Preparation of Hydrophobized Microfibrillated Cellulose Powder by Alkyl Ketene Dimer Treatment and Spray Drying

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Microfibrillated cellulose (MFC) is a sustainable, bio-based material with remarkable mechanical properties and a large specific surface area. However, its inherent hydrophilicity limits its application in hydrophobic composite systems. In this study, hydrophobized MFC powders were prepared by treating MFC suspensions with alkyl ketene dimer (AKD) and then spray-drying. The MFC suspensions were treated with varying AKD dosages (1 to 3%, based on oven-dried MFC) and spray-dried at different inlet temperatures to determine the best hydrophobization conditions. Contact angle measurements indicated that hydrophobicity increased with rising AKD dosage, and the maximum contact angle was obtained at an inlet temperature of 120 °C. Fourier transform infrared analysis supported the retention of AKD-derived hydrophobic alkyl groups in the spray-dried MFC powders. Field-emission scanning electron microscopy images further revealed morphological changes from fibrillated structures to compact particulate morphologies after AKD treatment and spray drying. Notably, high hydrophobicity was achieved without an additional curing process, indicating that spray drying provided sufficient thermal energy for AKD-mediated hydrophobization. These results demonstrate that spray drying combined with AKD treatment can effectively produce hydrophobic MFC powders potentially suitable for composite applications.

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Keywords: Microfibrillated cellulose (MFC); Alkyl ketene dimer (AKD); Spray drying; MFC powder; Contact angle; Hydrophobicity

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INTRODUCTION

Microfibrillated cellulose (MFC) has recently attracted considerable attention as a sustainable, renewable reinforcing material due to its high aspect ratio, large specific surface area, excellent mechanical strength, and biodegradability (Nishiyama 2009; Lavoine *et al*. 2012). Together, these distinctive characteristics have prompted extensive investigation of MFC for use in paper additives, coating materials, packaging, barrier layers, and polymer composites (Lavoine *et al.* 2014; Lendvai *et al.* 2016). Incorporating MFC into thermoplastic matrices, such as polypropylene, has been regarded as a promising strategy for developing lightweight, eco-friendly composite materials with enhanced mechanical performance (Iwamoto *et al.* 2014; Lendvai *et al.* 2016).

Despite these advantages, however, incorporating MFC into hydrophobic polymer systems remains challenging because cellulose fibrils contain abundant surface hydroxyl groups, which confer strong hydrophilicity and poor compatibility with nonpolar polymers. This hydrophilicity often leads to insufficient interfacial adhesion and nonuniform dispersion within polymer matrices, thereby limiting the reinforcing efficiency of MFC in composite applications (Iwamoto *et al.* 2014; Palange *et al.* 2019; Guo *et al.* 2025). Therefore, surface hydrophobization is essential to improve MFC compatibility with hydrophobic polymers and expand its applicability in thermoplastic composite systems (Yan *et al.* 2015).

Among various hydrophobization agents, alkyl ketene dimer (AKD) is widely used as an effective internal sizing material in the paper industry. AKD introduces hydrophobic alkyl chains onto cellulose surfaces. Specifically, AKD can react with cellulose hydroxyl groups through β-keto ester formation, thereby reducing surface polarity and improving water repellency. Hydrophobic surface modification with AKD may also reduce interfibrillar hydrogen bonding during drying, thereby influencing particle aggregation and powder formation behavior (Hubbe 2007; Liu *et al*. 2023; Aziz and Salleh 2024; Onodera *et al.* 2025). Notably, industrial thermoplastic processing typically requires MFC in dry powder form rather than in aqueous suspension (Yan *et al.* 2015). However, drying MFC remains challenging because strong capillary forces generated during water removal often induce irreversible fibril aggregation and structural collapse (Žepič *et al.* 2014; Silva *et al.* 2021). Thus, spray drying is a promising method for producing dry MFC powders, as it enables continuous operation, rapid drying, and industrial scalability (Peng *et al.* 2012; Lopes *et al.* 2021). Nevertheless, spray-drying conditions must be optimized because drying behavior strongly influences powder morphology and surface characteristics (Nardi *et al.* 2020; Hwang *et al.* 2023). Moreover, hydrophobic surface modification before spray drying may alter droplet drying behavior and influence the final morphology and hydrophobicity of the resulting powder (Wang *et al.* 2016).

Although several studies have examined MFC hydrophobization or powder production independently (Lopes *et al.* 2021; Blok *et al.* 2022; Yang *et al*. 2025), systematic studies on the combined effects of AKD treatment and spray drying conditions for producing hydrophobic MFC powders remain limited. In particular, the relationship among AKD dosage, spray-drying temperature, and the development of hydrophobicity remains unclear. To bridge this gap, this study investigated the preparation of hydrophobized MFC powders through AKD treatment combined with spray drying. The effects of inlet drying temperature and AKD dosage on hydrophobicity and powder morphology were evaluated using contact angle measurements, Fourier transform infrared (FT-IR) spectroscopy, and field-emission scanning electron microscopy (FE-SEM) to determine the best preparation conditions for hydrophobic MFC powders.

EXPERIMENTAL

Materials

Bleached hardwood kraft pulp (HwBKP), supplied by Moorim Paper Co., Ltd. (Jinju, Republic of Korea), was used to prepare MFC in a laboratory. AKD emulsion was supplied by LATON Co., Ltd. (Gongju, Republic of Korea), and its specifications are listed in Table 1. Ethyl alcohol (C_2H_5OH, 95.00%; Daejung, Republic of Korea), acetone (CH_3COCH_3, 99.9%; Fisher, USA), and n-hexane (C_6H_{14}, 86.18%; Daejung, Republic of Korea) were used for solvent exchange.

Table 1. AKD Specifications

Solid content (%)	Average particle size (μm)	Average zeta-potential (mV)
15 ± 0.5	0.68	+28.0 (at pH 7.0)

The zeta potential of the MFC suspension was not directly measured in this study; therefore, numerical MFC zeta-potential data were not added. However, cellulose microfibrils are generally expected to exhibit a negative surface charge in aqueous media (Lee *et al*. 2009). It follows that electrostatic attraction between anionic MFC surfaces and cationic AKD particles may have contributed to AKD adsorption before spray drying.

Preparation and Characterization of MFC

HwBKP with 1.57% solids was soaked in tap water and then beaten to 300 ± 5 mL Canadian Standard Freeness using a laboratory Hollander beater. To minimize clogging during high-pressure homogenization, the beaten pulp slurry was diluted to 0.7% solids and fibrillated for up to four passes using a high-pressure homogenizer (MN400BD; Micronox, Gwangju, Republic of Korea) at 20,000 psi. The MFC sample was collected after the fourth pass.

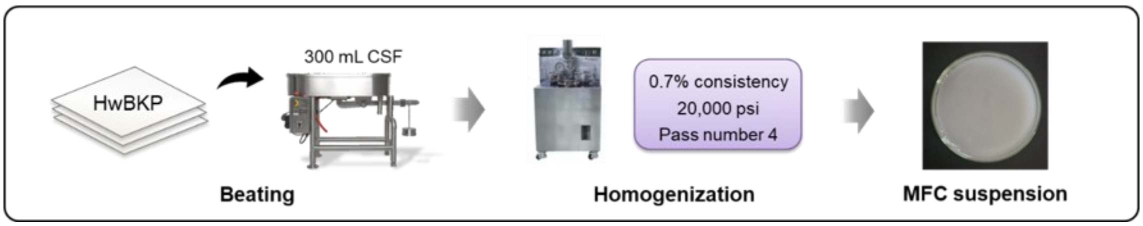

Fig. 1. Flow diagram of the mechanical treatments for preparing MFC made from HwBKP

Wet MFC test specimen pads were prepared by vacuum filtration for fiber-width measurement. To prevent deformation during drying, the test specimens were dried by solvent exchange with ethyl alcohol, acetone, and n-hexane (Stelte and Sanadi 2009; Kim *et al*. 2019). Before FE-SEM observation, the dried MFC specimen pads were sputter-coated with a thin conductive platinum layer to prevent charging during imaging. Because the conductive coating can slightly increase the apparent fiber width, all samples were prepared and imaged under identical coating and measurement conditions. FE-SEM images of the pads were acquired using a JSM-7610F instrument (JEOL, Tokyo, Japan), and the length and width of 100 individual microfibers from the MFC specimen were measured using 3D image software (MP-45030TDI; JEOL, Osaka, Japan). The aspect ratio of MFC was calculated from the measured fiber length and width.

The low-shear viscosity of the 1% MFC suspension was directly measured at room temperature using a low-shear viscometer (DV-IP; Brookfield Engineering Laboratories, Middleborough, MA). The reported viscosity value was obtained from the instrument reading after stabilization under the selected measurement condition.

Hydrophobic MFC Powder Preparation and Physical Property Measurement

MFC powders were prepared as follows to determine the best spray drying inlet temperature for MFC hydrophobicity development and to evaluate the effects of AKD dosage on MFC hydrophobicity and morphology. The MFC suspension was adjusted to

2%, and AKD was added at 1%, 2%, or 3% based on the oven-dried weight of MFC (Tak *et al*. 2025). The mixture was then stirred for 30 min at 600 rpm. Subsequently, the AKD-treated MFC suspension was spray-dried in a laboratory-scale spray dryer (Mini Spray Dryer S-300; Buchi, Switzerland) without an additional AKD curing step. This approach assumed that rapid moisture removal and high-temperature exposure during spray drying could promote sufficient AKD distribution and partial reaction on the MFC surface. The spray drying inlet temperature ranged from 110 to 150 °C. To determine the best inlet temperature, the feed rate was set to 10 mL/min using a silicone tube with an inner diameter of 2 mm. During spray drying, all operating parameters, except the inlet temperature, were maintained at the optimized conditions for MFC powder production.

The contact angle measurements were conducted according to TAPPI T 458 (2014), using pelletized MFC powder specimens prepared for analysis with a contact angle analyzer, with ferric chloride ($FeCl_3$) solution as the probe liquid (Lee *et al.* 2012, 2014). The colored ferric chloride solution was used instead of pure water to facilitate clear visual detection of liquid spreading and penetration on the pelletized MFC powder specimens. The concentration of the ferric chloride solution was 3%. Therefore, the contact angle values reported in this study should be regarded as apparent contact angles measured using a 3% ferric chloride solution as the probe liquid. FT-IR analysis was performed using an IS50 instrument (Thermo Fisher Scientific, USA) to confirm AKD incorporation into the MFC structure. For hydrophobicity evaluation, the powders were pelletized before contact angle measurement. Powder morphology was then examined by FE-SEM using a JSM-7610F instrument (JEOL, Japan) to evaluate the effects of AKD treatment and spray drying on MFC morphology.

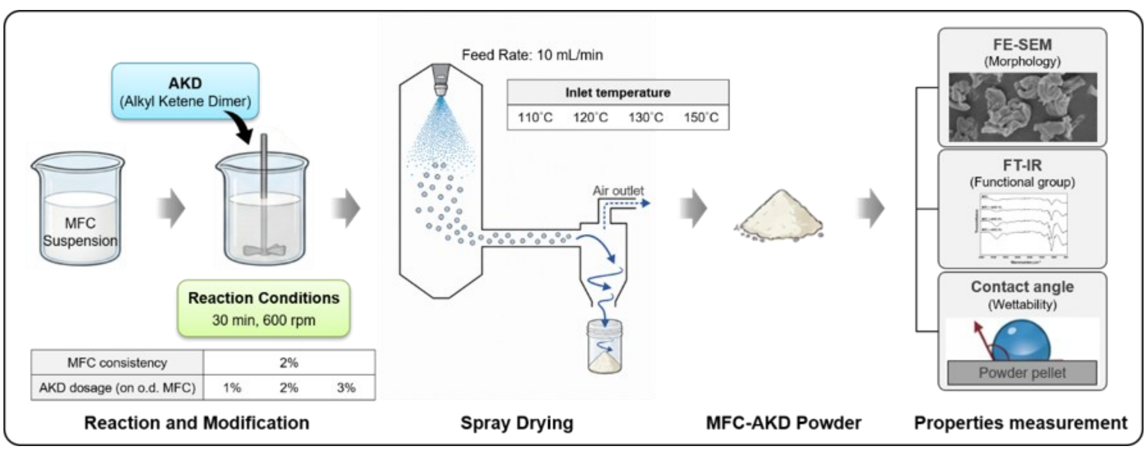

Fig. 2. Flow diagram of AKD-modified MFC powder preparation from an MFC suspension and characterization of the resulting powder

RESULTS AND DISCUSSION

Characteristics of MFC and Determination of the Spray Drying Inlet Temperature

Figure 3 presents FE-SEM images, and Table 2 lists the characteristics of the MFC prepared from HwBKP. The original HwBKP fibers were extensively delaminated into highly entangled fibrillar structures, forming a dense nanofibrillar network. The MFC had an average fiber length of 5.4 ± 1.5 μm and a nanoscale fiber width of 29.8 ± 15 nm, resulting in a high aspect ratio of 188 ± 111. Together, these results indicate that the mechanical treatment achieved sufficient fibrillation and successfully converted HwBKP into MFC.

The nanoscale fiber width and high aspect ratio further confirmed the production of a well-developed MFC with a highly fibrillated morphology. Furthermore, the high viscosity value of 890 cPs, measured at an MFC solids content of 1%, supported the formation of an entangled microfibrillar network with increased surface area and strong interfibrillar interactions. These results are consistent with previous studies reporting that mechanical fibrillation decreases cellulose fiber width to the nanoscale, increases aspect ratio, and increases suspension viscosity through the formation of an entangled microfibrillar network.

Although a few partially fibrillated fiber bundles remained, most fibers were effectively converted into microfibrillated structures. Therefore, the present results confirmed that the applied mechanical treatment effectively produced highly fibrillated MFC from HwBKP (Klemm *et al.* 2011; Ankerfors 2012).

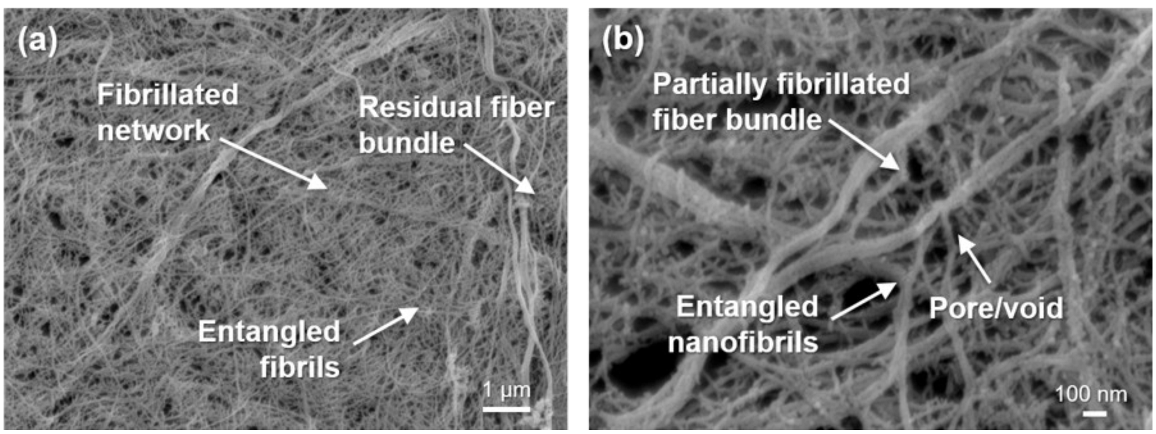

Fig. 3. FE-SEM images of MFC prepared from HwBKP at (a) 10,000× and (b) 50,000× magnification

Table 2. Characteristics of MFC

Pass number	Fiber length (μm)	Fiber width (nm)	Aspect ratio	Viscosity (cPs)
4	5.4 ± 1.5	29.8 ± 15	188 ± 111	890

Figure 4 shows the contact angle for hydrophobized MFC powders prepared by spray drying an MFC suspension containing 3% AKD at different inlet temperatures. All AKD-treated samples exhibited contact angles above approximately 100°, indicating sufficient hydrophobicity regardless of drying temperature. Among the tested conditions, the sample prepared at 120 °C exhibited the maximum contact angle, indicating the most effective hydrophobization of the MFC surface. The higher contact angle at 120 °C can be attributed to a balance between sufficient thermal exposure for AKD mobility and fixation and controlled moisture removal during spray drying. At lower temperatures, AKD distribution or fixation may be insufficient, whereas at higher temperatures, rapid skin formation and accelerated water evaporation may limit the uniform migration of AKD to the MFC surface. Notably, high hydrophobicity was achieved without additional curing, indicating that spray drying provided sufficient thermal energy for AKD hydrophobization. Therefore, 120 °C was selected as the best inlet temperature for producing hydrophobized MFC powder by spray drying without additional curing.

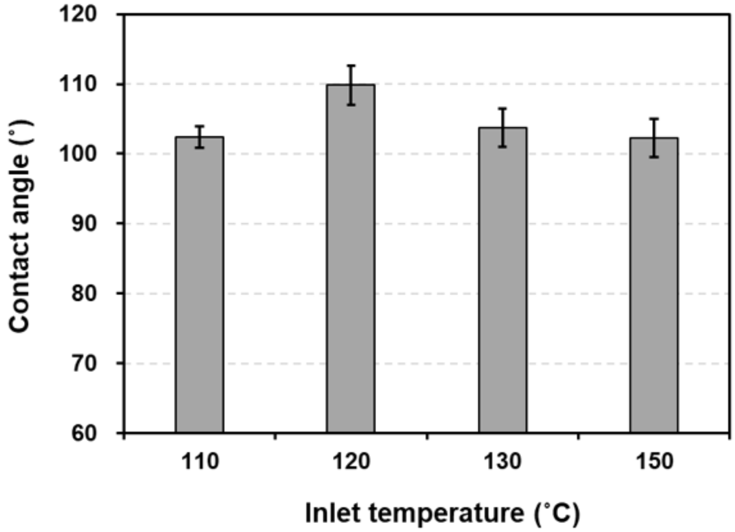

Fig. 4. Contact angle of AKD-modified MFC powder depending on the inlet temperature of the spray dryer

Hydrophobicity Measurement of AKD-modified MFC Powders

Figure 5 presents the contact angles of spray-dried MFC powders prepared with different AKD dosages. The contact angle increased progressively with increasing AKD dosage, indicating that AKD treatment during spray drying effectively enhanced the hydrophobicity of MFC powder. Untreated MFC exhibited a relatively small contact angle, likely because of the abundant hydroxyl groups on the cellulose surface. By contrast, the contact angle increased from approximately 42° for the untreated MFC powder to approximately 109° for the 3% AKD-treated powder.

The progressive increase in contact angles with AKD dosage indicates that more AKD-derived hydrophobic components were retained on the MFC surface. As the AKD dosage increased, the surface coverage of long alkyl chains likely increased, reducing the exposure of hydrophilic cellulose hydroxyl groups and decreasing surface polarity. This interpretation is supported by the increased intensity of C–H stretching bands in the FT-IR spectra (Fig. 6).

In particular, the sample containing 3% AKD exhibited the highest contact angle, suggesting effective AKD retention and possible fixation on the MFC surface during spray drying. Overall, these results demonstrated that spray drying combined with AKD treatment could effectively produce hydrophobized MFC powders without an additional curing process. The achievement of high contact angles without an additional curing step suggests that the spray-drying process supplied sufficient thermal energy for AKD-mediated hydrophobization. During spray drying, the atomized MFC droplets were rapidly exposed to hot air, which accelerated water evaporation, concentrated AKD near the cellulose surface, and may have promoted AKD fixation or strong retention on the fibril surface. Therefore, separate oven curing was not required to obtain hydrophobic MFC powder under the present conditions.

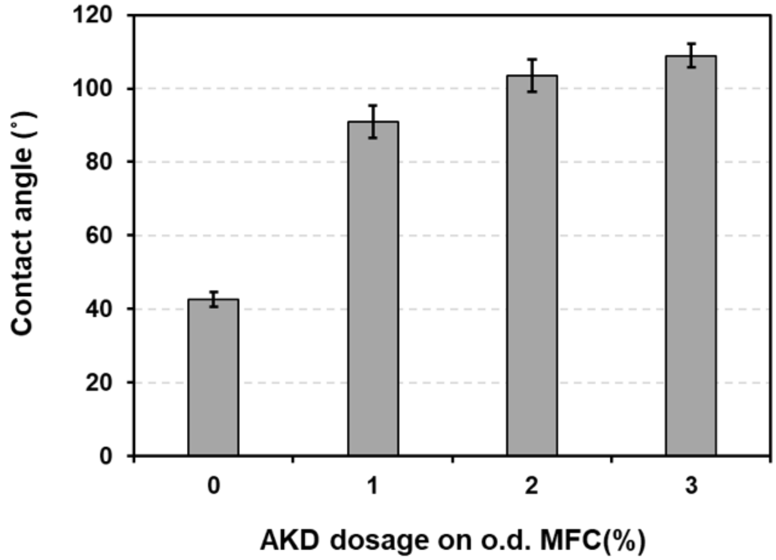

Fig. 5. Contact angle of AKD-modified MFC powders as a function of AKD dosage at an inlet temperature of 120 °C

Figure 6 shows the FT-IR spectra of spray-dried MFC powders prepared with different AKD dosages. The untreated MFC powder exhibited the characteristic absorbance bands of cellulose, including a broad band in the region of approximately 3,200 to 3,500 cm^{-1}, attributed to O–H stretching vibrations, as well as strong absorbance bands in the fingerprint region corresponding to C–O and C–O–C stretching vibrations of the cellulose backbone (Poletto *et al.* 2014). As the AKD dosage increased, distinct spectral changes became more evident. In particular, the absorbance intensity in the C–H stretching region at approximately 2,850 to 2,920 cm^{-1} increased with increasing AKD dosage. This change was attributed to the long alkyl chains of AKD, indicating that AKD-derived hydrophobic components were retained on the MFC surface after spray drying. The gradual increase in the alkyl-chain-related absorbance bands was consistent with the increase in contact angle observed for the AKD-treated MFC powders (Yan *et al.* 2016).

In addition, changes were observed in the carbonyl region at 1,700 to 1,750 cm^{-1} after AKD treatment. This region may be associated with carbonyl-containing groups derived from AKD, including possible β-keto ester linkages formed between AKD and cellulose hydroxyl groups. However, contributions from residual or hydrolyzed AKD cannot be excluded (Kumar *et al.* 2016; Onodera *et al.* 2025). Thus, while the FT-IR spectra support the retention and potential fixation of AKD-derived components on the MFC surface, they do not provide conclusive evidence for the formation of covalent β-keto ester bonds (Yan *et al.* 2016; Onodera *et al.* 2025).

Overall, the FT-IR analysis showed that increasing the AKD dosage increased the presence of hydrophobic alkyl moieties on the spray-dried MFC surface. These spectral changes, in conjunction with the contact angle measurements, suggested that AKD treatment combined with spray drying effectively improved the hydrophobicity of MFC powders without requiring an additional post-curing step.

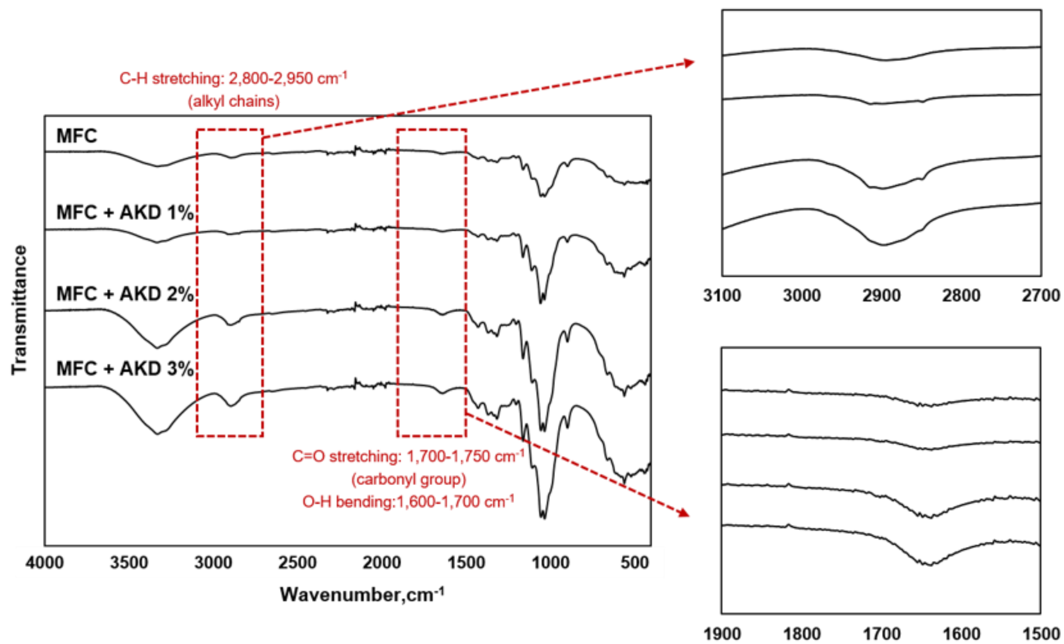

Fig. 6. FT-IR spectra of AKD-modified MFC powders depending on AKD dosage at the inlet temperature of 120 °C

Figure 7 shows the FE-SEM images of spray-dried MFC powders prepared with different AKD dosages. Untreated MFC exhibited a relatively fibrillated and aggregated morphology, with exposed fibrillar networks on the particle surface. In contrast, the AKD-treated samples formed more compact particulate structures with less exposed fibrillar networks. At higher magnifications, wrinkled surfaces and collapsed particle structures, which are typical morphologies of spray-dried cellulose-based materials, were observed.

The compact particle structure and smoother surface observed in the AKD-treated samples may have contributed to the increased contact angle by reducing the exposure of hydrophilic cellulose fibrils and hydroxyl-rich surfaces. As the AKD dosage increased, the particle surfaces became denser and smoother, suggesting that AKD-derived hydrophobic components were more uniformly distributed or retained on the MFC surface during rapid drying. This AKD surface coverage likely reduced the surface polarity of the MFC powders and increased the contribution of hydrophobic alkyl chains at the outer surface, thereby enhancing water repellency.

These morphological changes are consistent with the FT-IR and contact angle results, confirming that AKD treatment combined with spray drying successfully produced hydrophobized MFC powders. Although the FE-SEM images clearly showed the formation of compact, particle-like morphologies after AKD treatment and spray drying, quantitative particle-size distribution analysis was not performed in this study. Therefore, FE-SEM observations were used to qualitatively compare morphological changes among the samples. Quantitative particle-size analysis should be performed in future work to more precisely evaluate the relationship among AKD dosage, spray-drying behavior, surface morphology, and powder hydrophobicity.

Therefore, the contact angle, FT-IR, and FE-SEM results consistently confirmed that hydrophobic MFC powders were successfully fabricated using AKD treatment combined with spray drying.

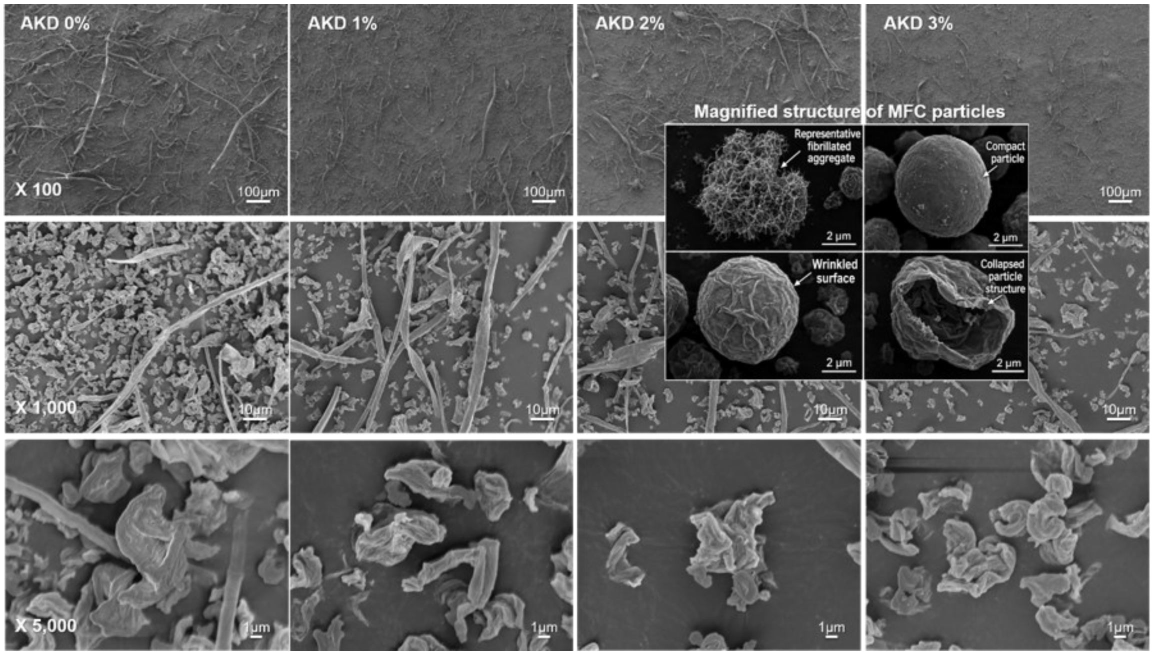

Fig. 7. FE-SEM images of AKD-modified MFC powders as a function of AKD dosage at an inlet temperature of 120 °C

CONCLUSIONS

1. Hydrophobic microfibrillated cellulose (MFC) powders were successfully prepared by combining alkylketene dimer (AKD) treatment with spray drying. The contact angle increased with increasing AKD dosage, indicating that AKD-derived hydrophobic components were effectively retained on the MFC powder surface.
2. Among the tested spray-drying inlet temperatures, 120 °C gave the highest contact angle. In addition, among the tested AKD dosages, 3% of AKD based on oven-dried MFC produced the highest hydrophobicity. Therefore, 3% AKD and 120 °C were identified as the best preparation conditions considered in this study.
3. High hydrophobicity was achieved without an additional curing process, suggesting that rapid moisture removal and thermal exposure during spray drying were sufficient to promote practical AKD-mediated hydrophobization under the present conditions.
4. Fourier transform infrared (FT-IR) analysis supported the retention of AKD-derived hydrophobic alkyl groups in the spray-dried MFC powders. However, FT-IR alone could not provide conclusive evidence for the formation of covalent β-keto ester bonds between AKD and cellulose.
5. Field emission scanning electron microscopy (FE-SEM) observations showed that AKD treatment combined with spray drying altered the morphology from exposed, fibrillated aggregates to more compact, particle-like structures. These morphological changes, together with AKD-derived hydrophobic surface components, contributed to the increased water repellency of the MFC powders.

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