Improved barrier performance of water-based barrier coatings with microfibrillated cellulose

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ABSTRACT: In this work, the impact of microfibrillated cellulose (MFC) on the properties of water-based barrier coatings intended for food packaging have been explored. Commercially available MFC was used for improving the rheology and water retention of three different commercially available dispersion coatings (acrylic, styrene acrylic, and polylactic acid). Coatings were applied by rod to paper, and barrier properties were tested by measuring air permeability and water barrier properties. Results clearly showed that addition of MFC to water-based dispersion coatings improved the barrier performance of the final coatings.

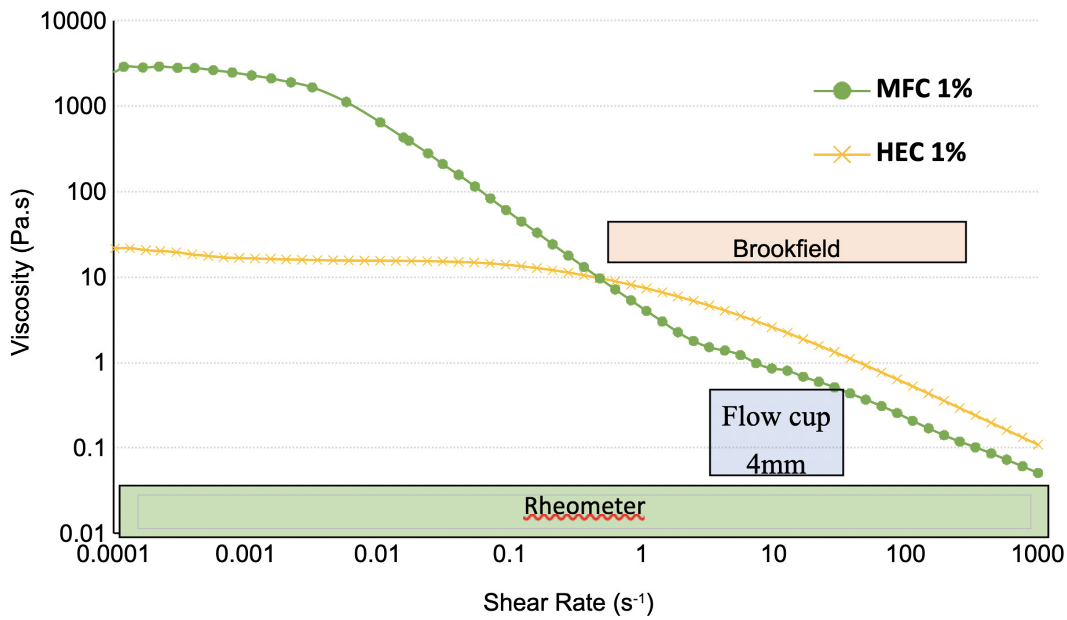
Application: The present work shows how MFC can be used to improve barrier performance, applicability, and drying properties of water-based barrier coatings. It is possible to reduce the coat weight and still obtain better barrier performance against grease and water, for example, even on a porous paper. This will have a strong impact on the packaging's sustainability and raw materials costs and help obtain a plastic:fiber ratio that meets the limits set by regulations.

Water-based barrier coating formulations for fiber-based food packaging are evolving to meet the demands for recyclable, bio-degradable, and compostable materials. However, replacement of traditional extruded coatings by dispersion coatings is not straightforward. In order to obtain optimal barrier properties, the coating needs to have a balance between water retention capacity and penetration rate into the paper substrate, among other requirements.

Dispersion barrier coatings are applied on various fiber substrates, ranging from wrapping papers to boards and molded fiber products. Due to differences in packaging surface properties, it is typically difficult to obtain good barrier properties with one dispersion coating formulation using traditional technologies. Influencing factors for a successful barrier coating result include the packaging material's porosity, smoothness, and whether the coating is applied on a horizontal or a vertical surface.

Microfibrillated cellulose (MFC) is a biobased and multifunctional product comprised of cellulose. It consists of fibrils with lateral dimensions below micron size and lengths up to micron scale [1]. The extremely high surface area of MFC, with numerous OH-groups adorning the three-dimensional fibril structure, impacts water retention capacity of barrier coatings. In addition, its structural properties help control the low shear viscosity of the coating without impacting high shear viscosity. Its low extensional viscosity allows easy spraying of the finished coatings. In addition, MFC reduces skinning of the drying film, allowing for faster drying processes.

The rheological properties of MFC suspensions are quite different compared to common low shear thickeners used in dispersion coatings. The impact of MFC on mid shear viscosity is low, and it basically has no impact on the high shear area. Kumar et al. studied the rheology of MFC suspensions using a slot geometry with different MFC concentrations up to shear rates of 100,000 s^{-1} [2]. At 1% MFC concentration, the apparent viscosity at a shear rate of 100,000 s^{-1} was around 4 mPas. Therefore, it can be concluded that MFC does not impact coating process runnability at high shear rates (> 500,000 s^{-1}). As can be seen from **Fig. 1**, MFC has a strong impact on low shear/no shear viscosity, showing 100 times higher viscosity compared to a highly viscous hydroxyethyl cellulose (HEC). This high viscosity at low shear is advantageous to coating. For example, MFC allows for control over film thickness on all surfaces of a 3D-shaped article when spray coating. The unique shear-thinning behavior causes some practical challenges for analysis of the thickening effects of MFC in dispersion coatings and its impact on runnability. When traditional methods such as Brookfield or Zahn cup are used, the impact of MFC on viscosity is often misjudged. One of the reasons is that flow cups are designed for Newtonian thickeners. They do not reliably work with pseudoplastic thickeners. Also, a Brookfield viscometer measures a narrow range of viscosities that do not represent the actual production conditions. To get a realistic picture of high shear viscosity, either a rheometer or capillary rheometer should be used. As discussed previously, MFC has extremely low impact on high shear viscosity. This means that in most of the cases, Newtonian thickeners, especially in high-speed coating processes, should be added to ensure good runnability.



1. Flow curves for 1.0% (wt%) of microfibrillated cellulose, MFC (dot); and 1% hydroxyethyl cellulose, HEC (square).

We will demonstrate the effect of commercially available MFC on the rheological properties and water retention of different water-based dispersion barrier coatings, including acrylic, styrene acrylic (SA), and polylactic acid (PLA). We will also show the impact of MFC on drying coated products and final barrier properties. Additionally, we will discuss the effect of the fibrillation degree on stabilization and film properties.

MATERIALS AND METHODS

Commercially available highly fibrillated MFC from Borregaard (Sarpsborg, Norway) was used in the study as a 10 wt% paste (Exilva F 01-V). The formulations used in this study (**Tables I–IV**) were prepared as follows. The MFC was dispersed in water, defoamer, and approximately a quarter of the corresponding binder using a Dispermat (Columbia, MD, USA) mixer at a tip speed of 6 m/s for 15 min. The remaining amount of binder was added during low shear mixing. Binders were received from their producers:

- NeoCryl A-2092 styrene acrylic, designated as SA1, from Covestro (Leverykusen, Germany).
- JonCryl HPB 4011 styrene acrylic, designated as SA2, from BASF (Ludwigshafen, Germany).
- Joncryl ECO 2189 styrene acrylic from BASF, designated as SA3.
- PL-1005 polylactic acid, designated as PLA1, from Miyoshi Oil and Fat Ltd. (Tokyo, Japan).
- PL-3000 polylactic acid, designated as PLA2, from Miyoshi Oil and Fat Ltd.

High shear viscosity of each formulation was measured with an MCR 301 rheometer from Anton Paar (Graz, Austria) equipped with stainless steel bob-cup. The shear rate range used was between 0.005–5000 s^{-1}. Papers were coated with the different formulations for analysis of air permeance (Gurley) and resistance to oil and water. The impact of MFC on the film properties was tested by applying 3 g of the Formulation 2 (Table III) on a plastic petri dish (90-mm diameter). The coatings were dried at

Ingredients	Reference	1% MFC	2% MFC	3% MFC
SA1 (binder)	96.0	96.0	96.0	96.0
MFC (10% paste)	-	1.0	2.0	3.0
Defoamer	0.2	0.2	0.2	0.2
Water	3.8	2.8	1.8	0.8
Total	100	100	100	100

I. Formulation for testing the impact of microfibrillated cellulose (MFC) on water holding of a dispersion coating (SA: styrene acrylic).

Ingredients	Reference	1% MFC	2% MFC	3% MFC
SA1 (binder)	325.6	325.6	325.6	325.6
Isopropanol (solvent)	17.2	17.2	17.2	17.2
Ethyldiglycol (coalescing)	8.4	8.4	8.4	8.4
Wax	36	36	36	36
MFC (10% paste)	0	4	8	12
Defoamer	0.8	0.8	0.8	0.8
Water	12	8	4	
Total, g	400	400	400	400

II. Formulation 1 based on modified styrene acrylic (SA) copolymer.

Ingredients	Reference	1% MFC	2% MFC	3% MFC
SA2 (binder)	322	322	322	322
SA3 (binder)	36	36	36	36
Rheology modifier	1.2	0	0	0
Defoamer	0.8	0.8	0.8	0.8
MFC (10% paste)	0	4	8	12
Water	40	37.2	33.2	29.2
Total, g	400	400	400	400

III. Formulation 2 based on styrene acrylic (SA) copolymer.

Ingredients	Reference	1% MFC	2% MFC	3% MFC
PLA1 (binder)	60	60	60	60
PLA2 (binder)	189	189	189	189
Isopropanol (solvent)	9	9	9	9
Wax emulsion	13.5	13.5	13.5	13.5
Defoamer	0.6	0.6	0.6	0.6
MFC (10% paste)	0	3	6	9
Water	27.9	24.9	21.9	18.9
Total, g	300	300	300	300

IV. Formulation 3 based on polylactic acid (PLA).

room temperature, and the final dry film thickness was around 180 μm.

RESULTS AND DISCUSSION

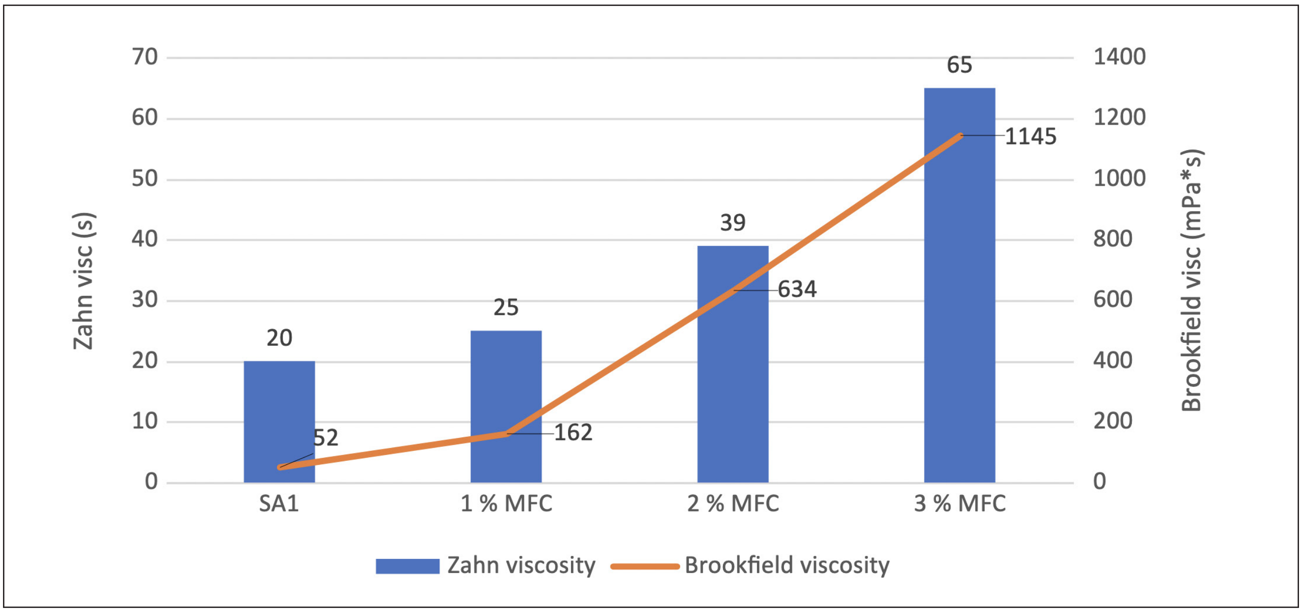
Impact of MFC on water retention of a water-based dispersion barrier coating

The effect of MFC on the water retention of barrier coating, as well as on the coating penetration into paper, was studied with a modified styrene acrylic copolymer. The MFC was tested at three different dosages ranging from 1% to 3% as delivered (0.1 to 0.3 of dry MFC). These dosages are 10 times lower than used in the study by Kumar et al. [2].

To meet target viscosity (Zahn cup #2 approximately 25–30 s), additional water (~10 wt%) was added into the formulation with an overhead stirrer (5 min, 300 rpm).

Water retention for each of the water-based barrier coat-

2. Analysis of the impact of MFC on water retention modified styrene acrylic copolymer (one reference; two with 1% MFC; three with 2% MFC; and four with 3% MFC).



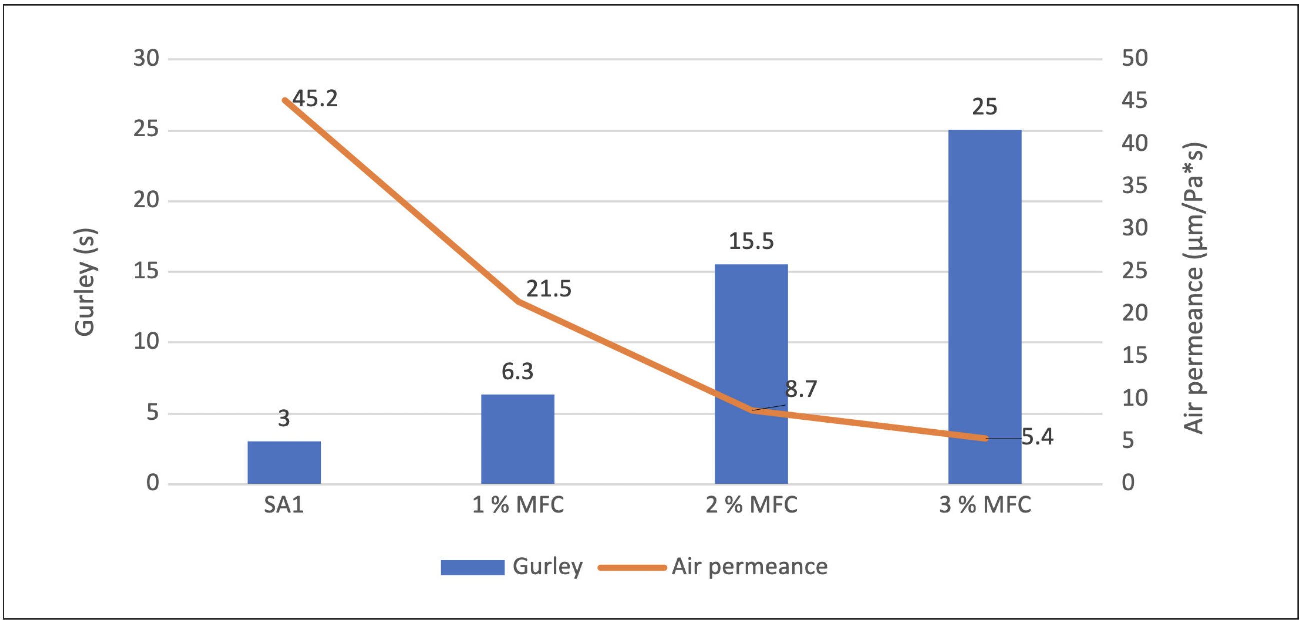
3. Zahn and Brookfield viscosity of Formulation 1 with different MFC dosages.

ing dispersions was analyzed by applying a droplet of each formulation onto a porous paper substrate.

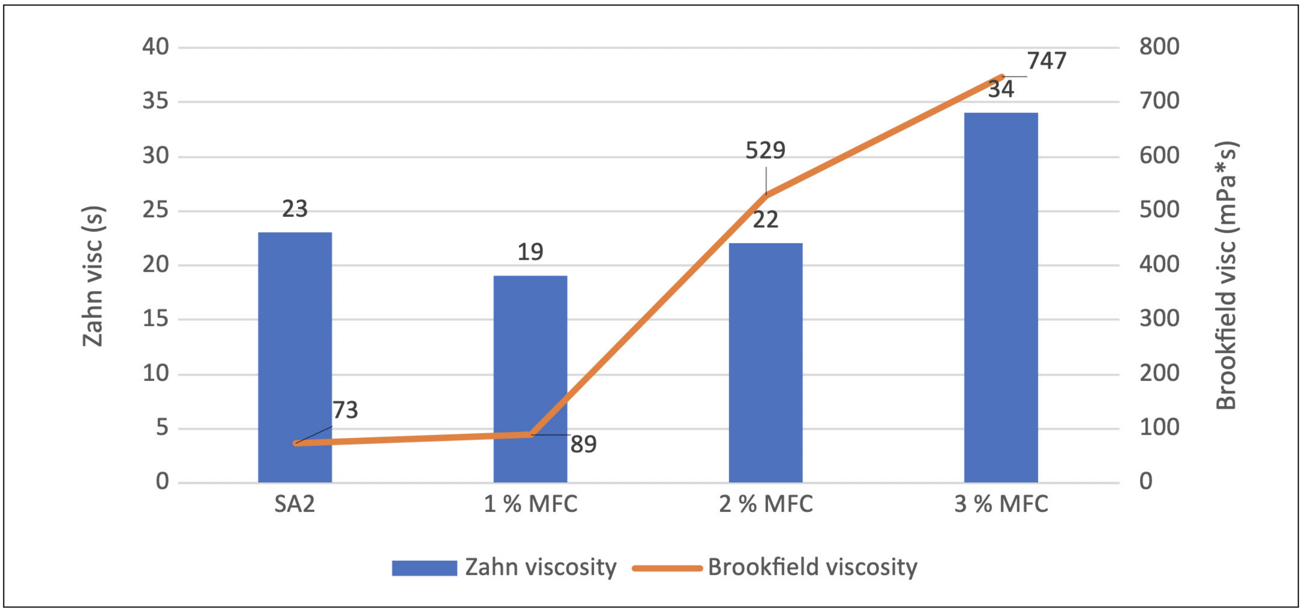
As seen from **Fig. 2**, by adjusting the amount of MFC in the dispersions using the acrylic based binder, the water holding capacity and coating absorption time can be controlled. Without MFC, the coating penetrates the paper in less than 30 s (upper left image). With only 1% MFC, the penetration of the coating is reduced. The change in water holding can be seen visually. With 3% MFC, there is essentially no leakage of water from the coating.

Impact of MFC on rheological and barrier properties of water-based dispersion barrier coatings

The effects of MFC on the rheological and barrier properties were studied in three different formulations based on a modified styrene acrylic copolymer dispersion, a styrene acrylic copolymer dispersion, and a polylactic acid dispersion (Tables II–IV). Reference formulations (1 and 3) did not contain any rheology modifier, and Formulation 2 contained a Newtonian thickener. The MFC was tested in all formulations at three different dosages (0.1 to 0.3 of dry MFC).



4. Air permeability results for Formulation 1.

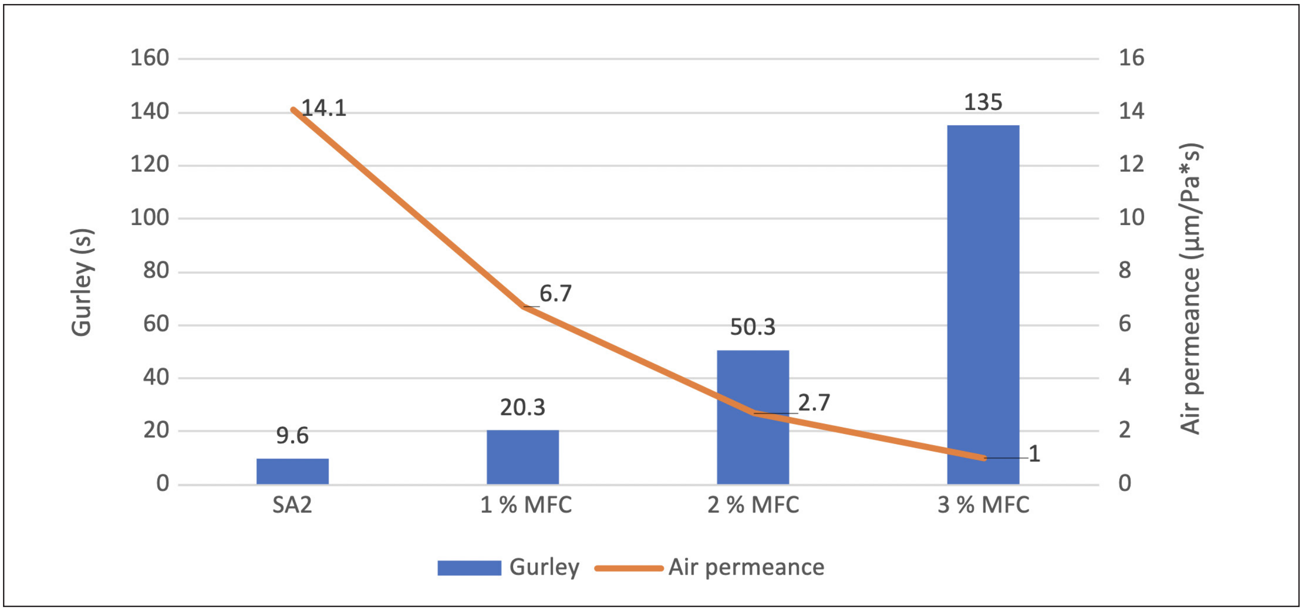


5. Zahn and Brookfield viscosity of the Formulation 2 with different MFC dosages.

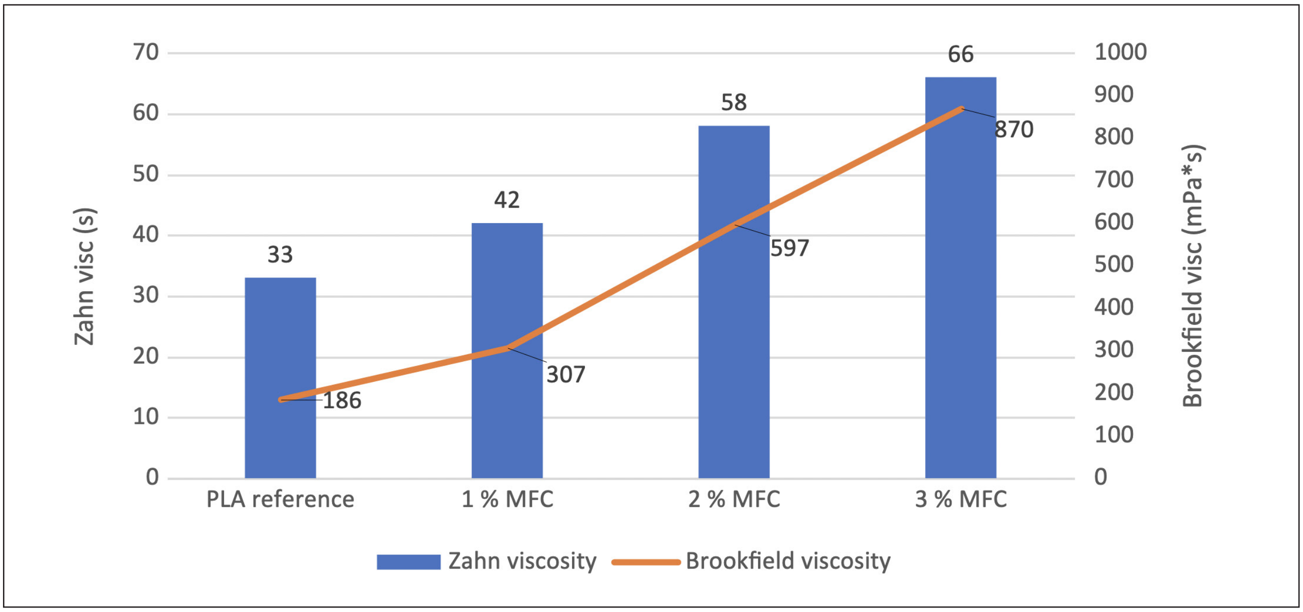
The impact of MFC on Zahn cup and Brookfield viscosity of Formulation 1 can be seen in **Fig. 3**. As the amount of MFC increases, viscosities increase accordingly. The high shear viscosity (5000 s^{-1}) of the reference formulation was 40 mPas, whereas for the formulations containing MFC, it varied between 40 and 50 mPas, depending on the dosage of MFC. The reason for the higher Zahn cup and Brookfield results at dosages higher than 1% MFC is because the formulation shows clear yield stress. This means that when at rest, these formulations are considered solids. However, after applying shear at a level higher than the yield stress, the formulations rapidly become liquids.

The impact of MFC on air permeability of the coated papers can be seen in **Fig. 4**. The reference Gurley value is 3 s. With increasing amounts of MFC, Gurley value increases up to 25 s. This indicates reduction in air permeability. The change effected is believed to be due to the reduced coating penetration into the paper, meaning that the coating is applied to the surface in a more uniform layer. The increased water holding of the coating will lead to improved film formation. It can also be expected that other barrier properties will also improve, e.g., oil, grease, and water.

The second formulation is based on two different styrene acrylic copolymer binders (Table III). The main difference from Formulation 1 is that reference Formulation 2 contains an acrylic thickener with a Newtonian rheology profile.



6. Air permeability results for Formulation 2.



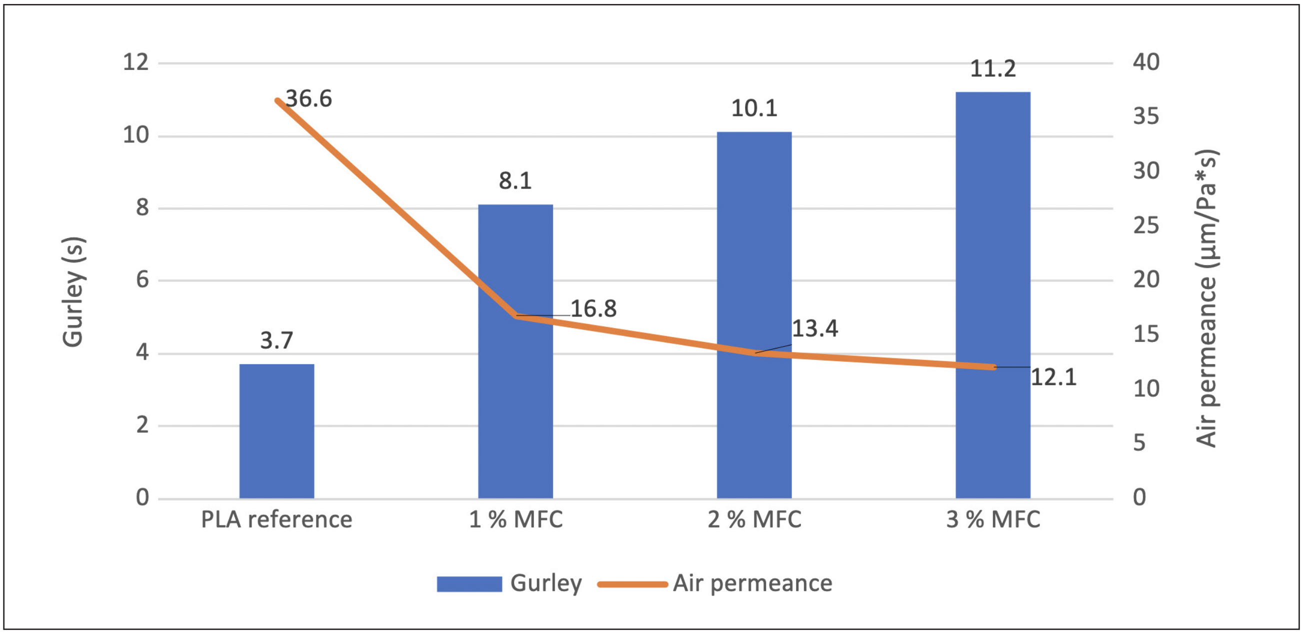
7. Zahn and Brookfield viscosity of Formulation 3 with different MFC dosages.

The difference between a Newtonian thickener and extremely shear thinning MFC can be seen in **Fig. 5**. At dosage levels of 1% MFC, Zahn cup values changed from 23 s (reference) to 19 s (MFC). At the same time, Brookfield viscosity changed from 73 mPas to 89 mPas, respectively. At dosage levels of 2% MFC, the Zahn cup value is comparable to the reference (22 s vs. 23 s), whereas the Brookfield value increases to 529 mPas. High shear viscosity (5000 s-1) of the reference formulation, as well as for the MFC formulations, was 30 mPas. This is a good example of the importance of analyzing a wide shear rate range when testing novel rheology modifiers with non-Newtonian properties.

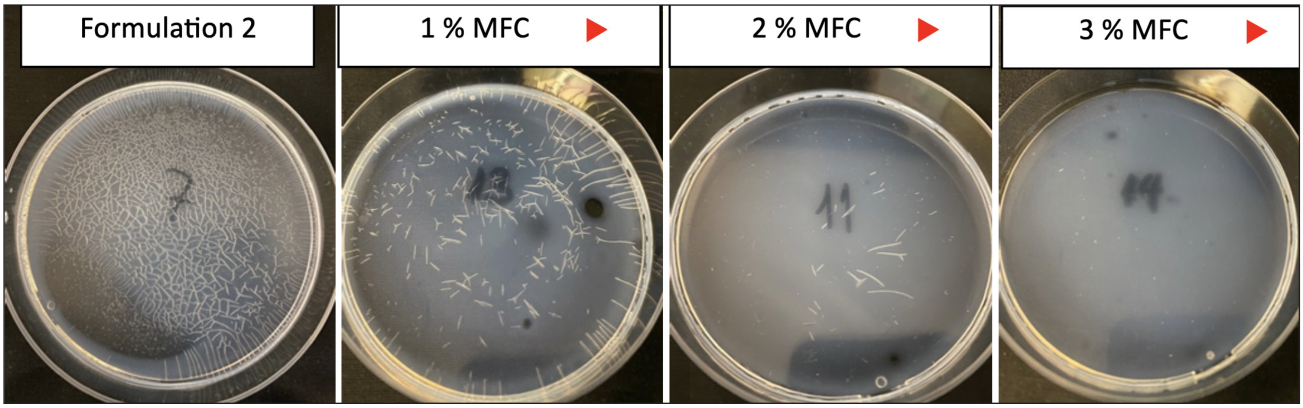
The impact of MFC on Gurley values is comparable to the results from Formulation 1. With 1% dosage of MFC, Gurley value doubled, as shown in **Fig. 6**. Figure 6 also shows that Gurley further increased five times at 2% and was 10 times at 3% of MFC.

The third formulation was based on two different water-based PLA binders (Table IV). Its accompanying reference did not contain any thickener.

As with Formulation 1, increasing the amount of MFC led to higher Zahn cup and Brookfield values, as **Fig. 7** shows. Zahn cup value increases about 30% with 1% MFC compared to the reference. A continued increase is observed at higher dosages due to yield stress.



8. Air permeability results for Formulation 3, with PLA as reference.



9. Impact of MFC on the cracking of SA copolymer-based barrier coating

Compared to the previous two formulations, the impact of MFC on Gurley results was lower, as shown in **Fig. 8**. One of the reasons might be that the morphology of the binder particle is different compared to the other ones tested in this study. If so, then this formulation would require a thicker coating than what was tested.

Impact of MFC on film formation and cracking of a water-based dispersion barrier coating

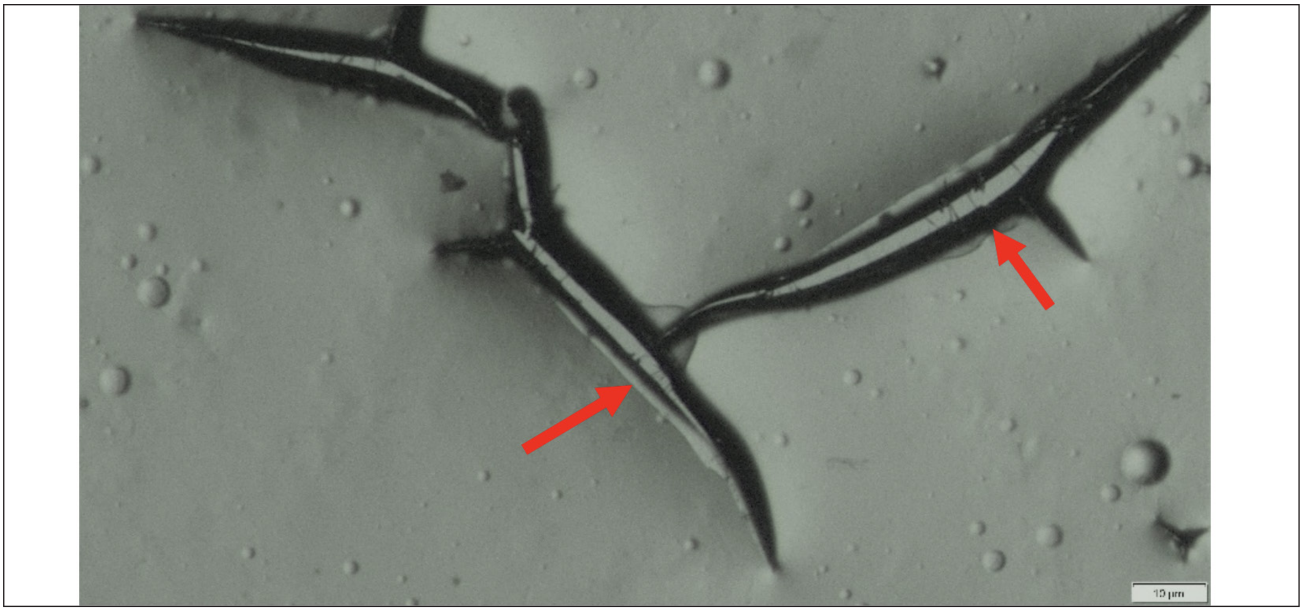
We have previously shown how MFC prevents mud cracking of an acrylic binder [3]. Improved mud crack resistance is a consequence of the effect of MFC on drying of the film. During drying, MFC prevents the formation of a "skin" on top of the film, allowing the solvents to freely evaporate. The MFC also increases the open time of the formulation and reduces film tackiness.

From **Fig. 9**, it is seen that the reference formulation suffered from pronounced cracking. At 1% MFC, the cracking reduced significantly. At 3% MFC, there was no visible cracking. It is also noticeable that MFC does not impart visual haziness to the films.

The prevention of cracking is believed to be due to mechanical reinforcement by MFC fibrils within the coating. The strong fibrils hydrogen bond with the binder, leading to a strong but flexible network. Tensile strength of single cellulose fibrils typically ranges between 1.6–3 GPa [4]. The single microfibrils in MFC consist of both crystalline and amorphous regions. Under stress, the amorphous regions offer flexibility due to their lower modulus [4].

Some fibrils can be microscopically seen existing within the cracks (**Fig. 10**). However, it has to be kept in mind that due to the size distribution of the fibril bundles, a major fraction of the fibrils is not visible under visible light.

As seen, MFC can prevent the cracking of coatings in two different ways: (1) through prevention of a skin forming during drying of the film, reducing mud cracking; and (2) by



10. Strengthening effect of MFC fibrils seen under microscope (2% MFC).

mechanically reinforcing the film and preventing the formation of cracks caused by stresses experienced during drying.

CONCLUSION

The use of MFC has been shown to impact the rheological properties of water-based barrier coatings, as well as their film properties. The addition of MFC increased water retention of the tested coatings, allowing for better control over coating penetration into fiber-based products. The yield stress, which can be observed at 2% MFC dosage (0.2% dry MFC) and higher, combined with extreme shear thinning, is beneficial for spray coating of molded fiber products. The yield stress prevents gravitational sagging of the coating on the sides of the molded fiber object, resulting in a uniform coating layer along the object. The unique shear thinning profile of MFC requires some extra attention from formulators. Reliance exclusively on Zahn cup and Brookfield viscosities does not provide a realistic picture about the runnability of a coating. We have also shown that addition of MFC into water-based barrier coatings prevents mud cracking and crack defects of the film during drying.

Due to these benefits, the overall barrier performance of coatings is significantly improved. We see MFC as a key ingredient in water-based barrier coating formulations based on these demonstrated benefits. **TJ**

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ABOUT THE AUTHORS

We performed this study because we wanted to see how microfibrillated cellulose (MFC) can solve issues related to water-based barrier coatings, as well as to see if MFC will improve barrier performance. The most difficult aspect of this study was narrowing down the subject, as MFC is a multifunctional additive.

Through this study, we gained further understanding about the rheology of MFC. Surprisingly, we also discovered how MFC can prevent the cracking of high glass transition temperature (Tg) polymers.

With the information presented here, mills can consider MFC as a way to improve the performance of their existing barrier coatings and coating color, reduce the consumption of coatings, and use lower quality (more porous) paper for barrier purposes. Our next step is to further explore MFC's impact in various binders, as well as its benefits in molded fiber barrier coating.

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