

Preparation of Polypropylene Nanofibers as a Function of the Thermal Exposure Time of the Polymer in a Melt Electrospinning/Melt Blown Hybrid System

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Abstract This study reports the fabrication of polypropylene (PP)-based microfiber webs (<1 μm) using a hybrid melt electrospinning/blown process with the aim of establishing a scalable and solvent-free platform for advanced lithium-ion battery separators. The primary objective was to address the inherent limitations of conventional melt electrospinning particularly the difficulty of achieving fiber thinning due to the high viscosity of polymer melts by incorporating auxiliary hot air flow and reducing the nozzle diameter from 1.0 mm to 0.3 mm. This modified configuration enables enhanced jet elongation and fiber diameter control under processing conditions relevant to industrial applications. The effects of nozzle temperature, hot air temperature, and applied voltage on fiber formation and jet behavior were systematically examined using high-speed charge-coupled device (CCD) imaging techniques. The results demonstrated that increasing both the hot air temperature and applied voltage significantly improved fiber thinning and uniformity, yielding an average fiber diameter of approximately 0.86 μm without evidence of thermal degradation. In contrast, elevated nozzle temperatures, while enhancing melt flowability, resulted in increased discharge rates and hindered fiber refinement when applied alone. These findings identify hot-air temperature as the most robust and controllable parameter for producing submicron fibers while maintaining the polymer integrity. Although the present study primarily focuses on morphological optimization and jet dynamics, future research will investigate the functional performance of fabricated microfiber webs as battery separators. Overall, the proposed hybrid process offers a technically feasible and environmentally sustainable route for the continuous production of fine PP-based fibers tailored for high-performance energy-storage applications.

Keywords Melt-electrospinning; Melt blown; Melt-electrospinning/blown; Polypropylene; Nanofiber

Citation: Moon, E.; Lim, J.; Kim, M.; Kim, H. S. Preparation of polypropylene nanofibers as a function of the thermal exposure time of the polymer in a melt electrospinning/melt blown hybrid system. *Chinese J. Polym. Sci.* 2025, 43, 2422–2431.

INTRODUCTION

In lithium-ion batteries (LIBs) and next-generation energy storage systems, the separator is a critical component that physically isolates the cathode and anode, while allowing the transport of lithium ions. They play a pivotal role in determining the electrochemical performance and safety of batteries.^[1–3] In particular, there is a growing demand for high-performance separators capable of stable operation under high-temperature and high-power conditions, such as those encountered in electric vehicles (EVs) and energy-storage systems (ESSs).^[4–6] To meet these requirements, it is essential to develop next-generation separator technologies that can overcome the limitations of conventional polyolefin-based microporous films, particularly in terms of their thermal stability and mechanical durability.^[7,8]

With the increasing demand for miniaturized and high-capacity lithium-ion batteries, the development of thinner

separators has become increasingly important. These separators must simultaneously satisfy multiple critical criteria, including reduced thickness, uniform pore structure, high mechanical strength, excellent electrolyte wettability, and thermal stability.^[9–13] Achieving these characteristics requires precise control over the fiber diameter within the separator, ideally at the submicron scale (<1 μm). Microfiber-based separators with submicron diameter offer distinct advantages over their conventional counterparts. Their smaller and more uniformly distributed pores shorten the ion transport pathways and improve the electrolyte uptake, thereby enhancing the battery power performance and cycle life. Moreover, these fibers enable the fabrication of separators with high porosity while maintaining a thin and uniform structure, making them particularly suitable for high-energy-density battery applications.^[14–18]

To address these technical demands, melt electrospinning/blown processes have garnered attention as promising fabrication methods.^[19–21] Unlike conventional solution electrospinning, this solvent-free technique is environ-

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Received June 30, 2025; Accepted September 11, 2025; Published online November 19, 2025

mentally friendly and compatible with continuous processing, making it well-suited for large-scale production.^[22–26] However, conventional melt electrospinning, which directly processes polymer melts, has a major limitation: the inherently high viscosity of molten polymers makes it difficult to produce fibers at the submicron scale. Although the use of high voltage can partially alleviate this limitation by promoting jet elongation, the resulting fiber diameter often remains within the submicron to micron range, owing to the limited stretching capacity of high-viscosity melts.

To overcome this challenge, our research group developed an advanced hybrid process that integrates melt electrospinning with a melt-blown system and auxiliary hot-air flow.^[27–29] This combination introduces additional aerodynamic stretching forces, which synergistically enhance jet attenuation and promote fiber thinning under melt-processing conditions. Furthermore, by systematically reducing the nozzle diameter from 1.0 mm to 0.3 mm, the polymer discharge rate significantly decreased, enabling the formation of finer fibers while maintaining continuous throughput. This hybrid configuration presents a scalable, solvent-free platform for submicron fiber fabrication, and offers a viable solution to the industrial limitations of conventional melt-based spinning technologies.

In this study, polypropylene (PP)-based microfiber webs (<1 μm) were fabricated using the proposed hybrid melt electrospinning/blown process, and the effects of various spinning parameters on the fiber diameter and web morphology were systematically investigated. Special attention was given to the interplay between the thermal and aerodynamic parameters, including the nozzle temperature, hot air temperature, and applied voltage, and their impact on the polymer jet behavior, fiber uniformity, and deposition characteristics. Through this approach, we demonstrated a feasible pathway for achieving submicron PP fibers under industrially relevant

melt-processing conditions, offering a promising alternative for advanced battery separator applications.

MATERIALS AND METHODS

Blending

Polypropylene (PP) was based on the PP FR-160 grade from Lotte Chemical (Melt index: approximately 1600 g/10min, ASTM D1238). To adjust the melt flow index (MFI), 2 wt% of a PP-based organic peroxide masterbatch (CM-100, containing 10% organic peroxide) supplied by Plus Chemtech was blended. The modified PP blend was provided by RA Co., Ltd., (Republic of Korea).

Spinning

The blended PP was fed into a metal nozzle with a nozzle tip diameter of 0.3 mm and used for spinning. The tip-to-collector distance (TCD) was fixed at 10 cm. The air pressure for polymer extrusion was maintained at 0.01 MPa, and the hot air pressure was fixed at 0.05 MPa for all spinning conditions. The hot-air-assisted melt-electrospinning/blown process was conducted by varying the nozzle temperature and hot-air temperature to investigate their effects on fiber formation. Fig. 1 illustrates the setup of the melt-spinning/blowing system.

The experimental conditions were categorized into five groups as follows:

C1: nozzle temperature=220 °C, hot air temperature=210 °C

C2: nozzle temperature=220 °C, hot air temperature=220 °C.

C3: nozzle temperature=220 °C, hot air temperature=230 °C.

C4: nozzle temperature=210 °C, hot air temperature=220 °C

C5: nozzle temperature=230 °C, hot air temperature=220 °C.

To further investigate the influence of electrostatic forces on fiber formation, each condition was tested, with and without an applied voltage. The applied voltage was set to 0 kV (no voltage) and 35 kV (with voltage) for each experimental category. The ejection pressure was applied to quantify the polymer throughput as a function of the nozzle temperature.

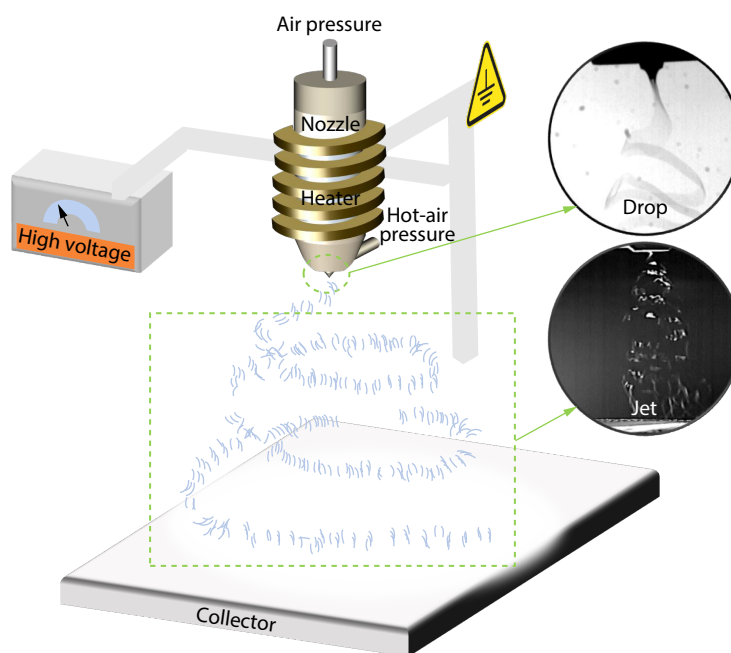


Fig. 1 Melt-electrospinning/blown set up.

The measured mass flow rates were 0.1413 g/m at 210 °C, 0.1732 g/m at 220 °C, and 0.2541 g/m at 230 °C.

These categorized conditions were used to systematically analyze the effects of nozzle and hot air temperatures, as well as the presence or absence of an applied voltage, on the fiber morphology. The fibers collected under these conditions were characterized to evaluate their structural and morphological properties.

ANALYSIS

Real-time Observation and Jet Behavior Analysis

The dynamic behavior of the spinning jet was monitored using a CCD camera (SCC-B2315, Samsung Electronics, Korea), and an image analysis was conducted using a MATLAB-based program. The jet area and distribution uniformity were quantified by binarizing the images and analyzing the distribution of white pixels. For a more detailed evaluation of the jet stability, each image was segmented into three vertical sections, and the standard deviation of the x-coordinates of the jet pixels was calculated over time. A high-speed camera (i-Speed 211, iX Cameras, UK) was used to capture the transient jet motion and flow dynamics.

Morphological Characterization of Electrospun Web

The morphological features of the electrospun nanofiber webs were characterized using field-emission scanning electron microscopy (FE-SEM, SUPRA25, Carl Zeiss, Germany). The fiber diameters were measured to evaluate the influence of the processing parameters on the fiber morphology.

Uniformity Assessment of PE Web Deposition

The deposition patterns of the PP web were digitized using a flatbed scanner (Epson L6190, SEIKO EPSON, Japan), and the uniformity was assessed using MATLAB-based image analysis. The light intensity from the scanned images was correlated with the fiber mass distribution, enabling quantitative evaluation of the deposition uniformity.

Ambient Temperature and Airflow Measurement

The ambient temperature profiles at different tip-to-collector distances (TCDs) were monitored using a temperature sensor (BK50, Beumyoung Technology, Korea). The hot-air velocity was measured at the corresponding TCD positions using a digital anemometer (GM8903, BENETECH, China).

Thermal and Chemical Structure Analysis

The thermal properties of the PP pellets and melt-spun fibers were analyzed using a differential scanning calorimeter (DSC, N-650, Sinco M&T Co., Ltd., Korea) to evaluate the possible changes in the crystalline melting point (T_m) induced by processing. FTIR spectroscopy (IRAffinity-1, Shimadzu, Japan) was employed to examine the chemical structure of the samples and identify any structural modifications resulting from the spinning process.

RESULTS AND DISCUSSION

In this study, the conventional 1.0 mm nozzle previously used in our laboratory was replaced with a 0.3 mm nozzle to reduce the polymer discharge rate under identical air pressure conditions, thereby enhancing the potential for fiber thinning. Although the influence of hot-air temperature on jet formation and fiber

morphology has been extensively examined—including in the prior work of our group^[27–29]—this variable was held constant at 220 °C to specifically evaluate the relative effects of nozzle temperature.

To clarify the role of thermal parameters in jet formation and fiber characteristics during the hybrid melt electrospinning and melt-blowing process, nozzle temperature and hot air temperature were selected as the primary process variables. Their individual effects and potential interactions were systematically analyzed with respect to the resulting fiber morphology. The behavior of the polymer jet varies markedly under different thermal conditions, directly influencing the uniformity and consistency of the produced fibers. In particular, unstable jet motion can result in defects, such as bead formation or fiber breakage, ultimately compromising the structural homogeneity of the deposited web. Therefore, precise thermal control is essential for achieving stable and uniform fiber formation.

In addition, the presence or absence of an applied electric field was identified as a critical factor that significantly affected jet stability, elongation behavior, and fiber diameter. Accordingly, we investigated the effects of nozzle temperature, hot-air temperature, and applied voltage on jet dynamics and web morphology to elucidate the underlying mechanisms governing fiber formation under solvent-free, melt-processing conditions.

Figs. 2(a) and 2(b) illustrate the variation in the jet area under different hot-air temperatures, nozzle temperatures, and applied voltages in the hybrid melt electrospinning/melt blowing system. The experimental results indicated that increasing the hot air temperature from 210 °C to 220 °C had little effect on the jet area; however, a further increase to 230 °C resulted in a noticeable reduction. At elevated hot-air temperatures, the polymer viscosity decreased, rendering the jet more susceptible to turbulence-induced oscillations. This increased instability leads to greater energy consumption during jet elongation and fiber shape retention. Additionally, higher hot-air temperatures lower the jet cooling rate, thereby extending the elongation window. This prolonged elongation promotes the formation of thinner jets, ultimately reducing the overall jet area. In contrast, at lower hot-air temperatures, the polymer maintains a relatively high viscosity, which limits jet elongation. Consequently, more pronounced bending and whipping motions occur, resulting in a broader jet area.

When the hot-air temperature was held constant, increasing the nozzle temperature led to a reduction in polymer viscosity, enhanced melt flowability, and an increased discharge rate. These combined effects contribute to the overall expansion of the jet area. However, when the nozzle temperature was further increased from 220 °C to 230 °C, no significant change in the jet area can be observed. This result suggested that beyond a certain temperature threshold, further reduction in viscosity has a diminishing effect on jet spreading.

A distinct difference in the jet area can be observed, depending on the presence or absence of an applied electric field. When a voltage of 35 kV is applied, the jet area increases significantly. This behavior could be attributed to enhanced electrostatic forces, which not only draw the jet more

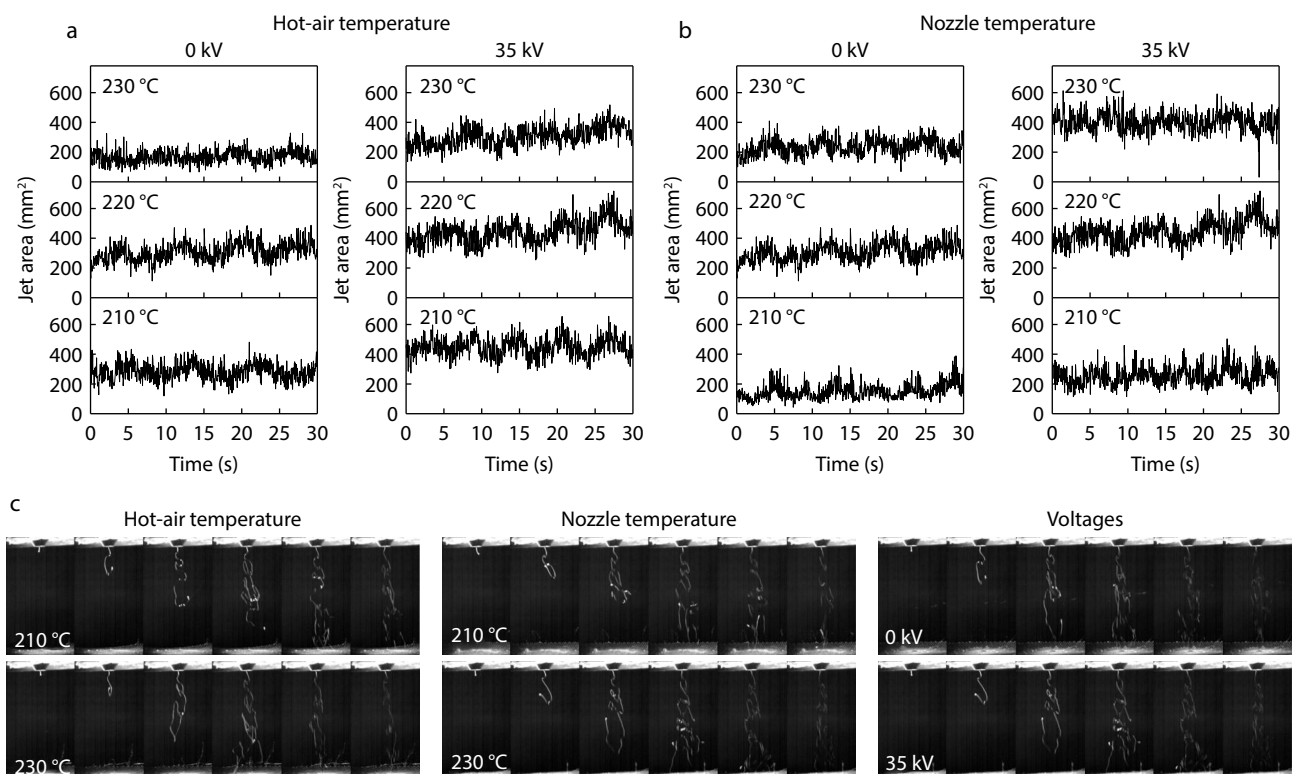


Fig. 2 (a) Effect of hot air temperature variation (210, 220, 230 °C) at a fixed nozzle temperature of 220 °C under two voltage conditions (0 and 35 kV) on the jet area; (b) Effect of nozzle temperature variation (210, 220, 230 °C) at a fixed hot air temperature of 220 °C under two voltage conditions (0 and 35 kV) on the jet area; (c) High-speed camera images under different processing parameter conditions.

strongly toward the collector, but also intensify repulsive interactions among the charged components within the jet. These repulsive forces promoted greater lateral oscillations, resulting in broader jet spreading. In contrast, in the absence of an applied voltage, such electrostatic interactions were eliminated, leading to a more focused and narrower jet area. These observations highlight the critical role of the electric field in modulating the jet dynamics and ensuring spatial stability during the spinning process.

Fig. 2(c) presents high-speed camera images that visually capture the dynamic behavior of the jet under varying process conditions, including the hot air temperature, nozzle temperature and applied voltage. The observed jet motion exhibits trends consistent with the jet area measurements obtained from the CCD camera analysis, providing visual confirmation of the influence of processing parameters on jet behavior. At elevated hot-air temperatures, the jet displays a more pronounced elongation in the downward direction, which is indicative of stronger drawing forces. In addition, motion blur can be observed in the high-speed images, suggesting that the jet velocity exceeded the frame capture rate of the camera. This phenomenon is particularly evident at higher hot-air temperatures. Under high nozzle temperature conditions, a visibly increased polymer discharge volume can be observed, indicating a higher flow rate and greater material throughput from the nozzle.

Fig. 3(a) shows a representative high-speed camera image of the jet. The binarized image was segmented into three regions—top, middle, and bottom—and the x-coordinates of

all the jet-associated pixels within each region were extracted. For each segment, the standard deviation of the x-coordinates was calculated for all the time-sequenced frames. This analysis enabled a quantitative assessment of the temporal stability of the jet.

The analysis revealed that under all tested conditions, the standard deviation of the jet was the highest in the lower region near the collector. This trend indicated that as the polymer jet travels farther from the fixed nozzle toward the collector, its lateral instability increases. The amplified fluctuations in the lower region are likely attributable to the increased exposure to external disturbances, such as ambient air turbulence and gravitational forces, during jet descent. These results suggested a reduction in the controllability of the jet motion at longer travel distances, highlighting the challenges associated with maintaining jet stability throughout the spinning process.

A comparative analysis of the jet standard deviation under varying temperature conditions revealed consistent trends for both the nozzle and hot-air temperatures. At the lower temperature of 210 °C, the polymer exhibits relatively high viscosity, which contributed to a stable and coherent jet trajectory. Consequently, shape fluctuations were minimized, and the standard deviation remained low. This behavior suggested that a high viscosity suppresses both elongation and lateral oscillations, thereby maintaining uniform flow characteristics. At an elevated temperature of 230 °C, the polymer viscosity was significantly reduced, facilitating easier jet elongation. This enabled more uniform stretching along the jet

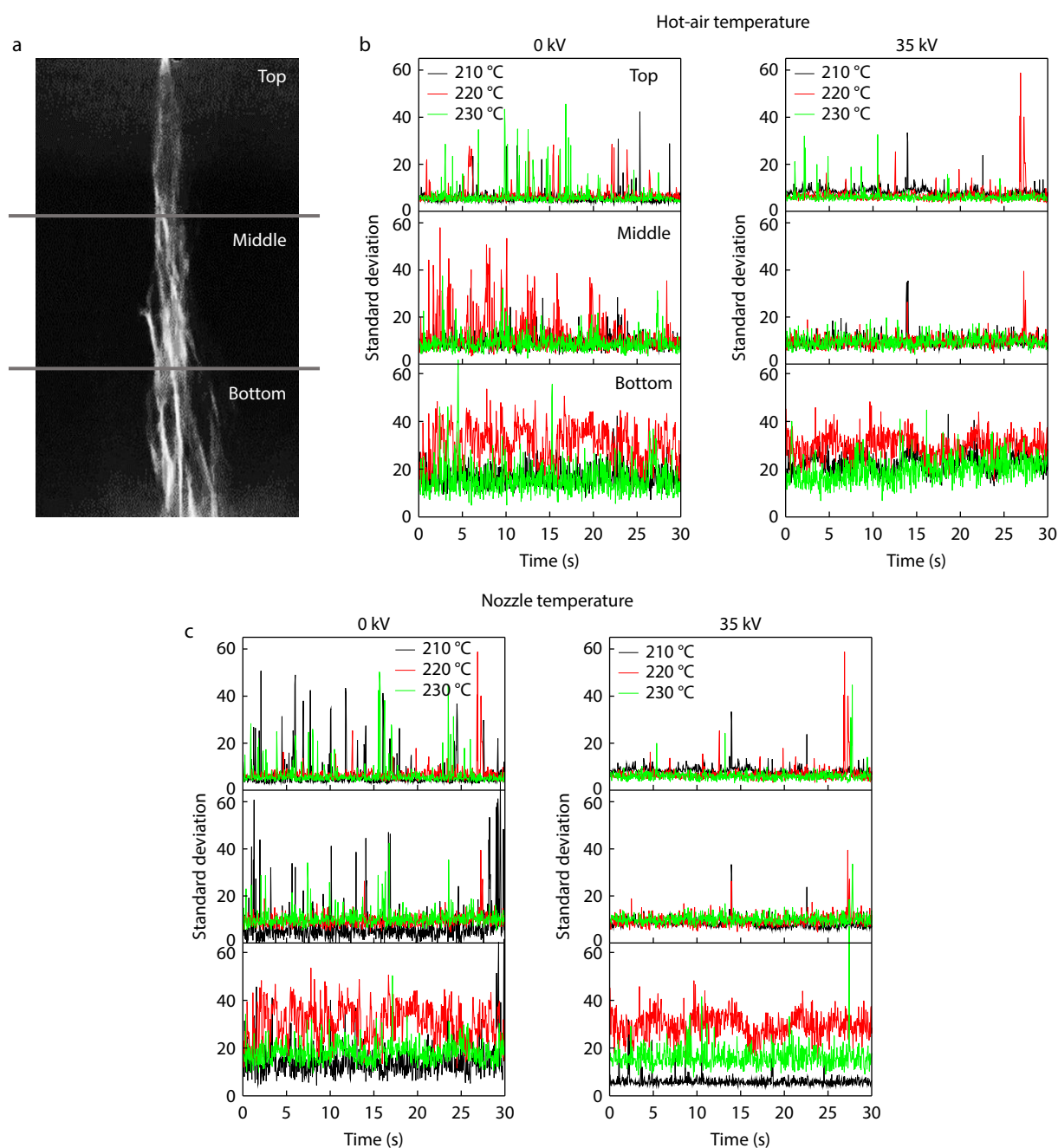


Fig. 3 (a) Jet standard deviation analysis method (b) Effect of hot air temperature variation (210, 220 and 230 °C) at a fixed nozzle temperature of 220 °C under two voltage conditions (0 and 35 kV) on the jet standard deviation; (c) Effect of nozzle temperature variation (210, 220 and 230 °C) at a fixed hot air temperature of 220 °C under two voltage conditions (0 and 35 kV) on the jet standard deviation.

length, resulting in a well-aligned trajectory with minimal fluctuations and low standard deviation. These findings indicated that a substantial reduction in viscosity under high-temperature conditions promotes dynamic alignment and stabilization of the jet. Interestingly, at an intermediate temperature of 220 °C, a transitional regime can be observed. Although the viscosity at this temperature was lower than at 210 °C, the jet exhibited spatially heterogeneous elongation and morphological development. In certain regions, the fibers were sufficiently elongated, whereas in others, the elongation was incomplete or irregular. This uneven behavior result-

ed in fluctuating jet motion and higher standard deviation values. These observations suggested that 220 °C represent a transitional thermal condition characterized by the coexistence of stable and unstable jet dynamics.

Furthermore, a distinct difference in jet behavior can be observed depending on the presence or absence of an applied electric field. In the absence of an electric field, the jet failed to maintain a consistent trajectory and exhibited unstable, irregular motion characterized by increased positional deviation and reduced coherence. This instability was attributed to the greater susceptibility of the jet to ambient disturbances

and the lack of electrostatic guiding forces. In contrast, when an electric field is applied, the electrostatic forces exert a stabilizing influence on the jet, guiding it along a more defined and focused path. Consequently, the jet flow became more aligned and uniform, leading to reduced fluctuations and a lower standard deviation. These findings confirm the critical role of the electric field in stabilizing the jet motion and promoting consistent fiber formation during the melt-spinning process.

Fig. 4 shows the calculated deposition area derived from the fiber deposition images. The experimental results revealed that the deposition area increased significantly when the applied voltage was increased to 35 kV. This behavior is attributed to the enhanced electrostatic repulsion between the charged fibers under a stronger electric field. As the mutual repulsive forces intensified, the fibers were pushed farther apart, resulting in a broader spatial distribution across the collector surface. These observations suggested that the electric field played a crucial role not only in shaping individual fibers but also in regulating their deposition and accumulation behavior.

Additionally, an increase in nozzle temperature led to an expanded deposition area. This trend was likely due to the enhanced melt flow rate at elevated temperatures, which resulted in a higher polymer discharge volume. The increased volume of the extruded material facilitated a more extensive distribution of fibers on the collector.

Overall, these findings indicated that both the electric field and nozzle temperature significantly influenced not only the fiber thinning but also the macroscopic deposition behavior and structural architecture of the resulting web. Therefore, precise optimization of these processing parameters is essential for controlling both the morphological characteristics of the individual fibers and the overall formation and uniformity of the deposited fibrous mat.

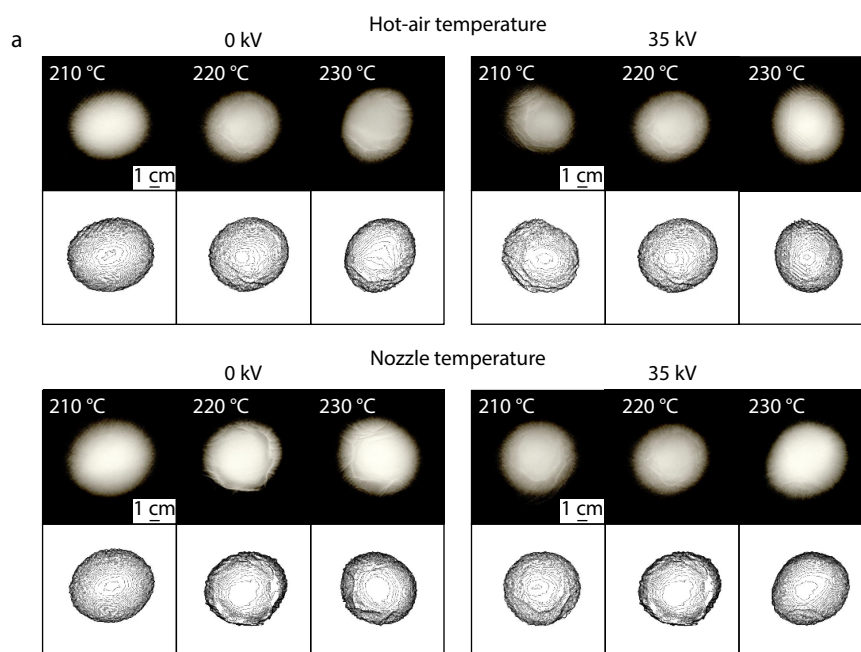
Fig. 5 shows the measured hot-air velocities at various tip-

to-collector distances (TCD). The experimental results demonstrated a gradual decrease in airflow velocity with increasing TCD. Immediately after the jet exited the nozzle, the airflow exhibited its highest velocity, which contributed to a more stable and uniform jet trajectory. However, as the jet travels further downstream, the surrounding airflow velocity decreased, resulting in weakened aerodynamic guidance. This reduction in airflow support led to increased irregularities in the jet path near the collector, as evidenced by the greater jet motion fluctuations and elevated standard deviation values.

According to the velocity measurements, the maximum and minimum airflow speeds were approximately 11 and 2 m/s, respectively. Based on these values, the estimated travel time of the polymer from the spinning nozzle to the collector ranged from approximately 0.009 s (at the highest velocity) to 0.05 s (at the lowest). These results indicated that the polymer jet is exposed to hot air for only a brief duration during its flight, particularly when compared with the residence time within the nozzle or extruder.

In contrast to the short exposure time, the polymer remained at elevated temperatures inside the nozzle for a substantially longer period. This prolonged thermal exposure increases the risk of thermal degradation or carbonization, thereby imposing a practical upper limit on the nozzle temperature in melt-based spinning systems. Conversely, the brief contact with hot air suggests that relatively higher hot-air temperatures can be applied without a significant risk of polymer degradation. Therefore, increasing the hot-air temperature represents a more effective and safer strategy for promoting fiber thinning while preserving the thermal stability of the polymer.

Fig. 6 presents SEM images of the polypropylene (PP) web fabricated using the hybrid melt electrospinning/blown process, which confirmed the successful formation of a uniform and stable fibrous network with no evidence of bead formation, indicating a consistent and defect-free spinning process.



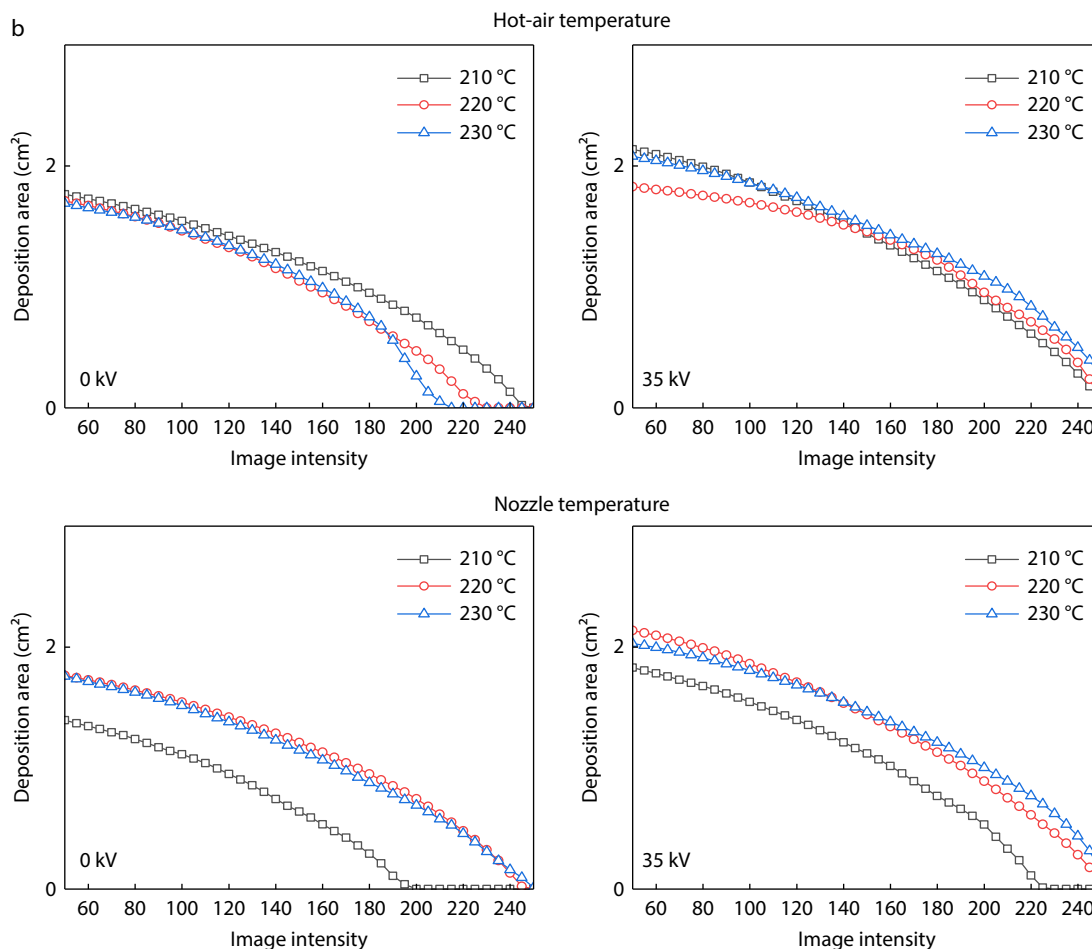


Fig. 4 (a) Processed web deposition images using thresholding and (b) the corresponding contour analysis results.

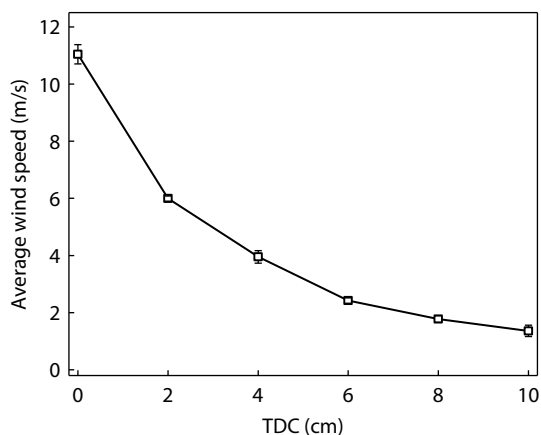


Fig. 5 Average air velocity as a function of tip-to-collector distance (TDC).

Fig. 7 shows the average fiber diameters of the resulting PP webs under various processing conditions. A clear trend was observed: increasing both the hot-air and nozzle temperatures led to a reduction in the fiber diameter. This behavior is attributed to two synergistic effects: (1) elevated hot-air temperatures increase the surrounding ambient temperature and reduce the cooling rate of the jet, thereby extending the effective elongation time; (2) higher nozzle temperatures re-

duce the viscosity of the polymer melt, enhancing its flowability and enabling more effective jet stretching.

In addition, the application of an electric field further contributes to the reduction in the fiber diameter. The enhanced electrostatic force between the charged jet and collector promoted additional elongation, facilitating the formation of finer fibers. Under optimized conditions, the finest average fiber diameter achieved in this study was approximately 860 nm, demonstrating the feasibility of producing nanoscale fibers using this solvent-free, melt-based hybrid process. Conversely, under the condition of a high nozzle temperature (230 °C) and no applied voltage (0 kV), a notable increase in fiber diameter was observed. This outcome is attributed to the increased polymer discharge rate caused by the reduced viscosity coupled with insufficient elongation in the absence of electrostatic forces. Furthermore, the lack of an electric field likely contributes to jet instability, leading to decreased fiber uniformity and a less homogeneous web morphology.

Fig. 8 presents the DSC thermograms and the FTIR spectra of PP fibers melt-spun under various nozzle and hot-air temperature conditions. DSC analysis revealed that all spun samples exhibited endothermic melting peaks at temperatures comparable to that of the PP pellet, with melting temperatures (T_m) ranging from 154 °C to 157 °C, indicating negligible variation. This suggests that, within the studied processing

temperature range, the spinning process did not induce any significant change in the crystalline melting point of PP, implying minimal thermal degradation or alteration of the crystalline structure.

FTIR analysis also shows no significant spectral deviations in the spun samples compared with the pristine PP pellet,

confirming that the chemical structure of the polymer was preserved during processing. The characteristic absorption bands of PP are clearly retained, including bands at 2950 and 2915 cm^{-1} corresponding to the asymmetric stretching vibrations of CH_3 and CH_2 groups, 1455 and 1375 cm^{-1} attributed to the bending vibrations of CH_2 and CH_3 , and 840 cm^{-1} asso-

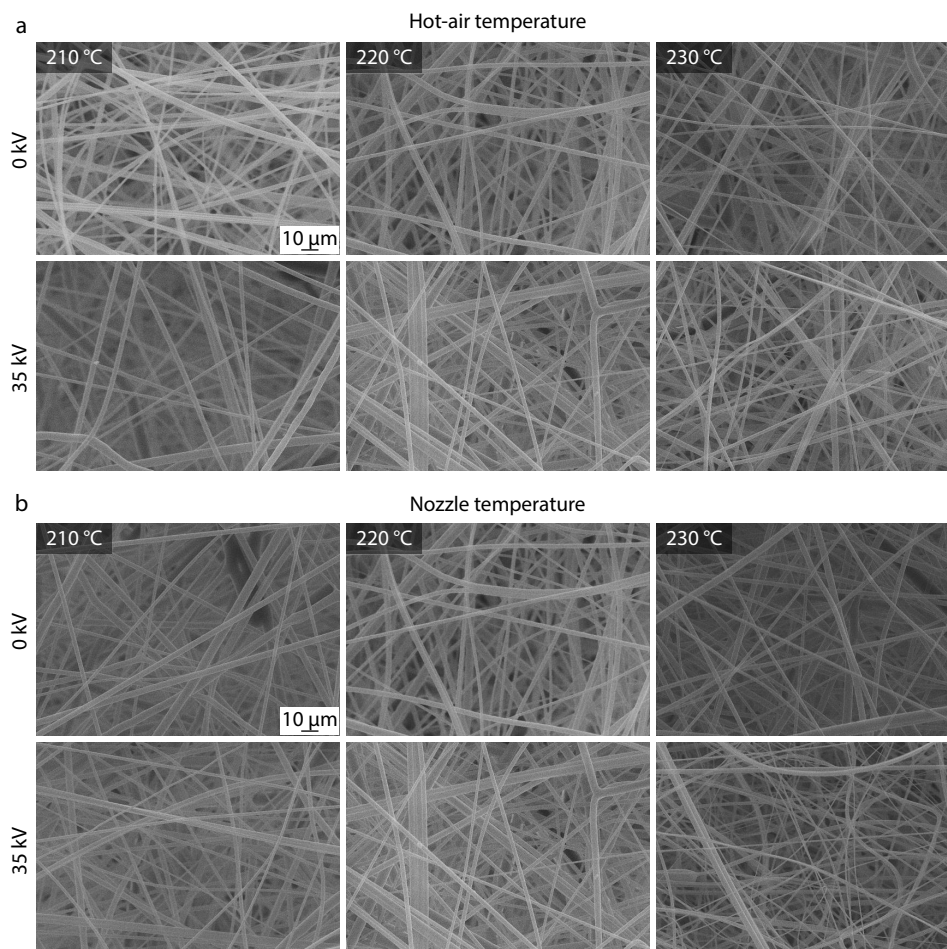


Fig. 6 (a) Effect of hot-air temperature (210, 220, and 230 °C) at a fixed nozzle temperature of 220 °C and (b) effect of nozzle temperature (210, 220, and 230 °C) at a fixed hot-air temperature of 220 °C on the SEM morphology of PP webs under two voltage conditions (0 and 35 kV).

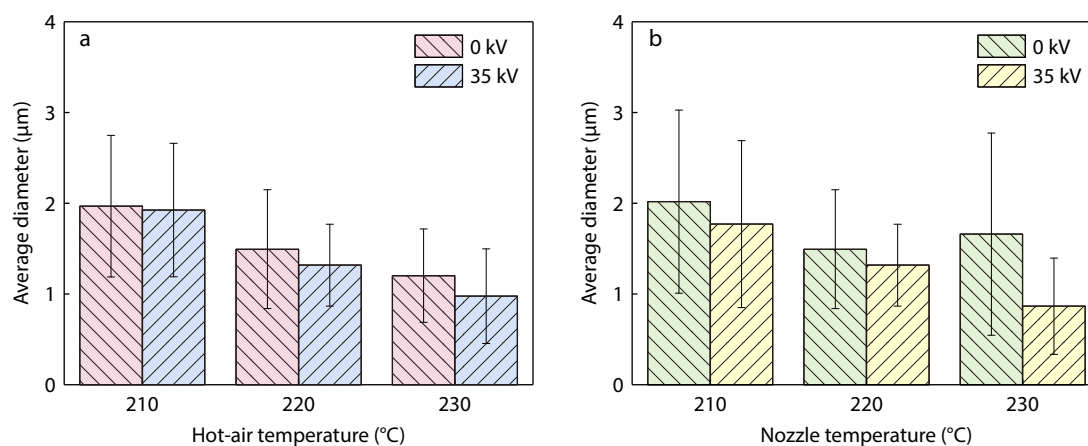


Fig. 7 (a) Effect of hot-air temperature (210, 220, and 230 °C) at a fixed nozzle temperature of 220 °C and (b) effect of nozzle temperature (210, 220, and 230 °C) at a fixed hot-air temperature of 220 °C on the average diameters of PP webs under two voltage conditions (0 and 35 kV).

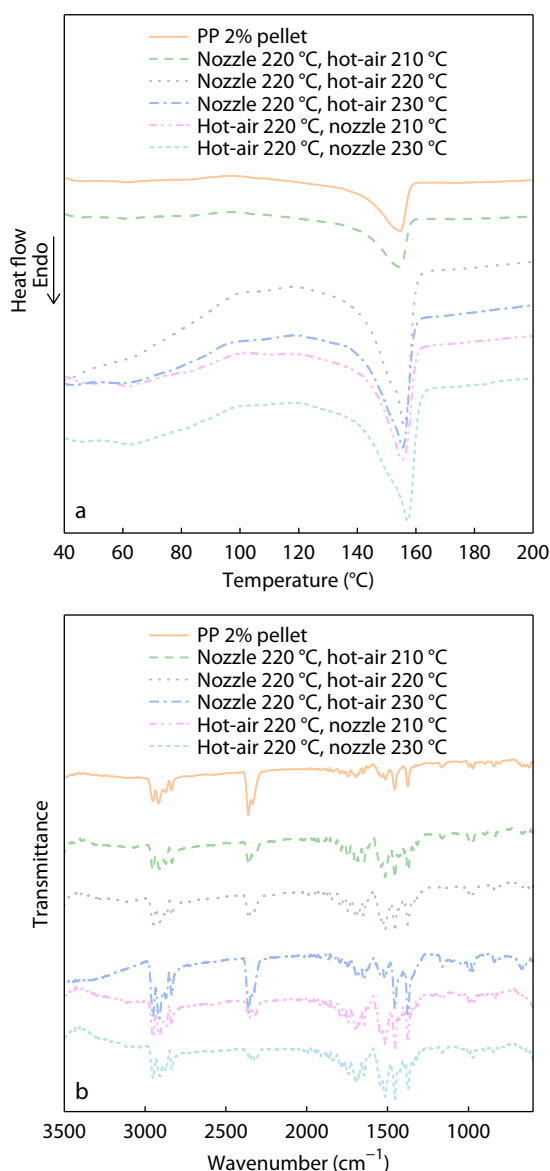


Fig. 8 (a) DSC curves and (b) FTIR spectra of PP pellet and melt-electrospinning/blown fibers.

ciated with the CH_2 rocking vibration in crystalline domains. All the spectra display a sharp band near 2349 cm^{-1} , assigned to the asymmetric stretching vibration of atmospheric CO_2 , which was excluded from the interpretation of the polymer structure.

Taken together, the DSC and FTIR results confirmed that the applied melt-spinning process conditions allowed fiber formation while maintaining the crystalline and chemical structure of PP.

CONCLUSIONS

This study demonstrated the feasibility of producing polypropylene (PP) microfibers ($<1\text{ }\mu\text{m}$) using a hybrid melt electrospinning/blown process. Among the three key processing parameters, nozzle temperature, hot-air temperature, and applied voltage, hot-air temperature was identified as the most effective

factor for promoting fiber thinning while maintaining polymer integrity and process stability. This could be attributed to the short exposure time between the polymer jet and hot-air stream, which minimized thermal degradation while facilitating stable elongation and jet alignment.

While increasing the nozzle temperature reduced the melt viscosity and improved flowability, it also increased the discharge rate and residence time, which introduced thermal degradation risks and limited its ability to refine the fibers beyond a certain threshold. Similarly, applying a high voltage enhanced electrostatic stretching but posed electrical discharge risks in upscaled systems and introduced lateral instabilities. In contrast, elevated hot air temperatures not only improved fiber fineness but also maintained uniform deposition and suppressed jet fluctuations.

The optimized process conditions enabled the formation of uniform, bead-free micro fibers with average diameters as small as $0.86\text{ }\mu\text{m}$, demonstrating the potential of this hybrid system for scalable, solvent-free, and thermally stable fabrication of advanced separator materials. Moreover, this study systematically quantified the effects of key thermal and electrical parameters on the jet behavior, fiber morphology, and web uniformity using real-time imaging and statistical image analysis.

Overall, these findings provide a comprehensive understanding of the parameter interplay in hybrid melt electrospinning/blown systems and highlight hot-air-assisted processing as a technically robust and industrially viable strategy for the development of next-generation lithium-ion battery separators.

Conflict of Interests

The authors declare no interest conflict.

Data Availability Statement

The datasets generated and analyzed during the current study are available from the corresponding author upon reasonable request.

ACKNOWLEDGMENTS

This work was financially supported by the Technology Innovation Program (or Industrial Strategic Technology Development Program) (No. 20017666) funded by the Ministry of Trade, Industry, and Energy (MOTIE, Korea).

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