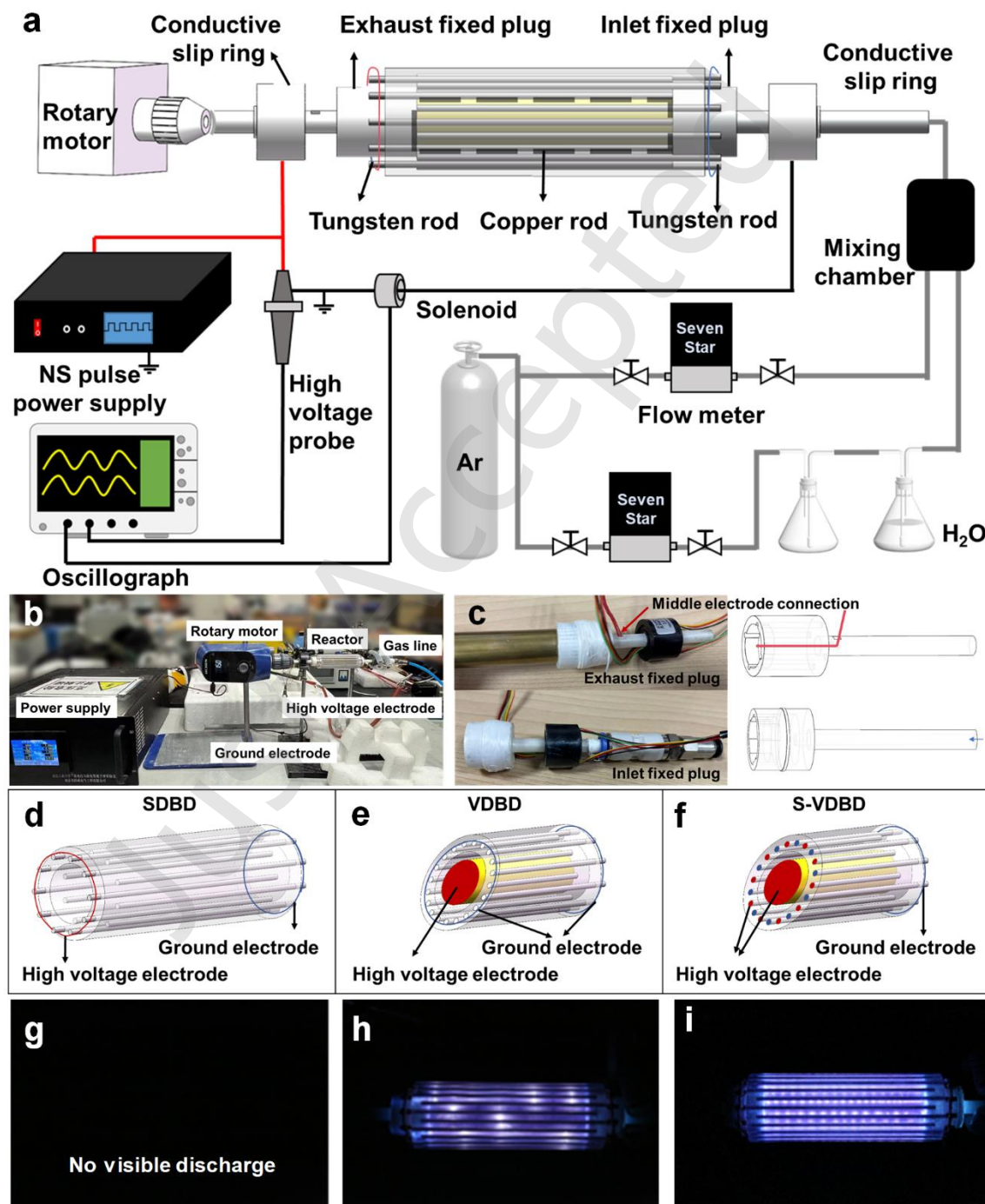


Figure 1. (a) Schematic diagram of the plasma treatment platform. (b) Photograph of the platform. (c) Photograph and perspective view of the fixing plug. Electrode wiring configurations (d–f) and the corresponding discharge optical emission images (g–i) of SDBD, VDBD, and S-VDBD under discharge parameters of 7 kV and 4 kHz.

3 Results and discussion

3.1 Optimization of plasma discharge performance

The intensity and spatial uniformity of the plasma discharge directly determine the efficiency and consistency of powder surface modification. Under identical nanosecond pulsed power parameters, the discharge characteristics of three modes were comparatively analyzed: SDBD (**Figure 1d**), VDBD (**Figure 1e**), and S-VDBD (**Figure 1f**). The corresponding optical discharge images are presented in **Figures 1g-i**. It is observable that the S-VDBD mode exhibits higher discharge brightness and a more uniform spatial distribution, demonstrating a pronounced discharge enhancement effect compared with the individual SDBD or VDBD modes. The corresponding voltage–current waveforms (**Figures 2a and 2b**) further reveal that under identical applied voltages, the discharge current amplitude in the S-VDBD mode exceeds those of the other two modes. Moreover, specific waveform features (e.g., the emergence of additional discharge pulses) cannot be explained by a simple linear superposition of SDBD and VDBD, indicating a nonlinear coupling interaction between the surface and volume discharges. This coupling behavior can be further understood from the distinct electric-field distributions and discharge-channel characteristics of SDBD and VDBD. In SDBD, the tangential electric field along the quartz dielectric surface promotes discharge propagation near the tube wall, whereas the radial electric field in VDBD drives discharge channels across the gas gap and extends the plasma region toward the reactor interior. When the two discharge modes are simultaneously activated in reactor, the associated surface and space charges further redistribute the local electric field, facilitating additional breakdown or re-breakdown events. This field–charge interaction is consistent with the higher current amplitude and more uniform discharge images observed in the coupled mode.

To further comprehend the discharge characteristics of the electric-field-coupled rotary S-VDBD reactor from an electrical perspective, an equivalent circuit model was introduced for a simplified analysis. The equivalent circuit of a conventional DBD reactor typically consists of a dielectric capacitance C_d in series with a parallel combination of a gas gap capacitance C_g and an equivalent discharge resistance R_p [26–28]. As illustrated in **Figures 2d and 2e**, which depict the equivalent circuits for SDBD and VDBD respectively, K_1 and K_2 represent the gas gap breakdown switches that conduct when the local electric field exceeds the breakdown threshold. Specifically, the SDBD branch can be equivalent to a second-order RC network consisting of a dielectric capacitance C_{d1} in series with a gas gap capacitance C_{g1} , which is further paralleled with an equivalent resistance R_1 . Similarly, the VDBD branch comprises a second-order RC network containing a dielectric capacitance C_{d2} and a gas gap capacitance C_{g2} in parallel with a variable resistance R_2 . During composite discharge, both branches are connected in parallel to the same power supply, forming a parallel system with two second-order branches, as shown in **Figure 2f**.

In a single branch, the equivalent series capacitance is given by (Eq. (1)):

$$C_{eq,1} = \frac{C_{d1}C_{g1}}{C_{d1} + C_{g1}}, C_{eq,2} = \frac{C_{d2}C_{g2}}{C_{d2} + C_{g2}} \quad \text{\textbackslash * MERGEFORMAT (1)}$$

During composite discharge, $C_{coupling}$ represents the additional equivalent capacitance arising from the electric-field coupling and space-charge interactions between the two discharge branches. When high-order nonlinear effects are neglected, the total equivalent capacitance of the composite discharge mode can be approximately expressed as the sum of the equivalent capacitances of each branch and the coupling capacitance (Eq. (2)):

$$C_{eq,hybrid} \approx C_{eq,1} + C_{eq,2} + C_{coupling} \quad \text{\textbackslash * MERGEFORMAT (2)}$$

Assuming that the equivalent capacitance remains constant within a single discharge cycle, the maximum capacitive energy storage under the applied peak voltage U_{max} can be approximately estimated as (Eq. (3)):

$$W_k \approx \frac{1}{2} C_{eq,k} U_{max}^2 \quad (k = 1, 2, \text{ hybrid}) \quad \text{\textbackslash * MERGEFORMAT (3)}$$

The average discharge power can be further expressed as (Eq. (4)):

$$P_k \approx W_k f \quad \text{\textbackslash * MERGEFORMAT (4)}$$

where f is the operating frequency. Because the composite discharge mode possesses a larger total equivalent capacitance, it can store more electrical energy and generate a higher average discharge power, thereby enhancing the local electric field intensity and the reactive species generation capacity. Meanwhile, the mechanical rotation of the reactor continuously tumbles and redistributes the powder within the discharge zone, allowing different particles and particle surfaces to be repeatedly exposed to the plasma. Combined with the broader active region provided by the coupled SDBD and VDBD, this continuous redistribution enhances the contact between the plasma and powder particles, mitigates agglomeration, reduces treatment “dead zones”, and ultimately promotes more consistent and efficient surface modification.

Building upon this electrode configuration and discharge mode, the operating parameters of the nanosecond pulsed power supply were systematically optimized. By comparing the optical discharge images (**Figure S3**) and optical emission spectra (**Figure S4**) under varying voltages (4–10 kV) and frequencies (3–6 kHz), it is observed that discharge onset occurs at an applied voltage of 5 kV, albeit with a relatively weak discharge intensity. As the voltage rises to 6 kV, the discharge region gradually expands and becomes increasingly continuous. At 7 kV, both the discharge intensity and spatial uniformity reach an optimal state. A further increase in voltage yields only a marginal enhancement in the discharge performance. The optical emission spectra exhibit a consistent trend; the intensities of the characteristic emission peaks of argon atoms progressively increase with the elevation of both the voltage amplitude and power frequency, demonstrating a positive correlation between the excited species intensity and the voltage amplitude. Consequently, by comprehensively considering the optical discharge images and spectral results, a voltage of 7 kV and a frequency of 4 kHz were selected as the optimal discharge parameters for subsequent powder modification. The corresponding voltage–current waveforms

are depicted in **Figure S5**.

Optical emission spectra of the DBD process were acquired utilizing a spectrometer over a wavelength range of 200–1100 nm. The spectral measurements were conducted under two distinct conditions: with only argon gas introduced and following the introduction of mBN powder, as depicted in **Figure 2c**. Compared with the background spectrum of pure argon, the spectral composition remained largely unchanged upon the introduction of mBN powder, predominantly consisting of a series of strong emission lines from argon atoms. Notably, within the characteristic emission regions of boron (~249.7 nm) and nitrogen (740–750 nm), neither the emergence of new emission peaks nor a significant enhancement of existing spectral lines was observed. This phenomenon can likely be attributed to the exceptionally high bond energy and thermal stability

inherent to boron nitride, which makes it challenging to be effectively sputtered or decomposed into gaseous atoms for excitation within the low-temperature plasma. Even if a minute fraction of B or N atoms were dissociated, their emission signal intensities would be far inferior to the background argon lines, thereby being masked. Consequently, the introduction of mBN powder did not induce any observable variations in the gas-phase composition of the plasma. This indicates that the modification mechanism of the plasma on the mBN powder is likely not governed by drastic gas-phase byproduct generation or etching, but rather by a milder approach realized through the physical bombardment by reactive species or the grafting of functional groups onto the powder surface.

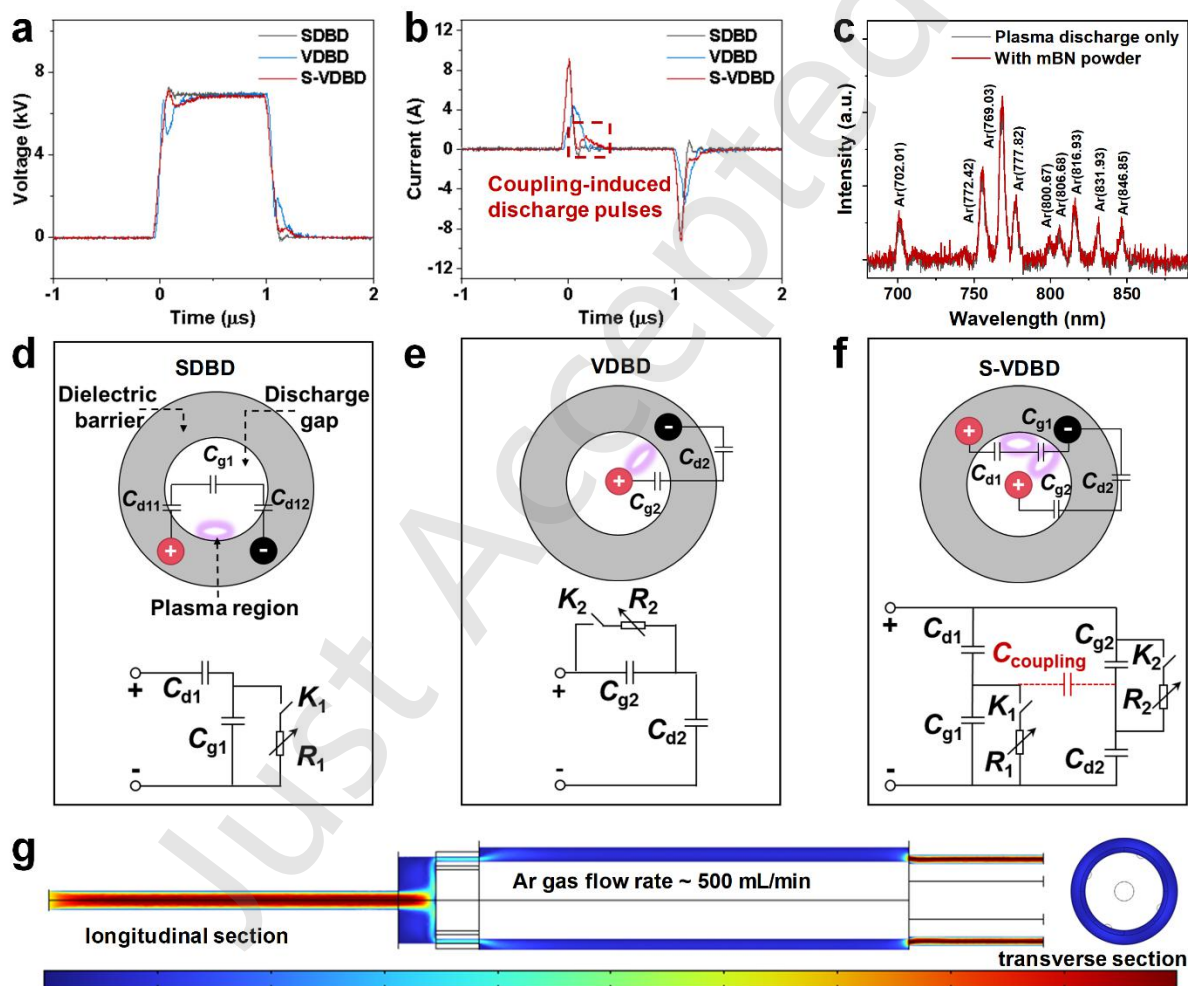


Figure 2. (a) Applied voltage and (b) discharge current waveforms of the SDBD, VDBD, and coupled S-VDBD modes. (c) Optical emission spectrum of the S-VDBD mode under discharge parameters of 7 kV and 4 kHz. (d–f) Equivalent circuit models of the SDBD, VDBD, and coupled S-VDBD. (g) COMSOL flow field simulation images of different cross-sections at 500 mL/min.

Furthermore, a three-dimensional flow field model of the reactor was established using COMSOL Multiphysics to numerically simulate the gas flow behavior within the discharge space (**Figure S6**). The model geometry, including the positions of the gas inlet and outlet, was constructed according to the actual reactor configuration. The effective discharge length of the reactor was 90 mm, and the discharge gap between the high-voltage electrode and the dielectric barrier was 3 mm. Flow field distributions under

various operating conditions were simulated by configuring low (100–300 mL/min), medium (400–600 mL/min), and high (700–900 mL/min) flow rates, respectively (**Figure S7**). The results reveal that within the medium flow rate range, the gas distribution in the discharge region is uniform and the flow remains stable, which is conducive to the sustained excitation of the plasma. Comprehensively considering the simulation results and experimental stability, an argon flow rate of 500 mL/min was ultimately selected, as illustrated in

Figure 2g. Under this condition, the stable excitation of the DBD plasma is guaranteed while simultaneously circumventing discharge instability caused by excessively low flow rates, as well as turbulence effects induced by excessively high flow rates. For the powder surface modification medium, a relative humidity of approximately 20% was employed, corresponding to a carrier gas flow rate of 125 mL/min.

Based on the above discharge comparison and parameter optimization, the S-VDBD mode was selected for the subsequent surface modification of mBN. The following material characterization and composite performance results further demonstrate the effectiveness of this selected discharge mode for mBN surface functionalization and interfacial regulation.

3.2 Analysis of the physicochemical properties of the powder products

Figures 3a and **3b** present the SEM images of h-BN before and after the ball-milling treatment, respectively. As observed in **Figure 3a**, the untreated h-BN raw material exhibits a smooth surface and a bulky stacked structure characterized by large lateral dimensions and substantial thickness. Following the high-energy ball-milling process (**Figure 3b**), the lateral dimensions are reduced, and the thickness is decreased compared to the pristine h-BN. This indicates that the intense shear and impact forces generated during ball milling effectively exfoliated the bulk h-BN into thinner mBN. **Figure 3c** displays the morphology of the plasma-treated mBN alongside its corresponding EDS elemental mapping. It is evident that the plasma-treated sample retains its typical lamellar structure without undergoing any noticeable structural collapse. Further elemental analysis reveals that, in addition to the intrinsic B and N elements, a uniformly distributed and distinctly outlined O element signal emerges on the mBN surface. This result compellingly demonstrates that the plasma treatment successfully introduced oxygen-containing functional groups onto the mBN surface, thereby achieving effective surface functionalization.

To further investigate the effects of exfoliation and plasma modification on the structure of mBN, XRD analysis was performed on the samples, as shown in **Figures 3d** and **3e**. The pristine h-BN raw material exhibits characteristic (002) and (004) diffraction peaks at $2\theta = 26.70^\circ$ and 55.09° , respectively. Following the mechanical exfoliation and plasma functionalization treatments, mBN and mBN-OH still retain distinct (002) and (004) diffraction peaks, indicating that the treatment processes did not disrupt the crystalline structural integrity of the h-BN. Compared with the pristine h-BN, the relative intensity of the (002) peak for mBN is enhanced. This is primarily attributed to the exfoliated material being more prone to undergoing a preferred orientation along the (002) crystal plane on the testing substrate, which substantially increases the scattering probability of this plane. In contrast, the intensity of the (002) peak for mBN-OH slightly decreases, suggesting that the plasma surface modification weakens the long-range interlayer order to a certain extent, further corroborating the successful functional modification of the mBN surface. As illustrated in **Figure 3e**, the (002) diffraction peak of mBN

exhibits a slight shift toward higher angles, with its corresponding interplanar spacing decreasing from 0.3336 nm ($2\theta = 26.70^\circ$) for h-BN to 0.3329 nm ($2\theta = 26.75^\circ$). This subtle contraction of the interlayer spacing is predominantly caused by the residual compressive stress introduced into the crystal lattice by the intense shear forces during the mechanical ball-milling exfoliation process. Furthermore, combined with the broadening of the full width at half maximum (FWHM) of the (002) diffraction peak after exfoliation, it demonstrates that the exfoliation process successfully reduced the crystallite size and effectively decreased the number of h-BN layers[29-32].

To investigate the effects of boron nitride exfoliation and plasma treatment on the surface chemical states of mBN, FTIR spectroscopy was initially employed for characterization, with the results presented in **Figure 3f**. Both the pristine h-BN and the mBN before and after plasma treatment exhibit intrinsic B-N characteristic absorption peaks at approximately 1370 and 800 cm^{-1} , which correspond to the in-plane stretching vibration of the B-N bonds and the out-of-plane bending vibration of the B-N-B bonds, respectively[33]. This indicates that both the high-energy ball milling and the plasma treatment perfectly preserved the intrinsic hexagonal lattice skeleton of the h-BN, without disrupting its fundamental thermally conductive structure. In the high-wavenumber region of 3200–3500 cm^{-1} , the pristine h-BN displays only a negligible signal corresponding to adsorbed water. In contrast, both the ball-milled mBN and the plasma-modified mBN-OH exhibit a distinct broad absorption peak in this region, which corresponds to the stretching vibration of hydroxyl (–OH) groups. This phenomenon indicates that following mechanical exfoliation or plasma treatment, the specific surface area of the material increased, and more active sites or oxygen-containing functional groups were introduced onto the surface. The hydroxyl signal of mBN primarily originates from the increase in physically adsorbed water due to the enlarged specific surface area during ball milling, as well as possible spontaneous hydrolysis at edge defects. Conversely, the signal of mBN-OH encompasses the combined contributions of physically adsorbed water and chemically bonded hydroxyl groups grafted by the plasma. Because FTIR spectroscopy is highly sensitive to surface physically adsorbed water, its broad peak signal can easily mask the subtle variations induced by chemical grafting, making it difficult to distinguish the origin of the hydroxyl groups based solely on peak intensity. To further elucidate the chemical bonding states of the surface functional groups post-modification and to quantify the grafting efficacy, XPS—a technique more sensitive to the surface chemical environment—was subsequently employed for an in-depth analysis.

Figures 3i and **3l** present the XPS survey spectra of the boron nitride before and after plasma treatment. Following the surface modification of the mBN powder, the O 1s content increases from 1.32% to 1.77%. A detailed XPS analysis of the B 1s electron binding energy (**Figures 3j**, **3m**, and **S8**) reveals the presence of an increased number of B–O bonds, which are manifested as a slight shift of the main peak in the B 1s spectrum toward a higher binding energy.

These results demonstrate that the plasma treatment effectively introduced hydroxyl groups onto the surface of the mBN powder, achieving successful surface modification that will facilitate interfacial bonding in the subsequent composites[29, 34].

Figure 3h displays the EDS spectra of the powders before and after plasma modification. It is observable that the atomic proportion of the O element increases following the modification, indicating that the surface of the mBN powder has been successfully grafted with hydroxyl groups. Based on the TGA (**Figure 3g**), both mBN and mBN-OH exhibit good thermal stability at elevated temperatures, with their mass varying gradually as the temperature rises. At an elevated temperature of 800 °C, the total weight loss of mBN is a mere 0.325%. This is primarily attributed to the desorption of surface-adsorbed water and the decomposition of certain unstable oxygen-containing functional groups. In contrast, the total weight loss of mBN-OH reaches 0.828%, slightly higher than that of the untreated mBN. This additional weight loss is predominantly ascribed to the condensation dehydration reaction of the hydroxyl functional groups grafted onto the mBN surface, indicating the successful introduction of hydroxyl groups

and further verifying the realization of surface functionalization[35, 36]. Meanwhile, the functionalized layer generated by this method is exceptionally thin. Only a minimal degree of surface modification is required to improve the interfacial compatibility between the filler and the matrix. Consequently, while strengthening the interfacial bonding, it minimizes the thermal resistance introduced by the interfacial layer against heat transfer, thereby providing favorable conditions for the synergistic optimization of the thermal conductivity and electrical insulation properties of the composites.

3.3 Thermal conductivity, electrical insulation properties, and applications of the composites

Figure 4a schematically illustrates the preparation process and interfacial mechanism of the mBN-OH/EP composites. First, h-BN is used as the raw material, and mechanical ball milling is employed to generate intense shear forces that overcome the interlayer van der Waals forces, thereby exfoliating it into mBN with a high aspect ratio. Subsequently, the exfoliated mBN is placed into the plasma reactor, where Ar is introduced as the working gas and the reaction medium (H₂O) is introduced via bubbling. Within the plasma environment, high-energy electrons

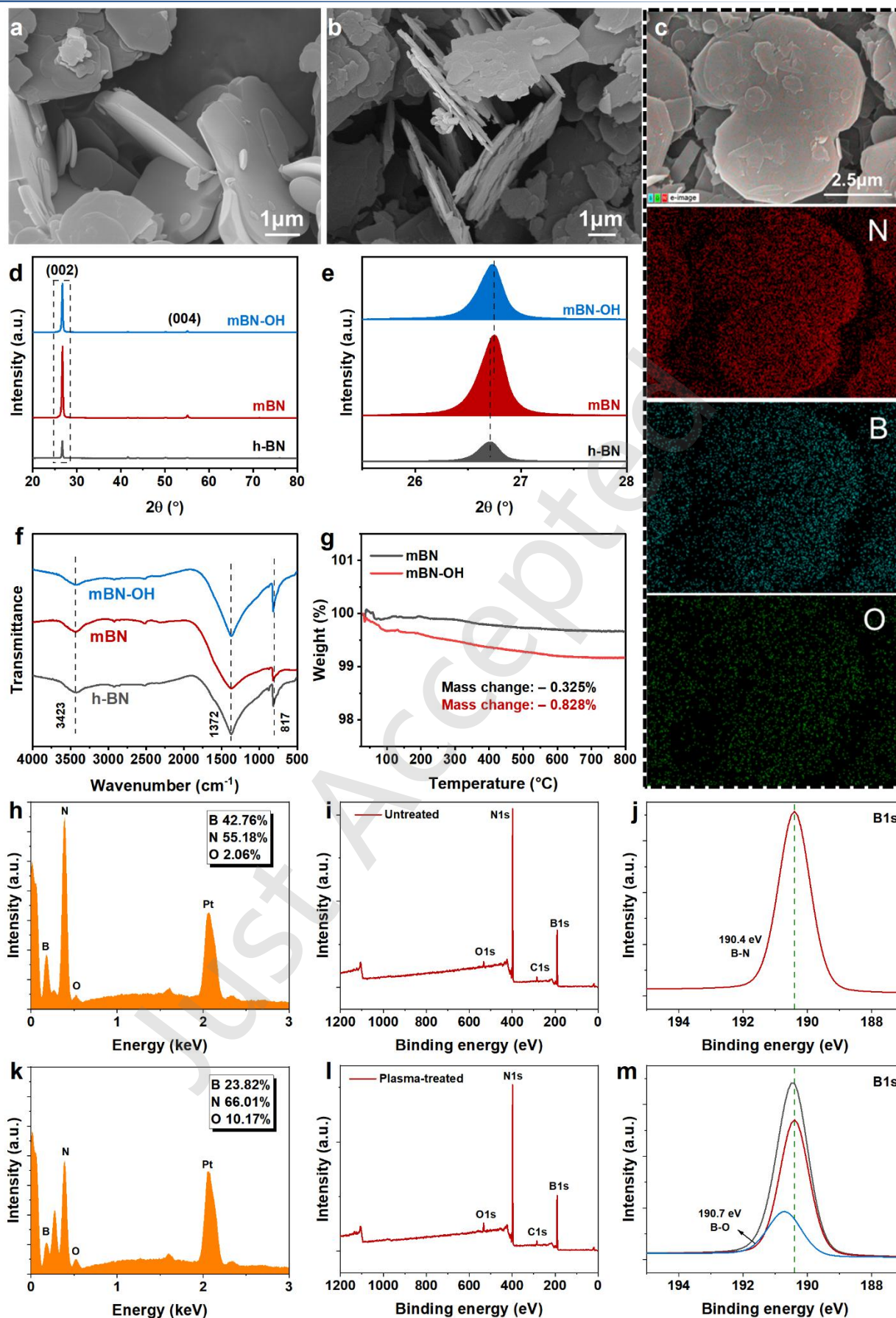


Figure 3. SEM images of (a) the raw h-BN and (b) mBN. (c) SEM image of mBN-OH and the corresponding EDS elemental mappings of B, N, and O. (d) XRD patterns of different samples, (e) magnified view of the dashed-box region, and (f) FTIR spectra. (g) TGA curves of mBN before and after plasma treatment. (h) EDS spectrum, (i) XPS survey spectrum, and (j) high-resolution B 1s spectrum of mBN. (k) EDS spectrum, (l) XPS survey spectrum, and (m) high-resolution B 1s spectrum of mBN-OH.

bombard the water molecules, dissociating them into highly reactive hydrogen radicals ($\text{H}\cdot$) and hydroxyl radicals ($\cdot\text{OH}$). These reactive radicals attack the surface and edge defect sites of the mBN, introducing hydroxyl ($-\text{OH}$) functional groups onto the mBN surface through chemical grafting to

yield surface-hydroxylated boron nitride (mBN-OH). Finally, the mBN-OH is blended with EP. The reactivity and affinity between the hydroxyl and epoxy groups promote uniform dispersion within the matrix, leading to the formation of mBN-OH/EP composites after curing.

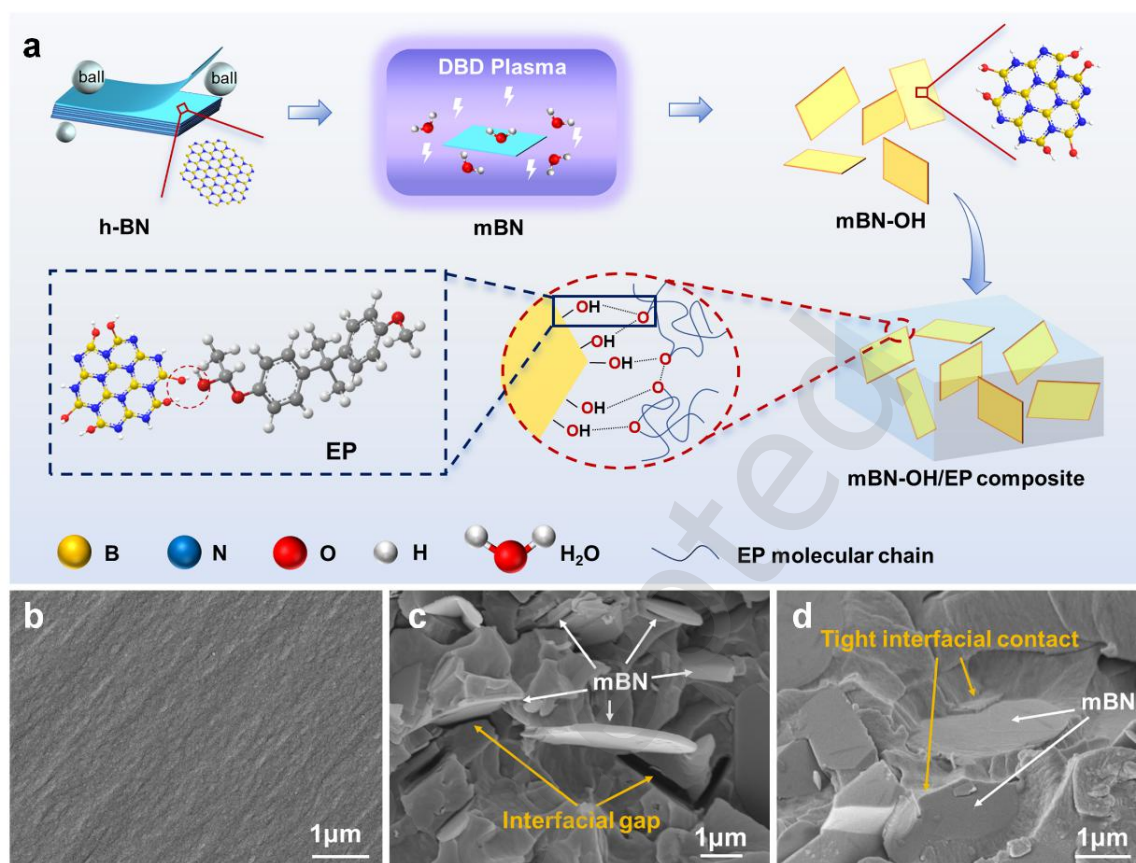


Figure 4. (a) Schematic diagram of the preparation process and interfacial interactions of the mBN-OH/EP composites. SEM images of (b) pure EP, (c) the mBN/EP composite, and (d) the mBN-OH/EP composite.

Figures 4b–4d present the cross-sectional SEM images of pure EP, mBN/EP, and mBN-OH/EP composites, respectively. As observed in **Figure 4b**, the fractured surface of pure EP is relatively smooth and continuous, exhibiting typical brittle fracture characteristics without prominent defective structures. In the unmodified mBN/EP composite (**Figure 4c**), obvious interfacial voids and localized debonding are present between the filler and the epoxy matrix, indicating insufficient compatibility and weak interfacial bonding between mBN and the epoxy matrix. In sharp contrast, the cross-sectional morphology of the mBN-OH/EP composite (**Figure 4d**) is improved. The filler is tightly encapsulated by the matrix, and the interfacial voids are drastically reduced. This improvement is primarily driven by the plasma-induced hydroxylation modification, which grafts hydroxyl groups onto the mBN surface, optimizes its surface properties, minimizes the interfacial polarity mismatch between the filler and the matrix, and effectively reinforces their interfacial compatibility.

To systematically evaluate the effects of filler content and plasma treatment on the thermal conductivity and electrical insulation properties of the composites, the thermal conductivity and breakdown characteristics of the composites prepared under different conditions were tested and comparatively analyzed. **Figure 5a** presents the thermal

conductivities of the composites at different filler loadings. The thermal conductivity of pure EP is $0.23 \text{ W}/(\text{m}\cdot\text{K})$, which is used as the baseline to evaluate the effect of mBN incorporation on the thermal conductivity of the composite. With the increase in mBN filler loading, the thermal conductivity of the composites improves, reaching a peak value of $0.50 \text{ W}/(\text{m}\cdot\text{K})$ at 15 wt%, representing an enhancement of approximately 117.4% compared to pure EP. This indicates that at this filler level, mBN can form more effective thermally conductive pathways within the matrix, thereby enhancing the heat transfer efficiency. When the filler loading is further increased to 20 wt% and 25 wt%, the thermal conductivity conversely decreases. This is primarily due to the fact that excessive filler content is prone to inducing the agglomeration of the flakes, which degrades the dispersion uniformity of the filler within the matrix. Simultaneously, the introduction of more filler/filler and filler/matrix interfaces exacerbates phonon scattering, thereby impeding heat flow. **Figure 5b** illustrates the Weibull probability distribution of the breakdown strength for composites with varying filler loadings, while **Figure 5c** intuitively presents the characteristic breakdown strength calculated from the Weibull distribution. The characteristic breakdown strength of pure EP is $120.83 \text{ kV}/\text{mm}$ and is used as the baseline to evaluate the effect of mBN incorporation

on the electrical insulation performance of the composite. Upon the addition of unmodified mBN, the overall breakdown strength of the composites exhibits a declining trend. This is because the untreated mBN has poor compatibility with the matrix, easily introducing structural defects such as micro-voids at the interface. These defective regions can cause localized electric field distortion and concentration, providing initiation sites for charge accumulation and partial discharge, thereby deteriorating the breakdown strength of the composites. Comprehensively considering both the thermal conductivity and breakdown performance, a fixed filler loading of 15 wt% was selected for the subsequent plasma hydroxylation modification in this study.

At the 15 wt% filler loading, the effect of the Ar/H₂O plasma DBD hydroxylation treatment time on the thermal conductivity and breakdown strength of the mBN-OH/EP composites was further investigated. In this stage, untreated mBN/EP was used as the direct reference to evaluate the additional effect of plasma surface modification. The thermal conductivity results are shown in **Figure 5d**. The thermal conductivities of all treated composites remain higher than that of pure EP, demonstrating that the mBN flakes can still play a dominant role in heat conduction within the matrix. At a treatment time of 1 minute, the thermal conductivity is 0.472 W/(m·K), representing a 105.2% enhancement compared to the pure EP. As the treatment time is prolonged, the thermal conductivity gradually declines. This can likely be attributed to the fact that the hydroxylation treatment forms a hydroxyl-containing interfacial layer on the mBN surface, and the thermal conductivity of this interfacial layer is substantially lower than that of the BN flakes themselves. When heat transfers among the mBN flakes, the interfacial layer, and the epoxy resin, excessive polar functional groups and the interfacial layer will increase the interfacial thermal resistance, thereby impairing the efficiency of cross-interfacial phonon transmission.

The breakdown performance results are shown in **Figures 5e–5f**. The hydroxylation treatment exerts a positive impact on the breakdown performance. With the prolongation of the treatment time, the characteristic breakdown strength of the composites exhibits a trend of initially increasing, subsequently decreasing, and eventually stabilizing. At a treatment time of 1 minute, the breakdown strength of the composite reaches 120.02 kV/mm, essentially recovering to the 120.83 kV/mm of the pure EP, which is higher than that of the unmodified mBN/EP composite. This indicates that moderate hydroxylation can effectively improve the interfacial bonding between the mBN and the epoxy matrix, reduce interfacial voids and debonding, and consequently mitigate localized electric field distortion.

High-power-density power electronic devices are required to operate within an appropriate temperature range to ensure high operating efficiency and long-term reliability. As an important thermal conduction medium between power devices and heat sinks, EP encapsulation materials serve as one of the primary pathways for heat transfer within the devices[37]. To evaluate the practical thermal management performance of pure EP, mBN/EP, and

the mBN-OH/EP composites, this study constructed a thermal management performance testing platform based on a simulated power chip. This testing system comprises an alternating current/direct current (AC/DC) power supply module (model K30-15, input 100–240 VAC, output 15 VDC/2 A), a heating control unit (model LM2596), and an infrared thermal imaging acquisition system. All tests were initiated at a controlled room temperature (approximately 25.6 °C), utilizing an infrared camera to record the real-time evolution of the surface temperature of the simulated power resistor over time. As shown in **Figures 5g–5h**, the surface temperature of pure EP exhibits the most rapid rise, reaching the highest final equilibrium temperature (approximately 110 °C). Following the introduction of the highly thermally conductive mBN filler, the surface temperature rise of the mBN/EP composites decelerates, with the final equilibrium temperature substantially dropping to around 103 °C. This indicates that at a filler loading of 15 wt%, the continuous thermally conductive network constructed by the unmodified mBN within the matrix effectively accelerates heat dissipation. After the plasma hydroxylation treatment, the final equilibrium temperature of the mBN-OH/EP composites (approximately 105 °C) slightly increases compared to the unmodified system. This practical heat dissipation testing result is consistent with the aforementioned thermal conductivity measurements. The plasma modification introduced abundant hydroxyl groups onto the mBN surface, forming a dense and strongly bonded interfacial layer with the matrix. While eliminating physical voids, this dense interfacial layer simultaneously acts as an additional phonon scattering barrier, thereby increasing the internal interfacial thermal resistance of the material. This subtle increment in interfacial thermal resistance partially constrains the overall heat transfer rate of the thermally conductive network. In macroscopic thermal management applications, this is manifested as a marginal deceleration in the heat dissipation rate from the chip surface and a slight rebound in the equilibrium temperature. Nevertheless, the mBN-OH/EP composites still demonstrate superior heat dissipation capabilities compared to pure EP.

To further investigate the underlying mechanism of the influence of hydroxylation modification on the breakdown behavior and partial discharge (PD) of the composites, PRPD measurements were performed on the mBN/EP and mBN-OH/EP composites, with the results presented in **Figures 5i–5k**. **Figures 5i** and **5j** illustrate the PRPD patterns of the mBN/EP and mBN-OH/EP composites, respectively. Evidently, the PD events of both materials are predominantly distributed in the high-electric-field phase regions corresponding to the positive and negative half-cycles of the applied alternating current (AC) voltage, exhibiting distinct phase-dependent characteristics. This indicates that the PD behavior is primarily modulated by the applied AC electric field. Compared with the mBN/EP composite, the mBN-OH/EP composite displays a more concentrated distribution of discharge points, a reduced overall discharge amplitude, and a significant decrease in high-amplitude discharge events during both half-cycles. This demonstrates that the hydroxylation treatment

suppresses the occurrence of intense PD events to a certain extent, which is primarily attributed to the dense interface eliminating air gaps prone to initiating ionization and introducing charge traps that restrict electron mobility. The further statistical results are depicted in **Figure 5k**. Compared to the mBN/EP composite, the apparent charge of the mBN-OH/EP composite decreases from 16.02 pC to 9.08 pC, representing a reduction of approximately 43%, which intuitively reflects the positive effect of the hydroxylation modification on suppressing PD activity.

In summary, at a filler loading of 15 wt%, the mBN-OH/EP composite prepared using the 1 min plasma treatment process exhibits the optimal comprehensive performance. The material not only achieves a markedly enhanced thermal conductivity but also essentially recovers its breakdown strength to the level of pure EP. This surface modification strategy successfully realizes the synergistic optimization of the thermal conductivity and electrical insulation properties of the composites, demonstrating great application potential in the field of advanced electronic packaging.

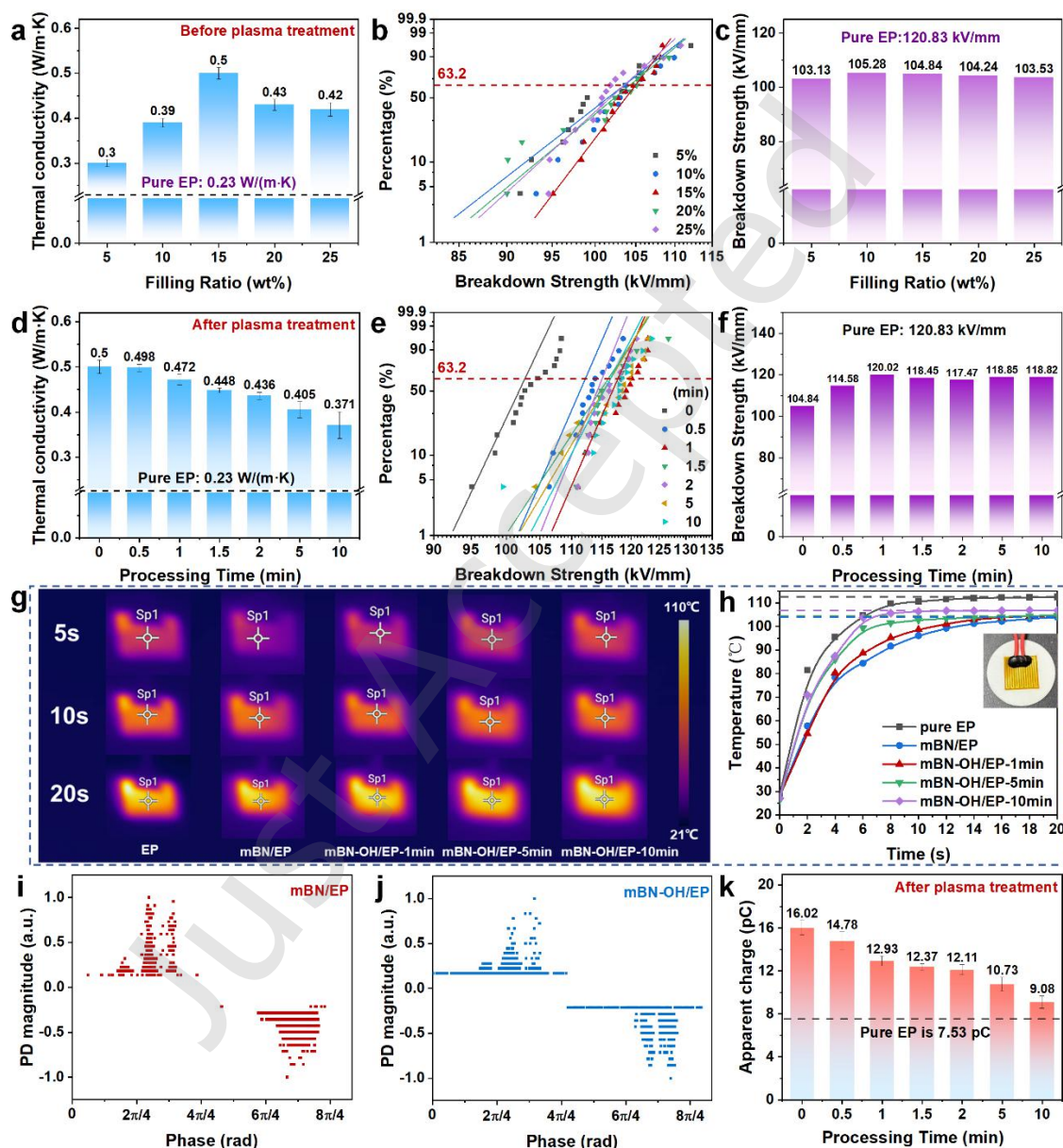


Figure 5. (a) Thermal conductivity, (b) Weibull distribution plots of breakdown strength, and (c) breakdown strength of the mBN/EP composites at different filler loadings. (d) Thermal conductivity, (e) Weibull distribution plots of breakdown strength, and (f) breakdown strength of the mBN-OH/EP composites at different treatment times. (g) Infrared thermal images and (h) temperature variation curves of pure EP and the composites treated by plasma for different times, upon heating for 5 s, 10 s, and 20 s. PRPD patterns of (i) the mBN/EP composite and (j) the mBN-OH/EP composite. (k) Apparent charge of the mBN-OH/EP composites at different treatment times.

To quantitatively compare the comprehensive efficiency of different powder modification strategies in terms of balancing performance enhancement against resource investment, this study introduces a Material Utilization Index (MUI). By taking into account factors such as thermal

conductivity enhancement, electrical insulation improvement, filler loading, and processing energy consumption, this index is employed to characterize the comprehensive performance gain achieved per unit of filler loading and unit of energy consumption. It is defined as

follows (Eq. (5)):

$$MUI = \frac{\eta_T \eta_E}{\Phi W} \quad (5)$$

where η_T and η_E are dimensionless relative performance factors for thermal conductivity and breakdown strength, respectively, and are defined as the ratios of the corresponding properties of the composite to those of the pure polymer matrix reported in the same study. This normalization facilitates relative comparisons among different material systems while partly reducing the influence of differences in baseline properties, specimen preparation, and testing conditions among literature sources. Φ denotes the filler loading, which is uniformly expressed in wt%, whereas W represents the estimated energy consumption associated with the powder modification process (kWh). Filler loadings reported on a volume basis are converted to wt% using the reported volume fractions and the densities of the filler and matrix. For literature data, the reported processing power or energy consumption is used when available or directly derivable. When the equipment power is not reported, a representative power value is consistently assigned to the same type of processing operation, and the energy consumption of individual steps is summed for multistep modification procedures. These treatments provide a unified basis for calculating the MUI across different studies, enabling the evaluation of the balance among performance improvement, filler utilization, and processing energy input.

Figure 6 compares the reported physical methods[10, 13, 38-44], chemical methods[14, 45-53], and plasma methods[21, 37, 54-61] based on the relationship between modification time and MUI. Since plasma modification represents the focus of this work, the detailed parameters of reported plasma strategies used for MUI evaluation are further summarized in Table S1. Generally, traditional physical and chemical modifications are concentrated in the lower-left region of the plot, with modification times mostly on the order of hours and generally low MUI values, indicating that traditional methods have limited comprehensive efficiency in enhancing material performance. Physical methods are restricted by a singular mechanism of action and insufficient interfacial regulation capability. Although chemical methods can improve compatibility by introducing coupling agents or grafting reactions, they involve multiple reaction steps, prolonged processing cycles, and higher energy consumption, which prevents a significant increase in the overall material utilization index. In contrast, the plasma modification methods are aggregated toward the upper-right region as a whole, demonstrating their capability to achieve higher comprehensive performance enhancements at a lower time cost. Notably, a minority of plasma data points located near the traditional method region belong to plasma-assisted chemical modification, which still relies on subsequent chemical grafting processes, thereby leading to longer processing times and relatively low MUI values. A further comparison of different plasma processes reveals that the data for dynamic plasma treatment are generally located in the upper-right of the static treatment region; that is, a higher MUI is achieved within a shorter treatment time. This

is primarily attributed to the continuous tumbling of the powder during the treatment process, which enables full contact between the filler surface and the active discharge species, thereby improving the surface modification uniformity and overall processing efficiency. Within the dynamic plasma system, the red five-pointed star representing this work is positioned at the optimal location among all data points. This demonstrates that the electric-field-coupled rotary DBD modification strategy can further enhance the surface activation efficiency, yielding a higher MUI while maintaining an extremely short treatment time, thereby achieving an favorable synergy between performance enhancement and resource investment. It is worth emphasizing that previous studies reporting relatively high MUI values employed low-pressure plasma systems, which, despite possessing strong surface activation capabilities, typically rely on vacuum environments and complex auxiliary equipment. In contrast, the atmospheric-pressure electric-field-coupled rotary DBD strategy proposed in this study not only achieves a higher MUI but also avoids the equipment and operational costs associated with vacuum processes, thereby offering a more promising solution for the large-scale, continuous, and green modification of powders.

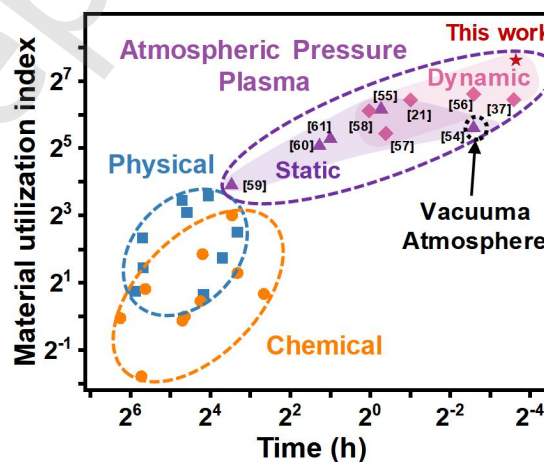
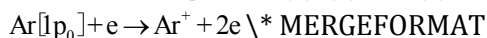


Figure 6. Comparison of the MUI for various treatment methods reported in the literature.

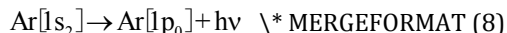
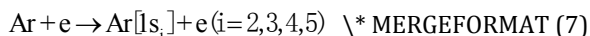
3.4 analysis of the discharge and enhancement mechanisms

The formation of active species in the plasma depends on parameters such as the type, proportion, and flow rate of the working gas and reaction medium, as well as the discharge electron energy and spatial temperature. Generally, in an atmospheric-pressure DBD plasma, the generation of active species originates from frequent inelastic collisions among high-energy electrons, excited species, precursor medium molecules, and their fragments. In this study, free electrons in the discharge region are accelerated by the electric field to become high-energy electrons, triggering a series of reactions including excitation, ionization, and dissociation. These high-energy electrons collide with ground-state argon atoms $Ar[1p_0]$, generating various high-energy species within the rotary DBD plasma. These include metastable atoms (e.g., $Ar[1s_5]$, $Ar[1s_3]$), resonant atoms (e.g., $Ar[1s_4]$, $Ar[1s_2]$), highly excited atoms

(Ar*), argon excimers (e.g., Ar₂[³Σu⁺, v=0]), as well as atomic ions (Ar⁺) and molecular ions (Ar₂⁺). The bombardment of these high-energy species on the surface of the boron nitride powder can break its surface chemical bonds, thereby creating active sites[62]. *MERGEFORMAT Typical reactions are shown in equations (6), (7), and (8).

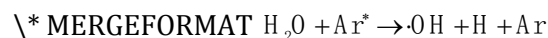
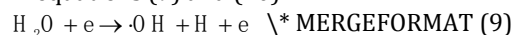


*MERGEFORMAT (6)



Upon introducing H₂O as the reaction medium, the plasma discharge reactions primarily encompass processes such as electron impact dissociation, Penning dissociation, and charge transfer. Due to the relatively high bond energy of the O–H bond in water molecules (approximately 4.8 eV), its dissociation requires sufficient energy input. In the plasma environment (as illustrated in the plasma discharge reaction section of **Figure 7a**), the dissociation of H₂O proceeds mainly through two dominant pathways: First, when the electron energy exceeds 4.8 eV, direct electron impact leads to the dissociative excitation of H₂O, generating highly reactive hydroxyl radicals (•OH) and hydrogen atoms (H); Second, energetic metastable argon atoms (e.g., Ar[1s₅], Ar[1s₃]) undergo Penning dissociation reactions with H₂O

molecules, which are a crucial pathway for the generation of •OH radicals within the system. Typical reactions are shown in equations (9) and (10).



*MERGEFORMAT (10)

It is evident that excited argon atoms (Ar*) and high-energy electrons serve as the primary driving forces for dissociating water molecules and generating the crucial reactive species, •OH radicals. The interactions between plasma and material surfaces typically encompass physical etching, chemical grafting, and thin-film deposition. In this study, the modification of the mBN powder using the H₂O-medium plasma is predominantly a surface chemical grafting process. On the one hand, the physical bombardment of the mBN surface by high-energy species in the plasma cleaves the inert B–N bonds, thereby exposing electron-deficient boron (B) atomic active sites. On the other hand, abundant •OH radicals migrate to the mBN surface and chemically bond with the electron-deficient B atoms via strong Lewis acid-base interactions. This results in the formation of stable B–OH covalent bonds, thereby accomplishing the successful grafting of hydroxyl (–OH) functional groups onto the mBN powder surface.

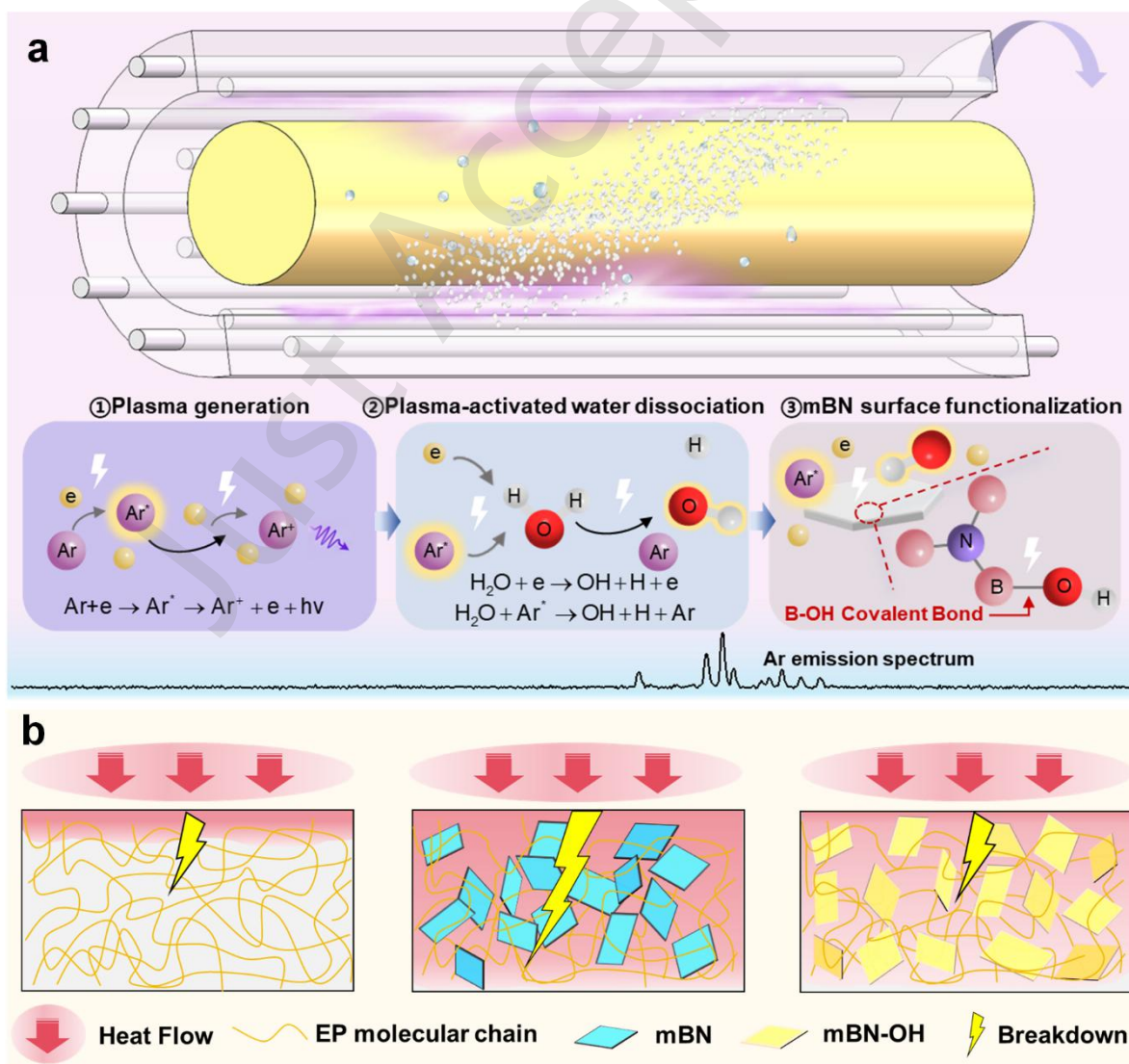


Figure 7. (a) Schematic diagram of the plasma-induced surface hydroxylation mechanism of boron nitride powder. (b) Schematic diagrams of the thermal conduction and breakdown mechanisms in pure EP, the mBN/EP composite, and the mBN-OH/EP composite.

As illustrated in **Figure 7b**, the interfacial state between the filler and the matrix is crucial in determining the thermal conductivity and electrical insulation properties of the composites. Pure EP, as an amorphous polymer, relies on the random vibration of molecular chains for heat conduction. This process is characterized by a short phonon mean free path and severe scattering. Consequently, its intrinsic thermal conductivity is extremely low; as shown on the left side of the figure, heat flow transfers slowly among the disordered EP molecular chains[62]. Upon the introduction of the highly thermally conductive mBN, the thermal conduction behavior of the composites undergoes a fundamental transformation. mBN possesses a perfect crystal structure and an extremely high in-plane intrinsic thermal conductivity, providing high-speed transmission channels for phonons. When the filler content reaches or exceeds the percolation threshold, the mBN flakes interconnect with each other, forming a three-dimensional continuous thermally conductive network within the matrix. At this point, the heat flow preferentially transfers rapidly along the low-thermal-resistance pathways constructed by mBN, bypassing the highly resistive resin matrix, which macroscopically manifests as a dramatic surge in the thermal conductivity of the composites.

However, the unmodified mBN exhibits poor compatibility with the EP matrix, and distinct microscopic voids exist at the interface. These defects easily trigger electric field distortion and concentration effects, thereby drastically reducing the breakdown strength of the material. After the plasma hydroxylation treatment, the -OH groups on the mBN surface enhance the affinity between the filler and the EP matrix, enabling the matrix to tightly encapsulate the filler and effectively eliminating physical defects such as interfacial micro-voids. Such a dense interface not only prevents the concentration of the electric field at defects but also introduces abundant deep traps to suppress the injection and migration of space charges, thereby homogenizing the internal electric field distribution and improving the macroscopic breakdown strength[63]. In terms of heat conduction, although the dense interfacial bonding theoretically reduces the interfacial thermal resistance, the introduction of the interfacial layer introduces additional phonon scattering. This, to some extent, masks the gains brought about by the interfacial improvement, resulting in a slight decrease in the thermal conductivity of the composite compared to the unmodified system. In summary, while largely maintaining the high thermal conductivity of the composites, the plasma surface modification strategy achieves a substantial recovery and leap in electrical insulation performance. This fully demonstrates the synergistic optimization role of interfacial engineering in tailoring the multifunctional properties of composite materials.

4 Conclusions

To address the issues of uneven treatment, low efficiency, and environmental pollution inherent in traditional powder surface modification processes, this

study successfully designed and constructed an electric-field-coupled rotary DBD reactor, achieving highly efficient, uniform, and green surface hydroxylation modification of mBN fillers. This reactor combines electric-field coupling with a dynamic rotary structure. Electric-field coupling is mainly responsible for enhancing the discharge characteristics, whereas dynamic rotation promotes repeated and more uniform exposure of powder particles to the plasma. Through voltage-current waveforms, discharge optical emission images, and optical emission spectroscopy diagnostics, the discharge parameters suitable for mBN modification were determined, and the optimal processing conditions were selected for subsequent research. The effectiveness of this method was verified through a series of physical and chemical characterizations. Analysis results from XPS and FTIR indicated that the plasma treatment successfully grafted hydroxyl functional groups onto the mBN surface and formed stable B-O chemical bonds, while fully retaining its intrinsic h-BN crystal structure, ensuring that the structural foundation for thermal conductivity remained intact. The modified mBN-OH exhibited improved interfacial compatibility with the EP matrix; cross-sectional SEM images of the composites revealed a reduction in interfacial defects and microscopic voids, achieving tighter interfacial bonding. This optimized interfacial structure ultimately led to the synergistic optimization of the thermal conductivity and electrical insulation properties of the composites. Under the optimal processing conditions of a 15 wt% filler loading and a 1 min treatment time, the thermal conductivity of the composite reached 0.472 W/(m·K), representing a 105.2% increase compared to pure EP. Simultaneously, its breakdown strength reached 120.02 kV/mm, an increase of 14.48% over the unmodified mBN-filled composite, achieving an favorable balance between high thermal conductivity and high electrical insulation. Furthermore, PRPD measurements demonstrated that the partial discharge activity in the modified composites was suppressed, with the apparent charge decreasing from 16.02 pC to 9.08 pC, a reduction of approximately 43%. This reflects the positive effect of the dense interface in eliminating air gaps and introducing charge traps to restrict electron mobility, thereby actively suppressing intense partial discharges. To quantitatively evaluate the process efficiency, the MUI was introduced. The results indicate that this method outperforms traditional wet chemical methods and conventional plasma technologies in terms of both energy consumption and raw material utilization. Overall, the electric-field-coupled rotary DBD technology proposed in this study provides a novel pathway for the surface functionalization of powder fillers, holding significant engineering guidance and application value for the development of polymer-based encapsulation materials that feature both high thermal conductivity and robust electrical insulation. Future work will further investigate the applicability of this method to other powder systems, explore strategies for larger scale processing, and evaluate the long-term reliability of the modified fillers and composites under practical service conditions.

Electronic Supplementary Material: Supplementary material (please provide the brief detail of the ESM) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909202>.

Data availability

The data supporting the findings of this study are available within the article and its Supplementary Information. Additional raw data related to the plasma modification process, material characterization, and performance evaluation are available from the corresponding author upon reasonable request.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author contribution statement

Juanjuan Ji: Investigation, Writing - Original Draft. Chixuan Fei: Writing - review and editing. Wenbo Zhou: Data curation. Weiming Yang: Validation. Gejiang Jin: Formal analysis. Pengyu Shang: Visualization. Longren Li: Conceptualization. Jialiang Huang: Methodology, Supervision.

Informed consent

Not applicable.

Ethics statement

Not applicable.

Use of AI statement

None.

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Electronic Supplementary Material

Electric-field-coupled rotary DBD plasma-modified mBN enables epoxy encapsulation with both high thermal conductivity and excellent electrical insulation: high material utilization index

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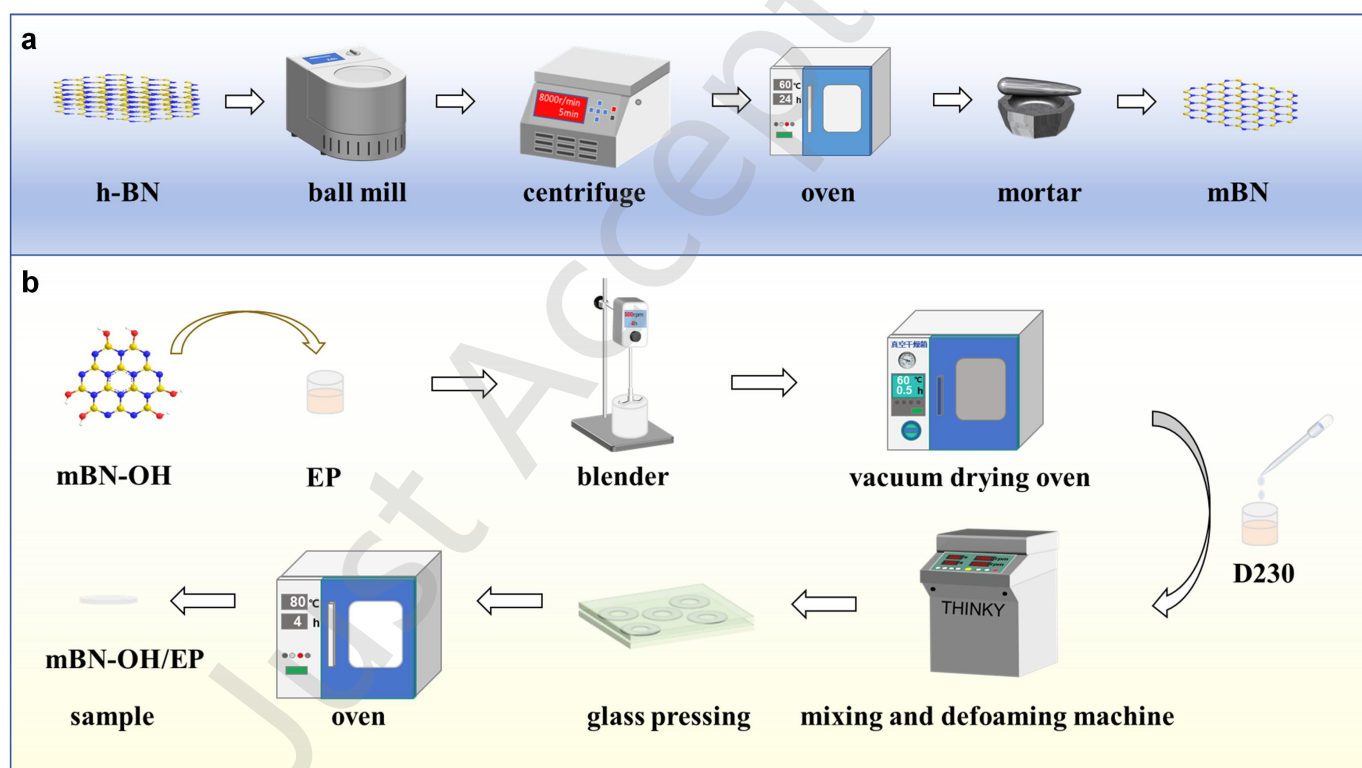


Figure S1. (a)Preparation of mBN, (b)Preparation of composite materials.

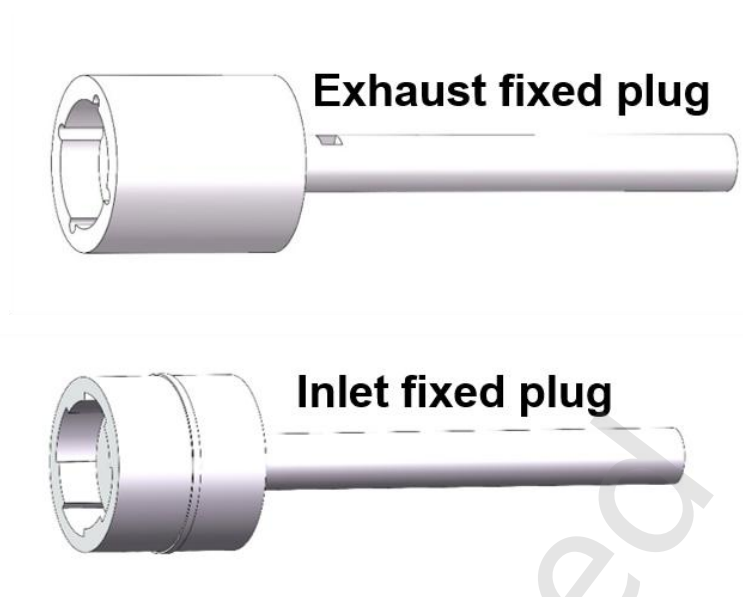


Figure S2. Design drawing of special fixed plug.

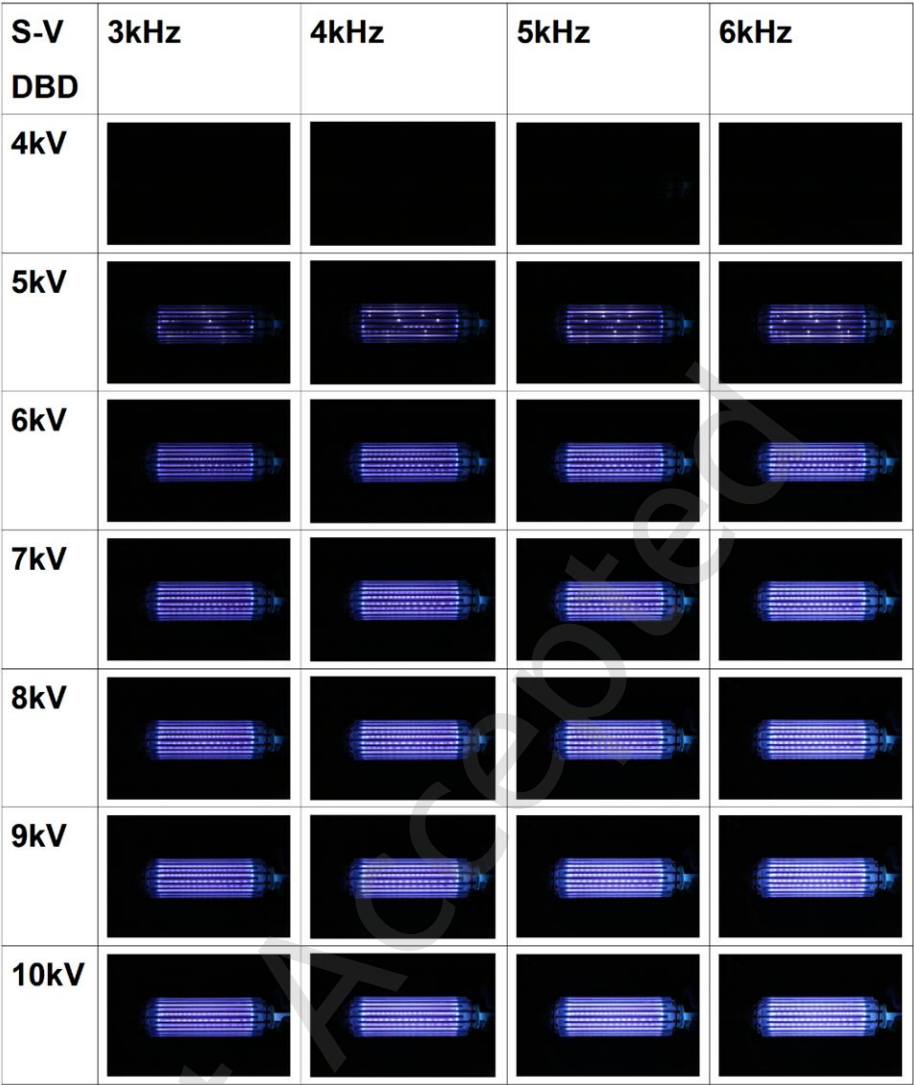


Figure S3. Light emission image in S-VDBD mode.

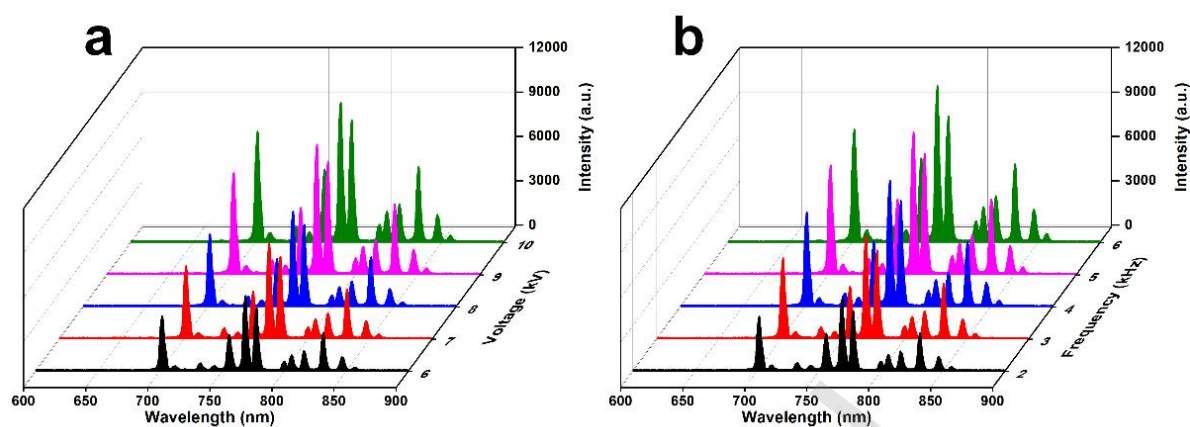


Figure S4. Optical emission spectra in S-VDBD mode under various (a) voltages and (b) frequencies.

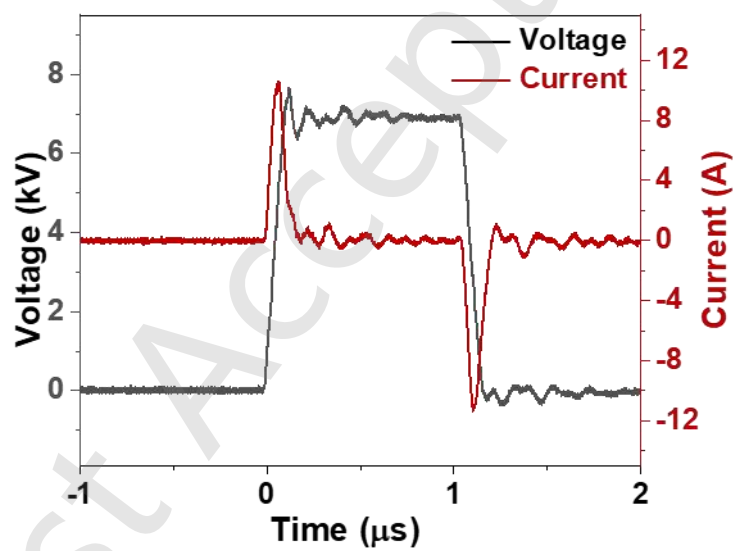


Figure S5. Voltage and current waveforms of S-VDBD.

For the flow-field simulation, the Laminar Flow module was employed. A normal inflow velocity boundary condition was applied at the gas inlet, with the inlet velocity adjusted according to the prescribed volumetric flow rate. A pressure boundary condition of $p_0 = 0$ Pa was imposed at the outlet, with backflow suppression enabled. The remaining solid boundaries were treated as no-slip walls. The inlet and outlet positions were defined according to the actual reactor geometry, as shown in Figure S6.

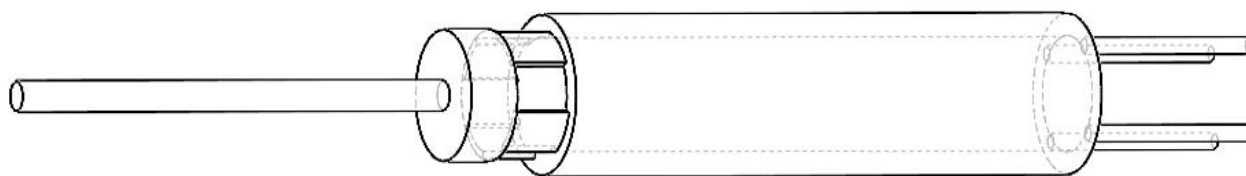


Figure S6. Geometry construction of the COMSOL simulation model.

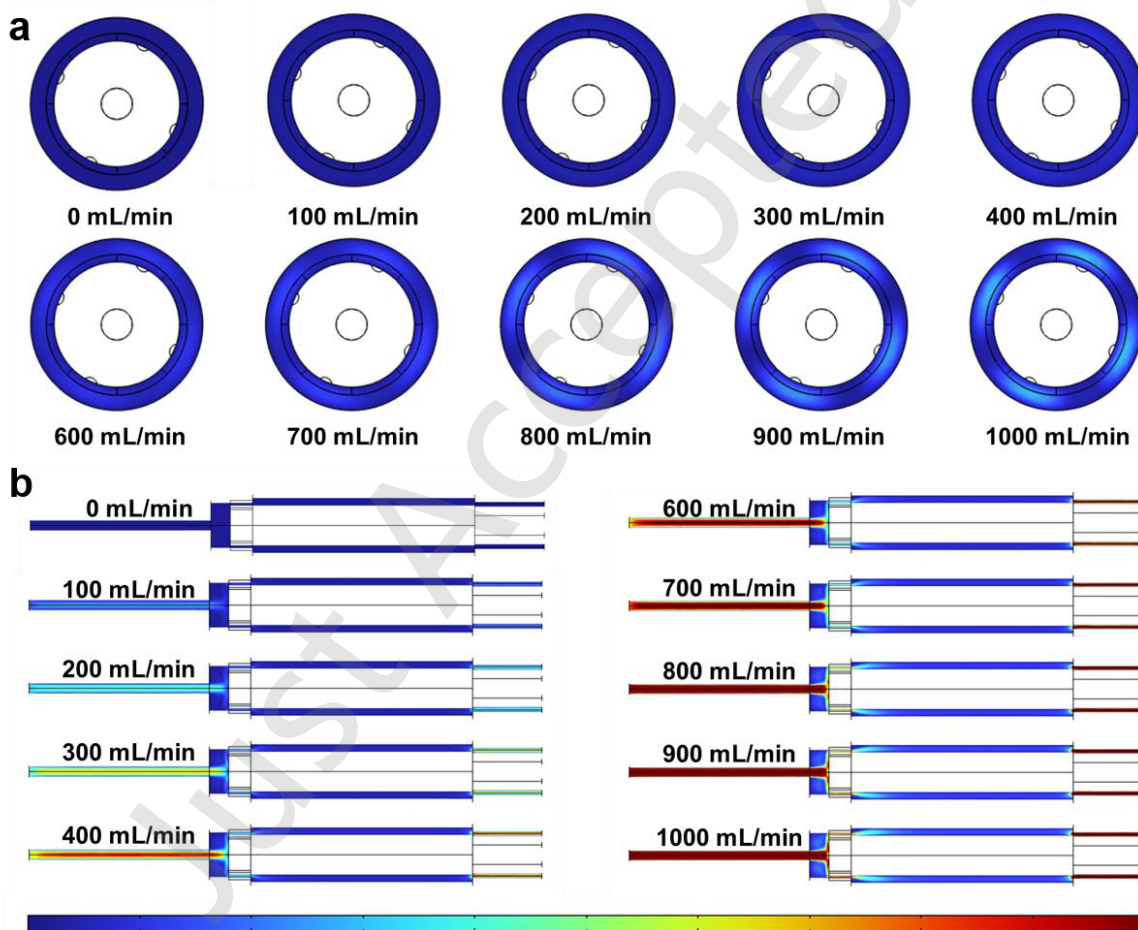


Figure S7. (a) Longitudinal and (b) transverse cross-sections of the COMSOL flow field simulation.

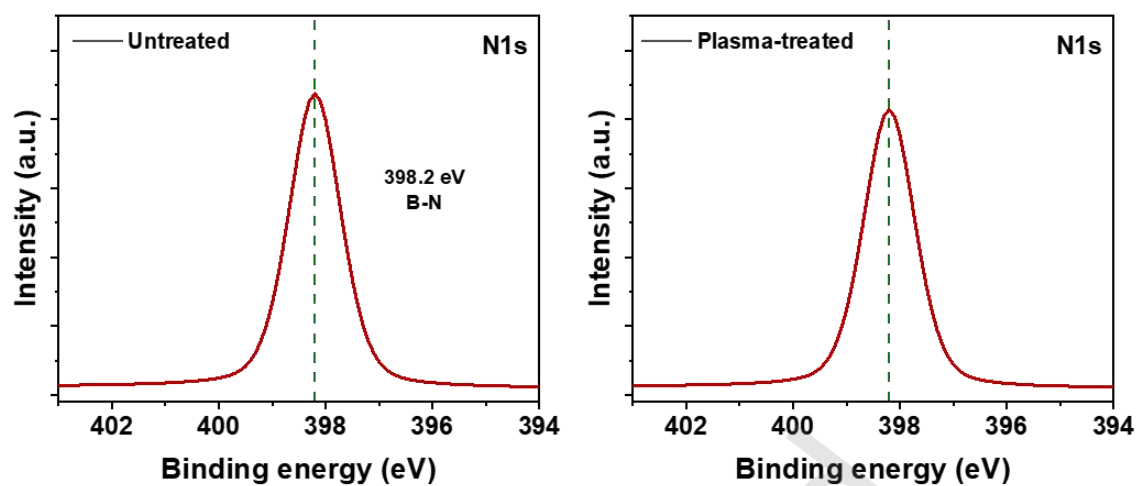


Figure S8. High-resolution N 1s spectrum of plasma-treated boron nitride.

Table S1. Summary of plasma-assisted surface modification strategies reported in the literature and corresponding parameters used for MUI calculation.

References	Filler(wt%)	Thermal conductivity improvement(%)	Breakdown strength improvement(%)	Energy input(kWh)	Treatment time(h)	MUI
54	40	3.07300885	1.265189421	0.2	0.166666667	48.59922859
55	20	4.87345679	1.210351967	0.415	0.83	71.06744592
59	1	1.064075942	1.183673469	8.405	11.1	14.98534755
61	7	4.352941176	1	1.6	2	38.86554622
60	10	2.112676056	2.547138047	1.6	2.416666667	33.63298478
58	20	3.055555556	0.906807335	0.2	1	69.27000474
56	20	3.278606965	1.191257669	0.2	0.166666667	97.64164225
37	56	14	1.166486486	0.333	0.0833	87.20034478
57	14	3.19047619	1.138590203	0.595833333	0.766666667	43.54819101
This work	15	2.04	0.993296367	0.069	0.08	195.7801535