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Improving water vapor barrier of cellulose based food packaging using double layer coatings and cellulose nanofibers

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ABSTRACT

Paper based packaging has the potential to replace many plastic-based systems if the required barrier properties can be obtained. Water borne barrier coatings have the potential to generate good barrier layers, but their performance is often less than expected. Recent work has shown improved performance of these coatings when applied on paper that has a cellulose nanofiber layer.

Here, papers with a cellulose nanofiber layer were coated with barrier coatings at different coat weights applied as single-layers, as double-layers, and single-layers pressed together in a hot press in order to generate a packaging system that has good barrier properties. The performance of double-layer samples resulted in moisture transmission rates that were 40–70 % of the value of the single-layer systems. Surprisingly, the hot-pressing of two dry layers showed no advantage compared to the single-layer system. A barrier pigment added to one formulation improved the performance further and followed the same trends. Three dimensional models of diffusion through layers that have defects help explain the results. The work shows a potential path to produce paper-based packaging that has both good oxygen and water vapor barrier properties.

1. Introduction

Society needs sustainable packaging that can be recycled and breaks down in the environment if discarded. Paper-based packaging satisfies these demands, but for some applications, paper does not have the needed barrier properties to keep some food products fresh. Paper can be extrusion coated with a polymer such as polyethylene (PE) to obtain these properties, but this makes the package harder to recycle and slow to break down in the environment. Water borne barrier coatings (WBBC) or water dispersed coatings are reported to be easier to recycle and to break down in the environment compared to PE coated paper (Brander & Ian, 2012; Mesic et al., 2015). However, these coatings do not give the same barrier properties as the extruded polymer films at the same film thickness. Multiple coating layers may improve the performance, but this is not well reported in the literature. The objective of this work is to understand the influence of applying WBBC in layers on paper that has a layer of cellulose nanofibers to give a paper-based food packaging system with good oxygen and water vapor barrier properties.

The concept of using multiple layers in the packaging is standard practice for several products, such as snack packaging with metalized layers (e.g., Snow, 1982). This packaging often uses polymers of different properties along with a metal layer to obtain the desired properties (Lagaron et al., 2004). Less is reported for the performance of paper-based systems with multiple layers of barrier coatings.

Several groups have described the excellent oxygen and grease resistance of cellulose nanomaterials (e.g., Aulin et al., 2010; Hult et al., 2010; Mousavi et al., 2018; Plackett et al., 2010, pp. 32254). The oxygen transmission rates (OTR) for pure cellulose nanofibers (CNF) without any additive were as low as 17 ml m^{-2} day^{-1}, obtained for films 30 μm thick (Syverud & Stenius, 2009). Kumar et al. (2014) show that a 25 μm film of CNF had an oxygen transmission rate of 1.4 ml m^{-2} day^{-1}. These values showed a better barrier performance to oxygen than many polymer such a PE. Wang et al. (2018) and Lavoine et al. (2012) reviewed the moisture and oxygen barrier properties of cellulose nanomaterials. Recent work has shown the great potential of CNF films for oil and grease barrier layers (Tayeb et al., 2020).

Abbreviations: PE, polyethylene; CNF, cellulose nanofibers; WBBC, water borne barrier coatings; CNC, cellulose nanocrystals; OTR, oxygen transmission rate; WVTR, water vapor transmission rate.

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CNF films are known to be water sensitive, losing oxygen barrier properties at high humidity. Layering CNF with a wax or other coating has been suggested (Aulin et al., 2010). Others have reported coating an extruded polylactic acid layer on CNF to keep the layer from the influence of the moisture (Koppolu et al., 2019): excellent results were obtained, but the CNF layer was still sensitive to moisture because only one side was coated. Another multilayer system of CNF and cellulose nanocrystal (CNC) was made to improve gas and grease resistance coating for packaging (Tyagi et al., 2019), but CNC layers are also sensitive to moisture. Vähä-Nissi et al., 2017, describe the use of CNF and biobased polyethylene to obtain pouches for dry food; these systems have good barrier properties and show excellent promise for sustainable packaging systems. Ferrer et al. (2017) review other potential methods, including layers, to utilize cellulose nanomaterials in packaging.

Therefore, it is logical to explore the potential to use of WBBC to obtain a good moisture resistance of the package and protect the CNF layer from moisture. Water borne dispersion coatings are known to impart good water vapor barrier properties (Rissa et al., 2002; Vähä-Nissi & Savolainen, 1999; Zhu et al., 2019). These coatings often depend on a latex that forms a film during drying. The value of the water vapor transmission rate (WVTR) of waterborne barrier coatings should be similar to a polyethylene film if the film is pinhole-free, but this value decreases significantly in the presence of pinholes (Brander & Thorn, 2012). In practice, WBBC generate WVTR values in the range of 20–100 g/(m^2 day). This value is not low enough to satisfy the demand of some food products. Recently, Al-Gharrawi et al. (2021) show that a CNF layer improves the performance of WBBC by over a factor of two, but further improvement is desired.

The use of multiple layers of WBBC to obtain improved results has limited reported studies. Kugge and Johnson (2008) report that when styrene-butadiene (SB) latex coatings were applied in two layers instead of one, the WVTR was half at the same total coat weight. For example, at 12 g/m^2 total coating weight, 50 % R.H., and 23 °C, the WVTR of two layers was 20 g/(m^2 day) while for single coating, it was 40 g/(m^2 day). Another group showed that coatings applied in thin layers gave better results than a single thick layer in terms of water barrier performance and fewer cracks (Mesic et al., 2015). Multilayer samples coated using the flexographic press showed better barrier performance and less cracks on the coated surfaces than samples coated using a conventional coating process for the same total weight of 6 g/m^2. The value of WVTR was 330 g/(m^2 day) by using a traditional coating of a rod coater, while this value dropped to 60 g/(m^2 day) at the same basis weight (Mesic et al., 2015). The rapid drying of thin layers results in more homogenous layers and reduced filter cake and skinning compared to a single layer.

Another group reported better grease barrier property when using a two-layer coating compared to a single layer (Larsson & Emilsson, 2019). When comparing two thinner coating layers with one thicker layer, the grease barrier properties increased significantly for two layers. The results indicate that the second layer was able to cover any lacking or defects in the first layer.

In the current work, the influence of applying two thin layers of water-based barrier coatings on paper with a CNF layer is compared to a single thick layer and for the case where two thin layers are applied on paper, dried, and hot-pressed together. A model is developed to characterize the influence of defects on the water vapor transmission rate.

2. Materials and methods

Coatings were applied on paper that was made at the Process Development Center at the University of Maine. The paper was 80 g/m^2 basis weight and had a top layer of CNF at 4 g/m^2. The base sheet was an 80:20 hardwood: softwood blend. The paper was manufactured on a pilot paper machine, consisting of a Fourdrinier forming section, press section, and drying section. The CNF used in this study was produced from bleached softwood Kraft pulp by two-stage disk refining at 3 % consistency, to a fines content of 90 % as measured on a fiber size analyzer (MorFI, Techpap Inc.). The CNF had a large number of fibrils that are on the order of 50 nm in diameter and 5 μm in length, but there was a large size distribution of material.

The surface application of CNF on paper was achieved via an in-house built secondary headbox. The CNF suspension at 0.5 % by weight solids drops onto the wet formed paper that has drained to around 20 % by weight solids. This combination goes onto the normal press and drying sections of the pilot paper machine. The CNF coated paper had a low air permeability compared to the base paper without the CNF layer: the permeability coefficients were on the order of 10^{-14} and 10^{-16} m^2 for the uncoated and coated paper, respectively. The rate of water uptake for the coated paper was over ten times slower than the uncoated paper. The CNF layer creates a closed dense layer on the paper. More details about the method to produce the paper are given by Johnson et al. (2019).

Three different WBBCs based on different polymers were used in this work. They were named polymer A, B, and C. Polymer A was supplied by Mantrose-Haeuser branded VerdeCoat WB-10-base. This polymer is FDA approved for food contact, meets ASTM 6400 and EN13432 standards for composability, and is repulpable, microwaveable, and heat sealable. It has a solid content of 36.5 % and a density of 1.03 g/cm^3. Polymer B was supplied by SYNTHOMER titled X12-185; it has a solid content of 52 % and is styrene-butadiene latex-based chemistry with a particle size of 180–220 nm, a glass transition temperature of 5 °C. It serves as water, grease, and moisture vapor barrier coating. Polymer C was supplied by Michelman (Michem Prime 4983R). The chemical nature of the preparation is ethylene- acrylic acid dispersion. It is a translucent liquid that has a solid content of 24.8 %.

Kaolin was the barrier pigment used in the coating formulation with polymer B in one case in this study. It was supplied by IMERYS Minerals Ltd (Barrisurf HX); it has a 64 % particle size less than 2 μm and a shape factor, linked to the aspect ratio, of over 90.

The coating process was performed using a laboratory bench-size automatic rod coater (model 21001, BYC Gardner USA) with standard Mayer Rods. Different rod size and pressure were used to end up with varying amounts of the coating. The speed was fixed at a moderate level. Coatings were dried at 105 °C for 5 min except for polymer A, that was dried at 125 °C as suggested by the manufacturer. All samples were coated on both sides, where the first side was dried before the other side was coated. The coat weight is determined by measuring the weight change after coating for a known area. Two of the samples with a single layer on each side were hot-pressed to each other using a hot press (Carver, model: C 41000-003) at 125 °C. Two aluminum (Al) foil sheets were sandwiched about the laminated films to keep the sample from sticking to the heated blocks. Then the Al films were peeled to end up with the required system.

The final double-coated films were coated twice on each side, drying the first coating before the second one was applied. The three polymers used in this work behaved differently during the coating operations. One issue was that on the second layer, polymers A and C would bead up or form drops that do not level out on top of the first layer. Polymer B did not have this issue. This beading up could be avoided for polymer A when the coating of the second layer was started directly after the drying of the first layer. This solution did not help for polymer C. Therefore, a brush was used to smooth out the drops in the second layer; after drying, brush marks were not visible for these samples.

The water vapor barrier property of the coating was measured according to ASTM 96/E96M-16. In a controlled temperature and humidity room (23 °C, 50 % relative humidity), jars were filled up with 30 ml of water and closed by the sample of interest as the lid. The sample was held against the jar with a silicone gasket and a metal screw top rim. Samples were conditioned at least one day to avoid any initial changes due to the humidity conditions. The weight change over at least 24 h was used to calculate the WVTR.

3. Diffusion models

3.1. Steady state Fickian diffusion

Under the assumption that the layers have no defects, Fick's law for steady-state diffusion through two layers is used to predict WVTR. The diffusion of water vapor through these films is likely through small regions that have low polymer density but it likely is not through a Knudsen mechanism. The CNF layer offers little resistance to diffusion of water vapor and is therefore neglected in the models (Wang et al., 2018). The standard equation to predict the total flux of water vapor or the WVTR for two layers in series can be written as:

$$WVTR = \left(\frac{M_w(C_0 - C_1)}{\frac{H_p}{D_p} + \frac{H_c}{D_c}} \right) \quad (1)$$

M_w is the molecular weight of water, C_o and C_1 are the concentration of water vapor inside and outside of the jar, respectively, H_p and H_C are the thicknesses of the paper and coating layer, respectively, and D_P and D_c are the effective diffusion coefficients of the paper and coating, respectively. The partition coefficient of water and polymer is assumed to be constant and becomes included with the diffusion coefficient. WVTR is usually measured by sealing the sample on the mouth of a jar with water inside in a room at 23 °C and 50 % relative humidity. Using the vapor pressure of water at these conditions, the concentration of water vapor inside the jar is 1.1 mol/m^3. The 50 % relative humidity would have a concentration of half of this value outside of the jar. The WVTR of the uncoated paper with the CNF layer is around 400 g/(m^2 day). The thickness of the paper was 0.098 mm. This thickness gives a diffusion coefficient for the paper of 4.6×10^{-8} m^2/s in Eq. (1). The coat weight of the barrier coating is used to calculate the coating thickness H_C. A least-squares method is used to find the diffusion coefficient of the coating that fits the experimental data.

3.2. Model of diffusion through parallel pores

When a barrier layer has a defect, such as a small pore, the diffusion through that defect can be estimated by the diffusion rate of water in the air. If the defects are idealized as cylindrical pores in the barrier film, then the diffusion rate through a film is given by the mass transport in parallel tubes. The pore size does not matter but the area fraction of defects becomes the controlling parameter. If the diffusion coefficient in the polymer is low, then all of the diffusion is through the pores. The water vapor transmission rate, assuming that transport through the paper layer is rapid, becomes

$$WVTR = \mathrm{Mw}(C_0 - C_1) \quad \left(\frac{A_d \quad D_{aw}}{H_c} + \frac{(1 - A_d)Dc}{H_c} \right) \quad (2)$$

where A_d is the fraction of the surface area that are defects, D_{aw} is the diffusion coefficient of water in the air that has a value of around 2.5×10^{-5} m^2/s at room temperature (Lee & Wilke, 1954), H_c is the thickness of the coating layer. If the diffusion coefficient of water in the polymer, including the partition coefficient, is on the order of 5×10^{-10} m^2/s, then only a small number of defects can dramatically change the barrier performance of the film. For example, for a barrier film with a thickness of 40 μm, the flux rate increases from 11 to 550 g/m^2 day if 0.1 % of the surface area has pores or some type of defect that runs through the barrier layer. This result shows how important it is to eliminate all defects from a barrier layer.

3.3. 3D model of diffusion in layers with defects

A finite element based computer program (COMSOL Multiphysics) is used to predict the diffusion rate through two barrier films that have defects. In one case, one layer has a defect and the second layer has no defects. Another case should represent the hot-press or lamination situation where each layer has defects, but they likely do not match up nor align at the interface. The basic geometry of interest and the boundary conditions are shown in Fig. 1. The meshes and typical results are illustrated in Fig. 2 for closely and widely spaced defects of radius 200 nm. The calculation is steady-state diffusion in a solid in three dimensions through two 20 μm-thick layers representing a total barrier layer thickness of 40 μm. The diffusion coefficient inside the defect is that of water-air given above. The diffusion coefficient of the polymer region is assumed to be 5×10^{-10} m^2/s. The water vapor concentration on each side of the region is the boundary condition with values given for saturated air and 50 % relative humidity at room temperature. No flux conditions are set on the edges of the region. If the defects happen to align, then Eq. (2) applies, and the flux rate would be for a situation where 0.1 % of the area has defects. Fig. 2 shows a case where the defects are close to connecting and another case where they are widely separated. The question of interest is how effective the lamination of two layers can be given that each layer may have defects but regions that do not have a defect cap the defect of the other layer. For the double coating simulations, the first layer has no defects: this assumes that the wet coating can cover or heal these defects, and the second layer has a specific size defect.

4. Results and discussion

For simplification, samples that were coated with a single layer on each side are denoted (DS), samples double coated or double layers on each side (DL), and samples with double films hot-pressed together (DF). Fig. 3 depicts these situations.

Figs. 4–6 summarized the results of the three types of samples for cases that involve the three polymers without a barrier pigment. As expected, the WVTR decreases as the coat weight of the barrier layer increases. Samples that were double coated on each side improved the moisture barrier of the films at similar total coat weights by 40–70 %. This result may come from the potential of the second layer to seal up or heal any defects that are in the first layer. Surprisingly, when films are hot-pressed together, the results are not improved and follow the results of the single layer on each side when the total coat weight is considered. This lamination was expected to also heal defects by closing defects with a solid film.

The lines in the figures represent the best fit to Eq. (1), where the diffusion coefficient of the barrier layer is adjusted to minimize the sum of the square of the residual error. In some cases, the data follows this trend well through the entire coat weight range, but the data deviate in other cases. For example, for polymer B, the DL results fall high of the best fit for high coat weights and are low for low coat weights. This result may come from the potential for defects to occur at high coat weights due to shrinkage of the layers during drying. There is much scatter in the data even though care was taken to be repetitive in the coating and WVTR results. This seems to come from random events or defects in the barrier layers as they are cut and mounted into the jars or from the coating event.

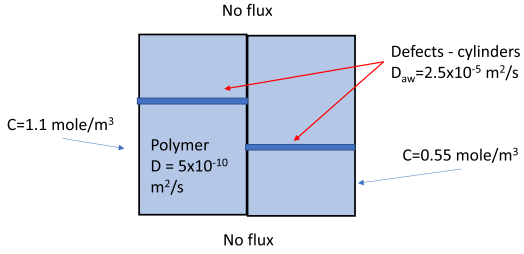

Fig. 1. Representation of the basic geometry and boundary conditions for the finite element model.

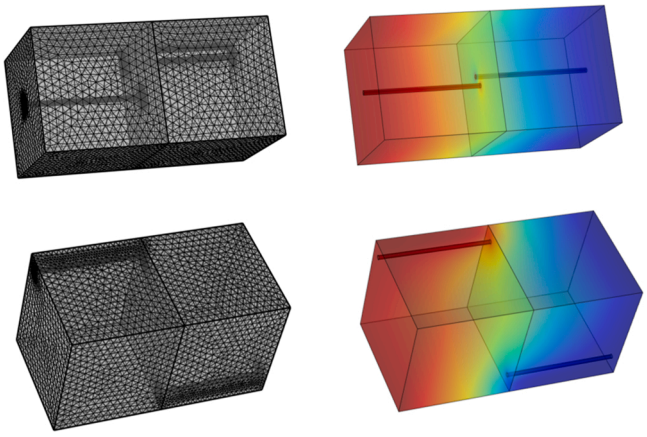

Fig. 2. Mesh (left) and concentration profiles (right) for the case of two barrier layers that have pore defects of 200 nm in radius, where the defects are close to connecting (top) and where they are widely separated (bottom). Red and blue represent high and low water concentrations, respectively. The model is also run with a defect only in one layer.

For polymer C, the hot-pressed structure had higher WVTR results compared to the samples with a single layer on each side. This polymer, when dried, tends to be brittle. Possibly the lamination of the samples can lead to an increase in the propensity for fine defects due to this brittle behavior. The net result is, after hot-pressing, the mechanical step generates more defects. This brittle behavior also explains the scatter seen in the data.

The behavior of the films with a barrier pigment added at 10 % by weight is shown in Fig. 7. Again, the double coating of each side had improved results compared to a single layer on each side and the hot-pressing of two films. The WVTR is only around 30 % of the value of a single layer when double coating on each side for the same total coat weight. As with the results above, the hot-pressing of two layers gives results similar to the single coating layer on each side. These results also indicate that the wet coating layer applied to a dry layer has the potential to seal up or heal defects in the first layer to improve the barrier performance, but when two layers are pressed together, little improvement is seen compared to a single thick layer. The barrier pigment at this level gives a nice improvement of the results: the WVTR of double coated systems is close to the target of some food packaging systems and is lower than most values reported in the literature for WVTR for these types of coatings.

4.1. Comparison of models with results

If the diffusion coefficient of water vapor in the polymer was 5×10^{-10} m^2/s, with no defects, then the WVTR through the film of thickness 40 μm would be 10.7 g/(m^2 day) following Eq. (2). This value is close to the results obtained with a double layer situation in Fig. 3. The kaolin pigment reduces this value to around 4 g/(m^2 day). If the fraction of the area with defects was 1×10^{-4} (0.01 %), then WVTR increases to

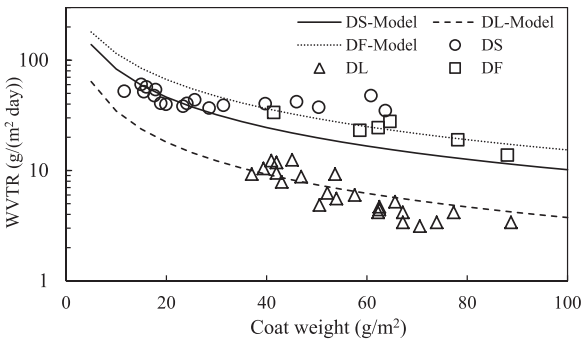

Fig. 4. WVTR of polymer A. Double side single layer coating only (DS), Lamination two single layer films (DF), Double layer on each side (DL). Lines are fitted to Eq. (1).

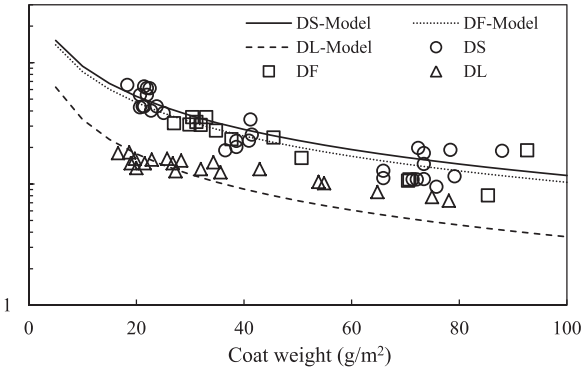

Fig. 5. WVTR of polymer B. Double side single layer coating (DS), Lamination of two single layer films (DF), Double layer on each side (DL). Lines are fitted to Eq. (1).

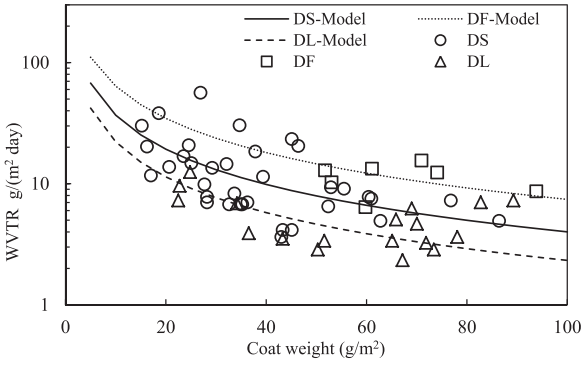

Fig. 6. WVTR of polymer C. Double side single layer coating (DS), Lamination two films (DF), Double layer coating on each side (DL). Lines are fitted to Eq. (1).

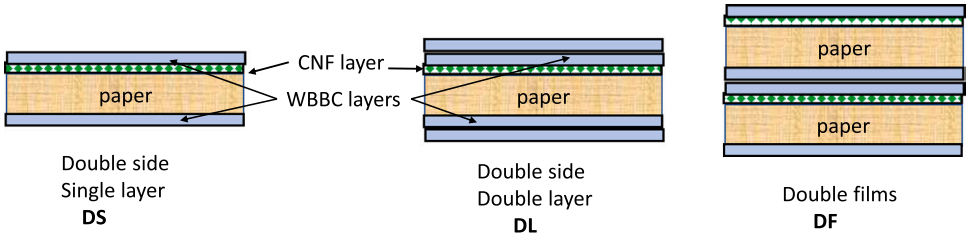

Fig. 3. The three methods to produced layered structures. DS is single layer on each side. DL is two layers, wet coated on dry, on each side, and DF is the hot-pressing of two single layer samples.

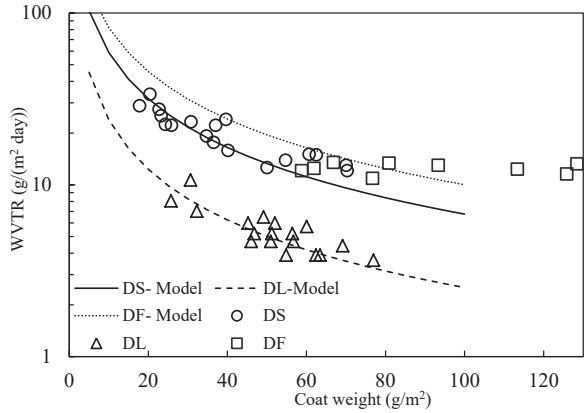

Fig. 7. WVTR of polymer B with 10 % Kaolin. Double side single layer coating (DS), Lamination two layers (DF), Double layer on each side (DL). Lines are fitted to Eq. (1).

values around 64 g/(m^2 day) using Eq. (2), a value near the result of the single-layer coatings and the laminated films.

When the WVTR is calculated with the finite element code, the results of single defects that line up and run through both layers of various sizes agree with Eq. (2). If the defects line up, there is a clear path for water to diffuse in air-filled pore through the sample. When the defects do not line up, as may occur when laminating two layers, the water vapor must diffuse some short distance at the interface of the two layers in the polymer to reach the other defect. Fig. 8 shows the results for different pore sizes for the different separation of pores. Clearly, if the defects do not connect, the flux rate drops, but for further separation of the defects, the flux tends to drop a limited amount to values that are around three times that of no defects (34 g/(m^2 day)). If there is a defect in just one layer, the WVTR is predicted to be around 50 % more than the defect free case depending on the pore size 16.5 g/(m^2 day).

The model results help explain the results in Figs. 4–7. If it is assumed that for double layer coating, that the defects in the first layer down are filled or repaired by the second layer, then defects are in the range of 100 nm in the second layer, will result in WVTR that are around 50 % higher than the no defect case. For the single layer coatings, it is assumed that defects span the whole layer and are on the order of magnitude of an area fraction of 0.01 %. This gives flux rates that should be a factor of six times the no defect case or in the range of 70 g/(m^2 day). For the hot-pressing of two layers, we assume that defects do not match and give results as represented in Fig. 6 for large separation distances, in the range of three times the no defect result. Also, for polymer A in Fig. 3 at 40 g/m^2, the double coating results give WVTR on the order of 12 g/(m^2 day). The single coating and lamination give WVTR on the order of 40 g/(m^2 day). This result is near the change expected from going to a defect in one layer, to defects in both that do not line up. For the double coating predictions, the defect in one layer is assumed to be filled, and the second layer has a single defect of a 30 nm radius. For the predicted laminated system, each layer has a 30 nm defect, but they are separated by 3 μm distance. The model predicts the general trends and magnitudes of the experimental results. For the laminated systems, the model predicts a smaller value of WVTR than what was measured, even if the defects were not aligned. This result may come from the potential of more defects generated in the experiments in the lamination process than what was assumed in the model. The double coated sample has the lowest WVTR because there is a layer that is defect free and the difference of these results compared to the single layer case is in the range seen experimentally.

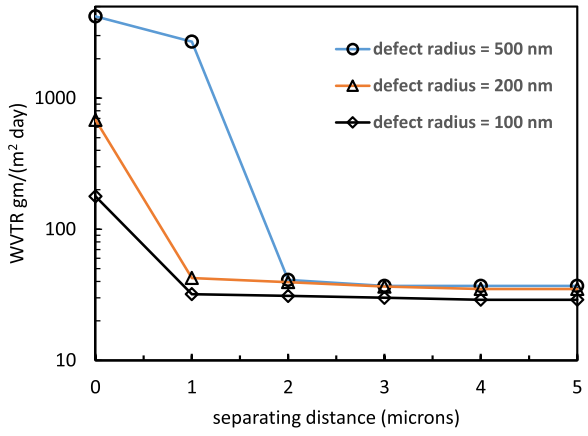

Fig. 8. The influence of the spacing of the defects on the WVTR predicted by the finite element model.

The mechanism that seems to explain these results is depicted in Fig. 9. If the second layer is a dispersion that can flow or penetrate into pores, then when the second layer is applied, it fills in the pores or defects of the first layer. Capillary flow into the defect is expected to drag more polymer into that region. When this flow occurs, after drying, the defect would be filled with polymer or the barrier pigment. If the second layer is a dried film that is laminated, the existing pores do get caped, but there likely are defects in the second layer that still can transport water vapor. Diffusion in the polymer still occurs, but now the distance for diffusion is small because the defects provide a path for water vapor to reach the interface in one layer and remove from the interface is the second layer.

5. Conclusions

Barrier coatings applied as two aqueous layers improved the barrier performance of the systems by 40–70 % compared to single-layer coatings or two thin coatings hot-pressed together. The ability of the second layer to heal pinholes or defects by capillary driven flow seems to explain the results. Hot-pressed dry films may cap defects, but this seems to have limited value in reducing the WVTR of these systems. Defects likely exist in both layers and provide a diffusional path to the interface. A barrier pigment added to the formulation improves the results further, but this coating still follows the same trend as the coatings without pigments. A model was developed based on diffusion in defects and polymers that agree with the experimental trends. The results indicate a good potential to produce a cellulose-based packaging system with good water barrier properties.

CRediT authorship contribution statement

All authors were involved in this project in many areas. **Mohammed Al-Gharrawi:** Investigation, Data curation, Validation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization.

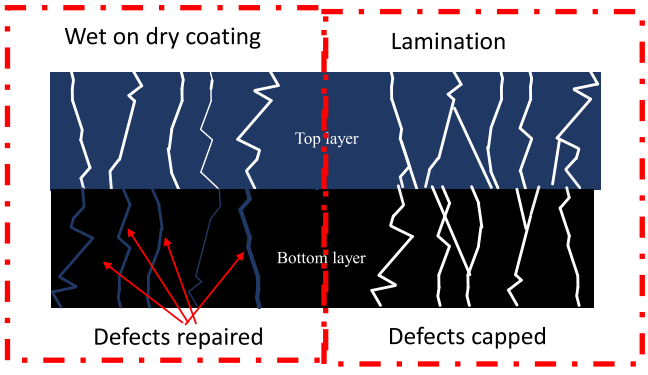

Fig. 9. Schematic of (left) the coating of the two layers wet on dry and (right) lamination of the two films that both have defects.

Jinwu Wang: Conceptualization, Methodology, Formal analysis, Writing – review & editing. **Doug Bousfield:** Conceptualization, Software, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Interests

None.

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