Effect of microfibrillated cellulose on paper barrier properties for Poly(vinyl alcohol)-based coatings

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INTRODUCTION

The food industry predominantly relies on fossil-based plastic for packaging and food storage due to its excellent barrier properties and low cost. However, there has been a shift in focus towards using eco-friendly alternatives like cellulosic fiber-based packaging materials. These materials are abundantly available, biodegradable, renewable and recyclable.

Despite its advantages, cellulosic fiber-based packaging faces challenge, especially in moisture-sensitive applications, due to its poor barrier properties and poor water resistance. Therefore, to broaden its application in food industry, extensive research is needed to improve its barrier properties and water resistance.

The aim of the study was to develop the coating formulations to improve the barrier performance of the paper. These formulations were developed as a unique blend of materials including poly(vinyl alcohol) (PVA), microfibrillated cellulose (MFC) and clay, in different concentrations.

The obtained PVA/MFC/clay coating suspensions were analyzed for their rheological behaviour and the coated papers were characterized for their thermal, barrier and strength properties.

MATERIALS AND METHODS

Preparation of coating formulation, paper coatings and characterizations

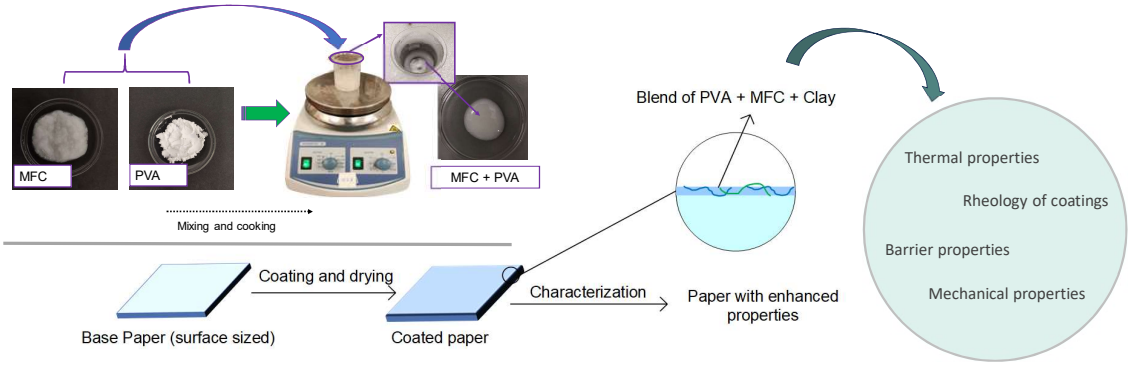


Table 1 Coating formulations to study the effect of PVA, MFC and clay on paper properties

Paper sample	Compositions (% wt.)
	PVA-MFC-Clay
Base Paper (BP)	-
PVA	100
PM5	95-5
PM10	90-10
PM15	85-15
PMC5	90-5-5
PMC10	85-5-10
PMC15	80-5-15

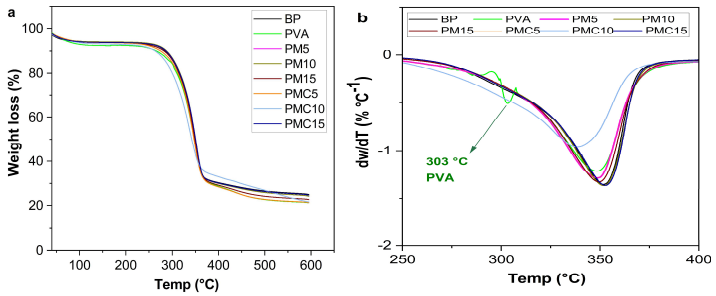
Coating formulations were prepared using specific concentrations of poly(vinyl alcohol) (PVA), microfibrillated cellulose (MFC) and clay as reported in Table 1.

First, PVA was dissolved in distilled water at 90ºC for 2 h, and used as a host material, followed by the addition of MFC and clay in the suspension. A coating layer was applied to the one side of the surface-sized base paper using a film applicator/casting knife aiming for a lower coating weight.

Coated papers were dried at the room temperature and characterized for their thermal, barrier (water, air and oxygen permeability) and strength properties.

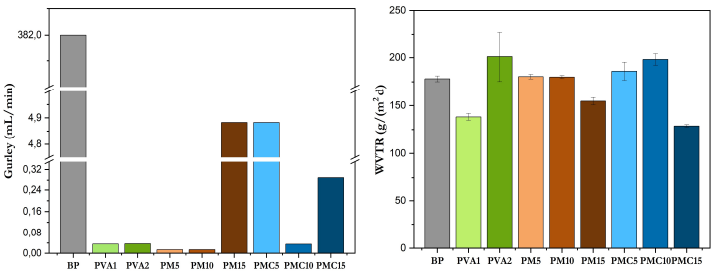
RESULTS

Thermal properties



In all the papers, water loss at low temperatures (50-100ºC) occurred due to their moisture content. Significant weight loss at 250-350ºC indicated the decomposition of starch and cellulose in these samples. In only PVA-coated paper, PVA degradation was noticeable at 303ºC. DTG curves suggested that in the composite coatings (PVA + MFC + Clay), the T_{max} (temperature at which maximum mass loss occurs) of PVA increased to around 350ºC. MFC and clay improved the thermal stability of PVA, except for PMC10, mainly due to lower pick up for this coatings. The highest T_{max} was observed for the PMC15 indicating the superior thermal stability.

Barrier properties



A thin layer of coating using PVA, MFC and clay (5-10 g/m^2) significantly reduced air permeability by closing of surface pores of coated paper. However, PM15 and PMC5 coatings showed a higher Gurley value, indicating some defects in the coating layer.

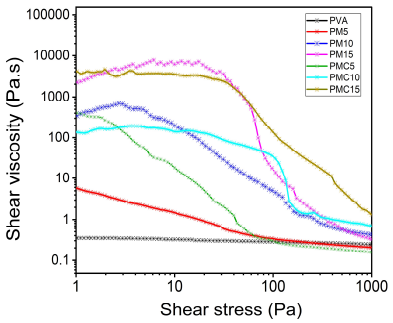
All the coated paper samples showed an oxygen transmission rate (OTR) above the detection limit of the equipment (OTR>61205 cm^3/ (m^2 d)) due to their low coating weight.

PVA coating resulted in a lower water vapor transmission rate (WVTR) compared to the base paper, suggesting that the thin PVA layer inhibited the penetration of water vapor through the coated surface. MFC (5 to 15%) reduced the WVTR to 154.5 g/ (m^2 d), which is 13.2% lower than the WVTR of BP.

Addition of 15% clay decreased the WVTR value to 128.8 g/ (m^2 d), showing a 16.6% and 6.8% improvement over BP and PVA coatings, respectively.

Rheology Results

Flow curves of shear viscosity of 20 wt% aqueous suspensions of the obtained for coating formulations as a function of shear stress.

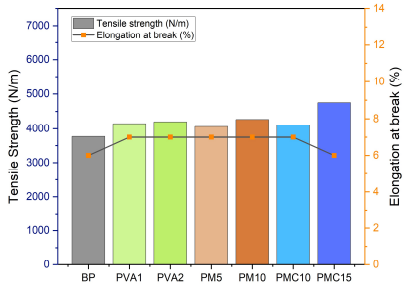


PVA solution displayed mainly Newtonian behavior across the range of shear stresses. At lower shear rates, the initial viscosity increased with increasing MFC content, likely due to the formation of a denser MFC fibrils network at higher concentrations.

Yield stress increased with increasing concentration of MFC. Further, viscosity decreased linearly as shear stress increased, attributing to the alignment of MFC domains in the shear direction after gel breakage.

Addition of clay to these formulations increased the initial viscosity at lower shear stress compared to PM5. This was due to more small particles interacting with fibers, boosting viscosity and also showed higher yield stress at increasing concentration of clay. It also improved the coating ability of formulations due to better rheological properties.

Strength properties



All coated papers showed improved tensile strength compared to BP.

PVA coated paper with higher grammage (PVA2) demonstrated slightly higher tensile strength than lower grammage (PVA1), attributed to PVA's the excellent film forming properties, coupled with its ability to reinforce the paper.

Highest increase of 26% in tensile strength was observed with the 15% clay, in combination with 5% of MFC in PVA-based coatings.

All coated papers showed almost equal elongation at break.

CONCLUSIONS

- All the coated papers exhibited excellent air resistance. In terms of WVTR, the addition of 15% clay (with 5% of MFC) to the PVA coatings increased it by 17% compared to the base paper.
- Composite coatings including PVA, MFC, and Clay demonstrate a notable improvement in PVA's thermal stability.
- The tensile strength improved with the addition of PVA, MFC, and clay, particularly with 15% clay when used in combination with MFC/PVA.
- The results show that the coating formulation with 15% clay 5% MFC and 80% PVA exhibits excellent properties in each aspect, thus making it the most optimized of the studied formulations to be used for the food packaging application.

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