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USE OF RENEWABLE CELLULOSE NANOFIBRILS AND WATER-BARRIER
LAYERS IN THE PRODUCTION OF SNACK PACKAGING

by

Thoalfakar Saheb Alsaady

A Thesis Submitted in Partial Fulfillment
of the Requirements for a Degree with Honors
(Chemical Engineering)

The Honors College

University of Maine

May 2018

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ABSTRACT

From potato chips to chocolate bars, snack packaging requires materials that have excellent water vapor and oxygen barrier properties, good mechanical behavior, and good grease resistance. The most common material used in snack packaging is polypropylene (PP) layer that has been aluminum coated or metalized on one surface. The problem is not only that non-renewable petroleum is used to produce the plastic, but the production of aluminum requires large energy inputs and generates toxic wastes. In addition, this current packaging does not break down in the environment and cannot be recycled. This research is looking at using layers of cellulose nanofibrils (CNF) and water-barrier layers on paper as a replacement for current packaging materials. This replacement will make the packing recyclable and renewable without compromising the characteristic of water, oxygen and grease resistance. The estimated production costs are similar to the current materials.

This work is part of a three-year project. The part presented in this thesis is part of the first year that is mostly the bench scale work that includes: coating CNF layers on papers, coating water-barrier layers, and exploring different paper types. In addition, all samples were characterized in terms of coat weight, air permeability, water-vapor permeability, and water vapor transmission rate using standard methods.

A number of paper types were used, including one that had CNF incorporated into the paper structure and one that had a CNF layer applied on the wet end of a paper machine. Three different water based barrier coatings were applied at a range of coat weights. A key focus here was the water vapor transmission rate, that dropped to a low level with a couple of different formulations. Papers that were coated on both sides had water vapor transmission rates that were below our detection limit.

DECICATION

DEDICATED TO MY PARENTS:

SAHEB ALSAADY & ELHAM ALSHAFEE

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- Special thanks to Dr. Douglas Bousfield for his willingness to have me as a member of his research group, even though he did not know me before I asked him to be my thesis advisor. Also thank you for your support, encouragement, and advice throughout the way!
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INTRODUCTION

Potato chips are the most popular snack food in the world. They account for one third of the global snack food market; it is a \$15 billion dollars industry (1). To keep potato chips crispy and increase shelf-life, special packing is needed. Some of the things that need to be kept in mind when engineering a packaging or bag for potato chips, include: moisture content, grease transmission rate, oxygen transmission rate, flexibility, sealing ability, shelf-life, toxicity, and price, see Figure 1. From potato chips to chocolate bars, snack packaging has been using material with excellent water, oxygen and grease resistance and has become the standard of the snack food industry. The most common material used in snack packaging is polypropylene (PP) plastic layers that are aluminum coated or metalized on one surface.

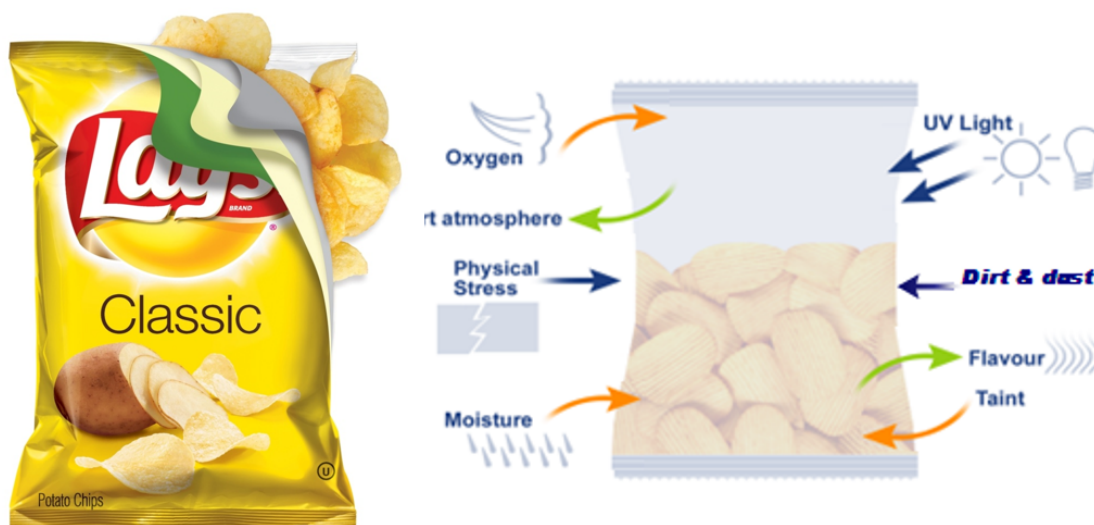


Figure 1. Potato Chip Bag Layers and Characteristics (2).

Most of today's potato chips bags have multiple layers that work together to provide the right environment for the potato chips. Typically, a potato chip bag has a Biaxially Oriented Polypropylene (BOPP) layer on the inside, which is used as a moisture

and grease barrier (3). The moisture barrier is needed to keep moisture that is in the air from getting to the chips and ruining their crispiness. The grease barrier is used so that bags do not get greasy on the outside. Next to the BOPP layer there is a Low-Density Polyethylene (LDPE) layer, which is used for its moisture and oil resistance, but also for its flexibility, toughness, and sealing ability (3). Some of the poor qualities of LDPE are that it has poor gas/oxygen resistance, has high static charge which can attract dust when stored, and has poor ink retainability. Following the LDPE layer there is usually a metalized BOPP layer, which is used as an oxygen barrier. BOPP alone has high oxygen transmission rates, which is why it needs to be metalized. An oxygen barrier is required to avoid oxidation of the fat in the potato chips, and in consequence spoiling. The choices are either to have a thick layer of PP or a different polyolefin, which is not desired, or to have a thin metalized layer. Metalizing the BOPP layer is done by placing the layer inside a vacuum chamber of 10^{-6} atm. The BOPP layer passes over molten aluminum; aluminum vapor is then sprayed or deposited on the surface of the film (2). The metalized layer does not only block gases, but it acts as an effective barrier to aroma and prevents light from entering. Light is avoided, because it can act as a catalyst for the oxidation of fat. Finally, an outer layer of Surlyn, which is a thermoplastic resin. It is mainly used for strength, flexibility, and ink retainability (3). Detailed layer formation of a typical potato chip bag is shown in Figure 2 below. The current desired criteria for packaging for snack foods provided by PepsiCo include an oxygen transmission rate (OTR) below 5 cc/m² day and a water vapor transmission rate of less than 0.1 g/m² day.

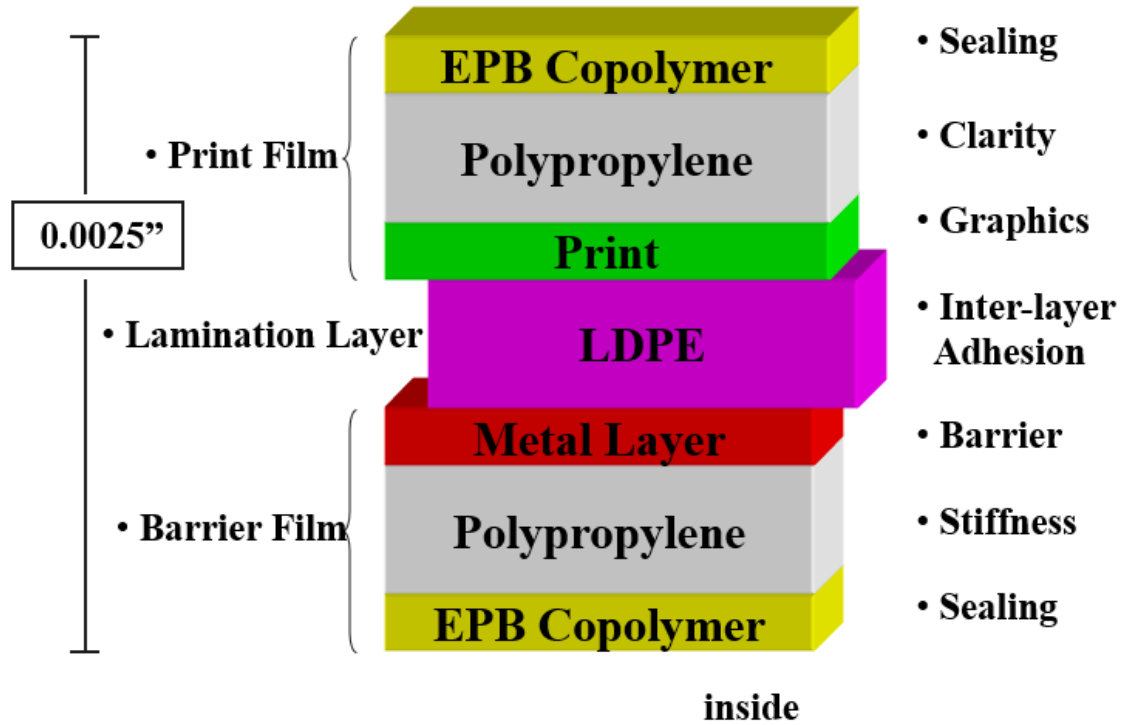


Figure 2. Detailed Breakdown of Layers of a Potato Chip Bag and their Functions (2).

While the current design is impressive, low cost, and widely used, the problem with current materials is that they cannot be recycled, even in the plastic stream, and do not break down in the environment (4). Polypropylene (PP) is a common plastic that is a member of the polyolefin family, which is produced from alkenes. PP can be recycled, but when other plastics are laminated to it, and the metal layer is present, current methods are not cost-effective and cannot separate the plastics types. The packaging material ends up in landfills at best, and if littered, they can end up in the landscape or ocean (4). Aluminum coated PP layers can take up to hundreds of years to decompose. In addition, non-renewable petroleum is used to produce the PP, and the production of aluminum requires a large energy input, mainly from the burning of fossil fuels. Aluminum production also generates a large amount of corrosive and toxic waste (4).

Plastic littering into the ocean is roughly seven million metric tons per year (5). This has created several garbage patches in the Pacific and Atlantic Oceans. One of the most well-known regions is the Great Pacific Garbage. Due to its chemistry, plastic is made to have a long life, which means that it does not degrade easily. Partial degradation over time leads to waste at macro and micro levels and the release of toxic chemicals in the ocean. Plastic and partially degraded parts may often be mistaken for food by many sea creatures, which leads to the death and deformities of a lot of marine life (5). Studies have shown that trace levels of plastic are being detected in human blood and cells (6).

This research is looking at using paper coated with layers of cellulose nanofibrils (CNF) and water based barrier coatings as a replacement for those aluminum coated PP layers (7). This will make the packing more easily biodegradable, recyclable, and renewable while keeping the same excellent characteristic of water, oxygen and grease resistance. Paper cannot be used alone because of its rough surface and high porosity. When coated with CNF or different barrier layers, some of the barrier material will fill in the pores, while also forming a smooth layer on top (7). Barrier coatings usually have excellent water vapor barrier properties, but they lack the oxygen barrier properties, which is why a combination of both CNF and other barriers are needed to do the job. The estimated production costs are similar to the current materials (7).

Cellulose is the most abundant organic biopolymer in the world. The annual production of cellulose worldwide is somewhere between 10^{10} and 10^{11} metric tons (8). It was first discovered in 1838 by Anselme Payen (8). Cellulose comes from wood, plant fibers, algae, fungi, and bacteria. Wood is primarily made of cellulose, so wood is the primary source. Cellulose is a white fiber-like structure with no odor. Its hierarchical

structure is composed of a linear homopolysaccharide of β -1,4-linked anhydro-D-glucose units (9). The cellobiose is the repeated segment of the structure, it contains the anhydroglucose units, which are the monomers that have three hydroxyl groups. These groups give cellulose its ability to form strong hydrogen bonds (8).

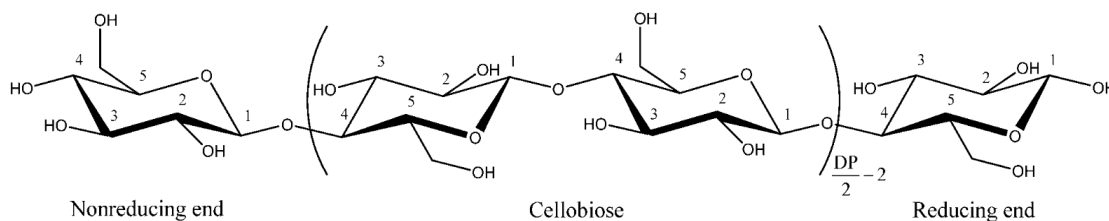


Figure 3. Chemical Structure of Cellulose (9).

What makes nanocellulose unique and a great material for packaging is the tight packing of cellulose chains (10). Nanocellulose is divided into two main groups, cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC). The process of making CNF involves, taking wood from trees turning it into small wood chips. The wood chips are then pulped to obtain wood fibers or pulp. The wood fibers are then mechanically ground to turn into nanofibrils. CNC is produced by strong acid hydrolysis.

Some of the strengths of CNF include excellent oxygen and grease barrier properties, good temperature resistance, and good tensile strength. In general, CNF has a temperature resistance of up to 200 °C, and a fiber tensile strength of 200 MPa (11). On the other hand, some of the weaknesses of CNF include: lack of flexibility, sealing ability, transparency, and moisture resistance (11). Previous research has shown that cellulose nanomaterials are excellent additives to base-paper products. Bulk addition in the mix and surface coating are both recommended. Bulk addition with pulp has shown that it changes the mechanical properties of the paper (10). When surface coated, CNF fills the voids between pulp fibers enhancing the properties of the base-paper; it provides

a non-porous layer. Figure 4 compares the surface of non-coated paper to that with a layer of CNF via a scanning electron micrograph conducted at the University of Maine. CNF made at the University of Maine has shown better oxygen barrier properties than higher grades of CNF and many plastics (12).

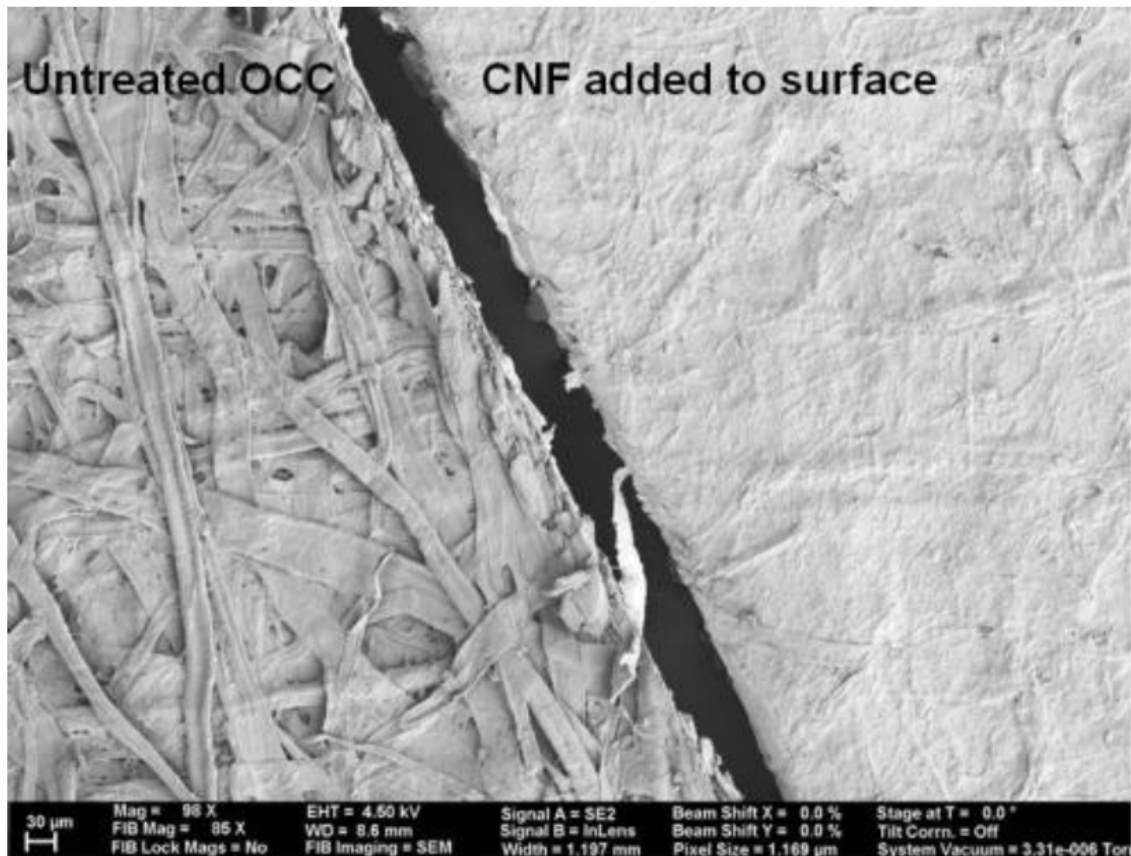


Figure 4. SEM images of paper and paper formed with a top layer of CNF applied in the forming section of the University of Maine pilot paper machine (7).

This research is part of the U.S. Endowment for Forestry and Communities (P3Nano). The endowment is a public-private partnership designed to develop and use wood-based nanomaterial for commercial products to keep the forests economy sustainable (13). The work presented here is part of a three-year project. The presented data are part of year one of the project which is mostly the bench scale that include: coating CNF layers on papers, coating water barrier layers, and exploring different paper

types. In addition, all samples were characterized in terms of coat weight, air permeability, water vapor transmission rate, and water vapor permeability using standard methods. After these tests are done, the most promising composition will then be improved and produced on a pilot scale during the second year. In the third and final year of the project, prototype packages will be produced, composting and recycling studies will be completed, and any other technical issues will be addressed.

The final goal at the end of the three-year project is to have pilot scale production of packaging material that uses CNF layering that would be able to compete economically and socially to replace the current aluminum coated PP design. The key question that is being addressed in this thesis is: can a layered structure of CNF, polymers, and pigments be cost effective and compete in performance with the industry standard?

MATERIALS

The three materials used in this study are: cellulose nanofibrils (CNF), polymers (latex) used as water vapor barrier, and pigments (clay) used as an alternative for the water barrier and to lower the cost of the polymers.

The CNF used in this research was made of pulp from St-Felicien Mill, Canada, NBSK: Softwood. The material was made from softwood components with deionization water as the water source. The pulp was mechanically refined or disengaged to turn into nanofibrils; the refining process is patented by the University of Maine and it was done using a stone refiner with a single rotating disc. The CNF was prepared by Gregory Yum using three different CNF samples, which were combined to form the mixture. Two of the components used in the making of the final mixture were made using continuous systems, and the third was produced using a batch process at 2.5%. The CNF mixture used had an 87.7% fiber fines with an average nanofiber bundle diameter of 54.4nm and a mean length of fibers above 200 μ m of 352 μ m. The full fiber size distribution is shown in Figure 5. The percent solid content of the CNF used was 2.1%.

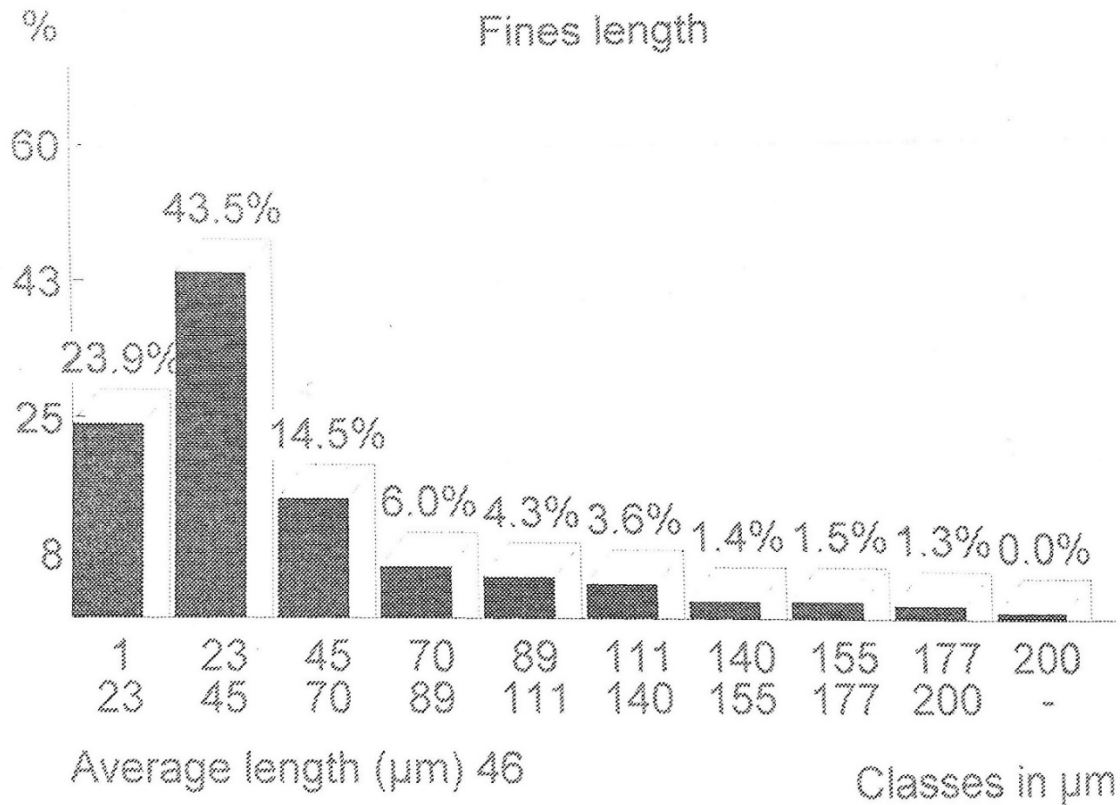


Figure 5. Cellulose Nanofibrils Breakdown of Fines Length.

Three types of polymers were used. Throughout the paper, they will be referred to as Polymers A, B, and C:

- Polymer A is a latex that serves as a water, grease, and moisture vapor barrier coating supplied by Mantrose-Haeuser titled VerdeCoat WB-10. This biopolymer barrier coating is made for biodegradable, compostable packaging. The VerdeCoat line of coating was tested by Western Michigan University for pulpability and recyclability and successfully passed (14). The polymer was tested under Vincotte EN13432 (ASTM 6400) standards for composability and biodegradability (15). They are designed to coat surfaces like cellulose and paper for packaging usage. The latex is an opaque white liquid that has a solid content of 36.5%. According to the supplier, the

liquid has a density of about 1.03 g/cm³ and a viscosity of 350-550 cps. Polymer A is FDA approved for food contact (aqueous, dry, and fatty food).

- Polymer B is a latex that also serves as a water, grease, and moisture vapor barrier coating supplied by OMNOVA Solutions titled X12-185. The latex is a milky white liquid with slight odor that has a solid content of about 42.0%. The liquid has a density of about 1.00 g/cm³. Little information is known about this product.
- Polymer C is a polymer that also serves as a water, grease, and moisture vapor barrier coating supplied by Michelman titled Michem Prime 4983R. The latex is a milky white liquid with slight odor that has a solid content of 24.8%. The liquid has a density of about 0.98 g/cm³. The chemical nature of the preparation is ethylene-acrylic acid dispersion. Some of this polymer's properties include: adhesion promotion, chemical resistance, and heat sealing ability (16).

Two kinds of pigment were tested:

- Pigment A is a tan liquid clay made by BASF and named Nuclay slurry that has a solid content of 71.1%.
- Pigment B is a green powder supplied by Minerals Technology and is called a Bentonite PGN clay. The clay looks and works like powdered cement. It absorbs great amounts of water when mixed with other materials.

Four different types of paper were tested:

- Paper A is made at the University of Maine, code T2303, that has a final base-weight of 80 g/m². This paper has 5% internal CNF to 95% paper stock. Paper A has an average thickness of 0.1033 mm.
- Paper B is also made at the University of Maine, code T2309, that has a final base-weight of 80 g/m². This paper has 5% CNF that is surface coated on the wet end of a paper machine. Paper B has an average thickness of 0.0987 mm.
- Paper C is made by Centre International de Couchage (CIC) in Canada. This is a specialty coated paper that was made in June of 2011, trial point 237. The base paper is BS 2011-61-18. Paper C has an average thickness of 0.0669 mm and an unknown base-weight.
- Paper D is regular Laser Print paper made by Hammermill. The paper has a weight of 90 g/m² (24 lb) and 98 brightness. Paper D has an average thickness of 0.1026 mm.
- Paper E is a thick purple divider paper. Paper E has an average thickness of 0.1327 mm, and unknown base-weight.

PROCEDURE

Before conducting tests, paper thickness tests were performed to find the thickness of the different base-paper that was being used. A TMI Model 49-87 Digital Micrometer was used to conduct the test. To perform the test, the paper was placed between the two plates of the machine, and the machine automatically display readings of thickness. Results are given in the descriptions of the different papers in the materials section above.

The coating process was performed using a laboratory bench-size automatic rod coater with standard Mayer Rods. Papers A and B were available on machine rolls that were 11 inches wide. When using Papers A or B, the paper was pulled and cut from the roll at a length of 15 inches. Papers C and E were pre-cut and had similar dimensions of 15 inches by 11 inches. Paper D was also pre-cut and had dimensions of 8.5 inches by 11 inches. After obtaining the correct type of paper to test, the paper was taped and secured to the top of rod coater surface. Taping played a critical role in preventing wrinkling; therefore, the entire width of the top of the paper was taped to the rod coater surface. After securing the paper, the automatic rod coater speed was adjusted to 3.5 rpm. Then the correct rod size was installed into the rod coater and the pressure knob was adjusted. The pressure knob changes the height between the rod and the rod coater surface, the numbers on the knob are unitless. Rod size and height of the rod play a critical role in determining the coat weight. After the rod was installed, the desired coating material was poured on top of the tape at the top of the paper. The rod coater was then turned on to

have the rod draw the coating on top of the paper. Once the rod reached the end and stopped, the rod coater was turned off. The coating process is completed at this point.

To dry the coated paper, the top of the paper was un-taped from the rod coater surface, leaving the tape attached to the paper. The paper was lifted from the rod coater, while it was kept as flat as possible, and placed on a drying surface. The paper was then taped from all four edges to the drying surface to prevent shrinking and wrinkling due to the water content of the coating material. The paper was left to dry overnight. When coating with a polymer, after the overnight room drying, the paper was dried in an oven for 15 minutes at 105 °C (125 °C was the desired). The drying process is completed at this point.

Through trial and error, it became apparent that it was best to pour excess coating material, more than what was theoretically calculated or needed. Usually, as the rod draws the material down the paper, some material gets pushed to the side and lost; therefore, excess amounts were used to insure equal coating. It was also extremely difficult to target a specific coat weight, especially when mixing more than one material. Therefore, I developed a general standard of rod size and pressure knob adjustment to use to obtain rough coat weight ranges. To obtain a very high coat weight, roughly 45+ g/m², the #8 Mayer rod was used with pressure knob setting of 7.50. Similarly, for a high coat weight, roughly 20-30 g/m², the #7 Mayer rod was used with pressure knob setting of 7.25. For a medium coat weight, roughly 15-20 g/m², the #5 Mayer rod was used with pressure knob setting of 7.00. Finally, for a low coat weight of roughly 4-15 g/m², the #3 Mayer rod was used with pressure knob setting of 6.75.

When coating CNF, 0.25 was added to the pressure knob settings stated above. CNF is thicker than the polymers used and it did not coat at a pressure knob setting below 7.0; the space between the rod and the sheet of paper becomes so small that the CNF simply gets pushed off the paper. When drying a paper that was coated with CNF, it was best to tape the paper along all four sides, because CNF is only 2-3% solid and the high-water content wrinkled the paper significantly. To prevent as much of that wrinkling as possible, extra tape to hold the paper was used. After drying the paper, wetting the paper a little with water using a spray bottle and placing it under a book or something heavy de-wrinkled the paper even more.

The viscosity of the different coating materials was not taken into account when coating the paper. Since we were targeting simply “high, medium, and low” coatings and not specified numbers, viscosity did not matter a lot, even though it impacted the coating a little. It is also important to note that the Mayer Rods used were old and had some old coating material cured on some regions. This prevented getting truly equal coating throughout the paper, but for the most part it was even. Some of the rods did not have numbers on them, so an educated guess was used to determine the size of the rod.

When comparing results, most graphs were plotted and fitted with power curves. The power curve trendline was chosen because we saw that as coat weight (x-axis) increased the WVTR (y-axis) reached an asymptote near zero. Similarly, when a power law is raised to a negative exponent, it gives a very similar trend. The power trendline curves were not used to represent physical characteristics or statistical performance, but simply for visual purposes only; to have an easier way to demonstrate the performance

and compare the results visually. That is why error-bars, r-squared values, and p-values were not used to determine how well the curves statistically-fit the data.

For some tests, CNF films were made and coated with polymers. Two different methods were used to make a CNF film. The first method included diluting the CNF until it became liquid-like (so that it can be easily distributed on a dish). Then depending on the thickness of the film, a calculated amount was poured into a petri dish. After pouring the CNF, the petri dish was shaken and spun to evenly distribute the CNF. Then it was let it dry for 24-48 hours inside a hood, to speed the process. Once dry, the CNF usually self-peel off the dish. At low weight the CNF film was fairly flexible and somewhat transparent; however, at high weight it becomes more rigid, brittle, and non-transparent milky-white, as seen in Figure 6.



Figure 6. Dry CNF films made in a petri dish (method 1).

The second method was developed by a member of this research team. The first method included diluting the CNF until it became liquid-like (so that it can be easily distributed on a dish). Then using a vacuum, funnel, and flask, a calculated amount of CNF was poured on top of a filter paper and drain the water. Once the water was drained, the filter paper with a layer of CNF was removed from the funnel and a ring was placed on the edges of the filter and a weight is placed on top to avoid wrinkling. Once dry, the CNF film was easily peeled off the filter. The final CNF film may be seen in Figure 7 below.



Figure 7. Dry CNF film made using vacuum and funnel (method 2). The filters used had dotted texture, and the texture printed on all the film.

Once it was time to test the WVTR, a minimum of four 70 mm circles were cut from each of the dry coated paper. Cutting near the edges of the paper was avoided, because the edges sometimes folded in the process of drying or transferring to the drying surface. After cutting the coated paper into 70mm circles, the circles were weighed to determine their coat weight:

$$\text{Coat Weight } \left[\frac{g}{m^2} \right] = \frac{m_{\text{coated}} - m_{\text{base}}}{A} \quad [1]$$

In the Equation 1 above, m_{coated} is the mass of the coated paper in grams, m_{base} is the mass of the base paper before in grams, and A is the area of the circle (diameter 70mm) in m^2 .

After determining the coat weight of each circle, they were tested for air permeability. This test did not only classify the air permeability and porosity of the paper and coating, but it also informed us if there was a hole in the space being tested. Air permeability can be a direct representation of the uniformity of material (17). The test was done using a Gurley Model 4340 Automatic Densometer & Smoothness Tester. Unlike the traditional, more popular manual Densometer, the Model 4340 is oil-free, so it did not ruin the samples. All the tests were done using a volume of 100 cc and a 1.0 in² orifice. The testable range for the Model 4340 is between 0 and 50,000 Gurley Seconds (17). Porous material gave low values for Gurley Seconds, and when the air permeability was low (not porous material) the readings were typically high. When air permeability was lower than the testable limit (high numbers), the machine read as “Too Dense” and gave a reading of 999,999,999.99. This was a constrain we faced when testing high coat weights and CNF films, but it showed that the sample was likely be a good barrier.

After determining the air permeability of each circle, a water vapor transmission rate (WVTR) test was conducted to characterize the paper and coatings for moisture resistance. WVTR is the volume of water vapor passing through the paper and its coating per unit of area and time under specified condition. The WVTR test was conducted according to TAPPI Standard conditions: T 448 om-09 “Water Vapor Transmission Rate of Paper and Paperboard at 23°C (73°F) and at 50% RH. (18) ” Using a modified combination of T 448 om-09 and ASTM E96 methods with Mason Jars, see Figure 8. ASTM E96 is a “Standard Test Method for Water Vapor Transmission of Material. (19)”

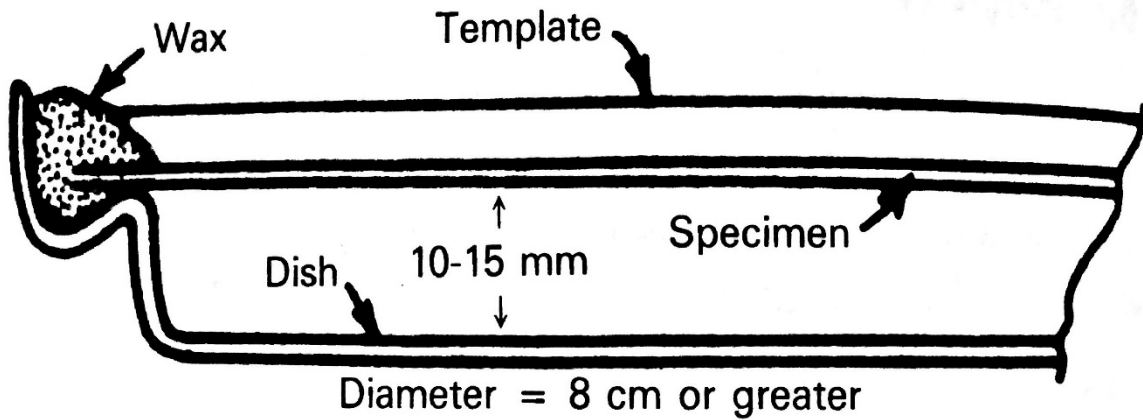


Figure 8. WVTR Test according to T 448 om-09 (17).

The WVTR tests were conducted in a special room set to TAPPI standards. Small Mason Jars (4 oz.) were used as the container to carry the test in, Figure 9 below. To decrease variation, 50 grams (± 0.05 g) of water was added to each jar. The cut-out circular paper was placed on top of the jar, followed by a silicone seal, and finally the metal screw band of the jar (the lids were not used). The sides of the jar and the metal screw band took 10mm off the paper, leaving the circle with a diameter of 60mm as the area used for permeability.

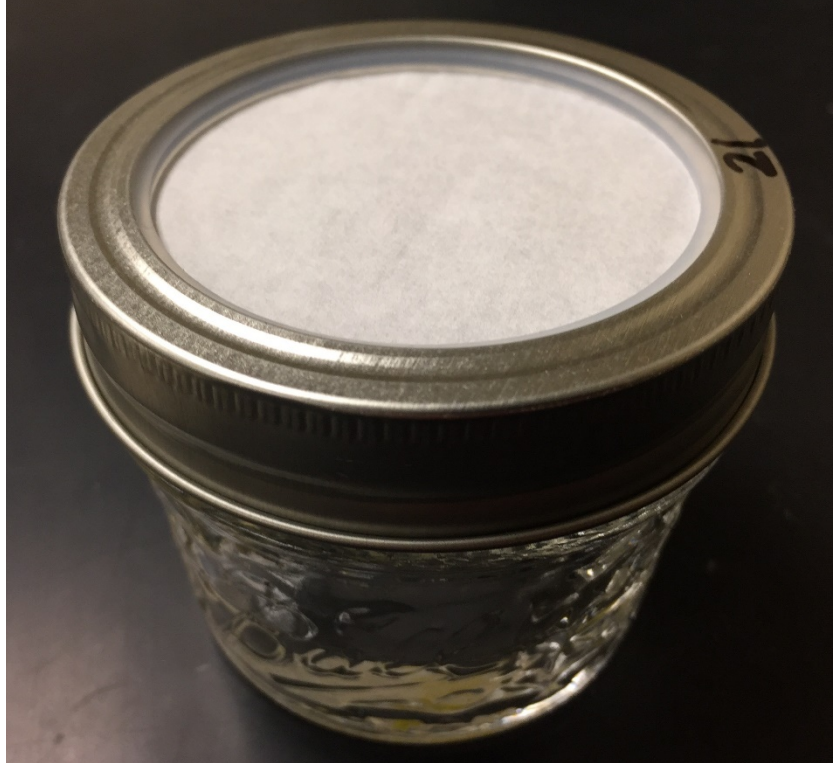


Figure 9. Mason jar apparatus.

After assembling the apparatus, the jar (with all of its attachments) was weighed and left to sit in the room under standard conditions for 24 hours. After 24 hours ($\pm 0.5h$), the jars were weighed again. The difference between the final and initial mass gives us the WVTR.

$$WVTR \left[\frac{g}{m^2 \cdot day} \right] = \frac{m_f - m_i}{A_p} \quad [2]$$

In the Equation 2 above, m_f is the mass of the mason jar apparatus at the end of the test; m_i is the mass of the mason jar apparatus at the beginning of the test; and A_p is the area of the transfer surface. The area of the transfer area is different than the area of the circle, this is the area of the permeable section, which has a diameter of 60mm. The term A_p is multiplied by the number of days the tests was conducted (usually 1 day).

Standard methods recommend using gel or wax to securely seal the paper and eliminate any leaks from the jar. Previous research groups members have modified the lid of the mason jars and used them as seals, while others used rubber seals. I found that Amazon sells professionally-made Silicone Gasket Sealing Rings that are used for Mason Jars specifically, Figure 10, more information may be found using the link in the References section (20). The seals are made of food grade silicone and are claimed to not leave off any chemicals. The silicone seals are perfect and eliminate leaks almost completely (WVTR = 0 for the first 24 hours).

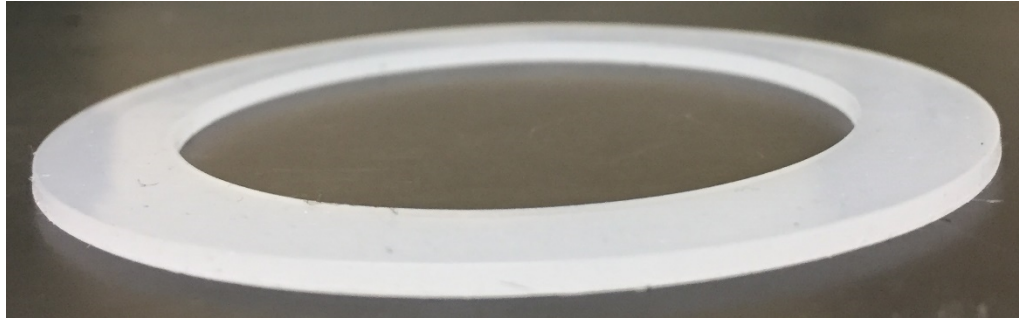


Figure 10. Silicone Gasket Sealing Ring

There is high vapor pressure inside the Mason Jar and low vapor pressure in the outside controlled environment. This causes water molecules to pass through the permeable material resulting in a mass loss. The weight over 24 hours gives the WVTR for 24 hours.

Water vapor permeability (WVP) was then calculated using WVTR data. Permeability gives a different prospective to look at the collected data.

$$\Delta P^* [Pa] = P_{sat.}^* \times \Delta RH \quad [3]$$

$$Permeance \left[\frac{g}{m^2 \cdot Pa \cdot day} \right] = \frac{WVTR}{\Delta P^*} \quad [4]$$

$$WVP \left[\frac{g}{m \cdot Pa \cdot day} \right] = Permeanace \times t_{coated} \quad [5]$$

In the Equation 3, ΔP^* is the difference in vapor pressure in Pa; P_{sat}^* is the saturated vapor pressure in Pa; and ΔRH is the difference in relative humidity inside and outside the jar. In the Equation 5, water vapor permeability (WVP) is calculated by multiplying permeance by the final thickness of the coated paper in m².

To examine the effect of coating alone on permeability, the effect of base paper had to be eliminated. Therefore, the above equations were modified to be:

$$WVP_{coat} \left[\frac{g}{m \cdot Pa \cdot day} \right] = \frac{t_{coat}}{\frac{WVTR}{\Delta P^*} - \frac{t_{base}}{WVTR_{base}}} \quad [6]$$

Equation 6 was derived by Randy Raditya, a PhD student of this research group (21). In the above equation, WVP_{coat} is the water vapor permeability of the coating alone, t_{base} is the thickness of the base paper, and $WVTR_{base}$ is the water vapor transmission rate of the base paper. t_{coat} is the thickness of the coating, which is calculated by dividing the coat weight (g/m²) by the density of the dry coating material (g/m³), given by Equation 7:

$$t_{coat} [m] = \frac{w_{coat}}{\rho} \quad [7]$$

CONDUCTED TRIALS

The following trials testing for WVTR were conducted:

- **Experiments testing different polymers:**
 1. Polymer A coated on Paper A at four different coat weights.
 2. Polymer B coated on Paper A at four different coat weights.
 3. Polymer C coated on Paper A at four different coat weights.

- **Experiments testing different paper types:**
 1. Polymer A coated on Paper A at four different coat weights.
 2. Polymer A coated on Paper B at four different coat weights.
 3. Polymer A coated on Paper C at four different coat weights.
 4. Polymer A coated on Paper D at four different coat weights.
 5. Polymer A coated on Paper E at four different coat weights.

- **Experiments testing the addition of clay (pigment) to polymers:**
 1. Polymer A mixed with Pigment A coated on Paper A at four different coat weights.
 2. Polymer A mixed with Pigment B coated on Paper A at four different coat weights.
 3. Polymer C mixed with Pigment A coated on Paper A at four different coat weights.
 4. Polymer C mixed with Pigment B coated on Paper A at four different coat weights.

- **Experiments testing other coatings:**

1. Double coating of Polymer A (one layer on each side) on Paper A at four different coat weights.
2. Heavy coating of Polymer A on Paper A (4-day experiment).
3. Polymer A coated on Paper A (side 1), then CNF was coated on the second side of the paper (four different coat weights total).
4. Polymer C mixed with CNF and coated on Paper A at four different coat weights.
5. CNF films covered with Polymer A and coated on Paper A (4-day experiment).
6. Sun Chips Original potato chips bag alone.

RESULTS AND DISCUSSION

The first test that was conducted examined the performance of the different polymers being tested. Paper A was used as the base-paper for all three polymers. When conducting the Gurley Air Permeability test on the papers coated with the three polymers, the machine read “Too Dense” for Polymer A and B at all coat weights, but all of Polymer C’s readings were below 25,000 Gurley Seconds. Detailed readings are given in Table 1-3 in the Appendix. Figure 11 below shows the WVTR performance of the three polymers. Polymer C performed in a different manner than Polymers A and B. It was hard to get very high coat weight using Polymer C, mainly because it had the least solid content of 24.8%. The polymer beaded-up like rain drops on top of a water resistance surface, when dried, it was cured in patches. It was very liquid-like, a lot of Polymer C was added to the base-paper that the paper was completely soaked, but the coat weight was relatively lower than the two other polymers.

These trials were not done at 50% relative humidity, as set by TAPPI Standards. The room in which the test was carried out in had trouble controlling humidity. The humidity in the room was about 25% RH. We used linear interpolation of vapor pressure to correct for the change. The TAPPI Standard states that linear interpolation is not ideal and not recommended, but previous lab members had seen that changes for the first 24 hours could be modeled linearly. Equation 8 was used to calculate the correction factor:

$$\text{Correction Factor} = \frac{P_{sat.} - P_{standard}}{P_{sat.} - P_{actual}} \quad [8]$$

The correction factor was multiplied by the amount of water loss to correct for humidity. In the expression above, $P_{sat.}$ is the saturated vapor pressure, $P_{standard}$ is the desired vapor pressure at 50% RH, and P_{actual} is the actual vapor pressure at the current relative humidity.

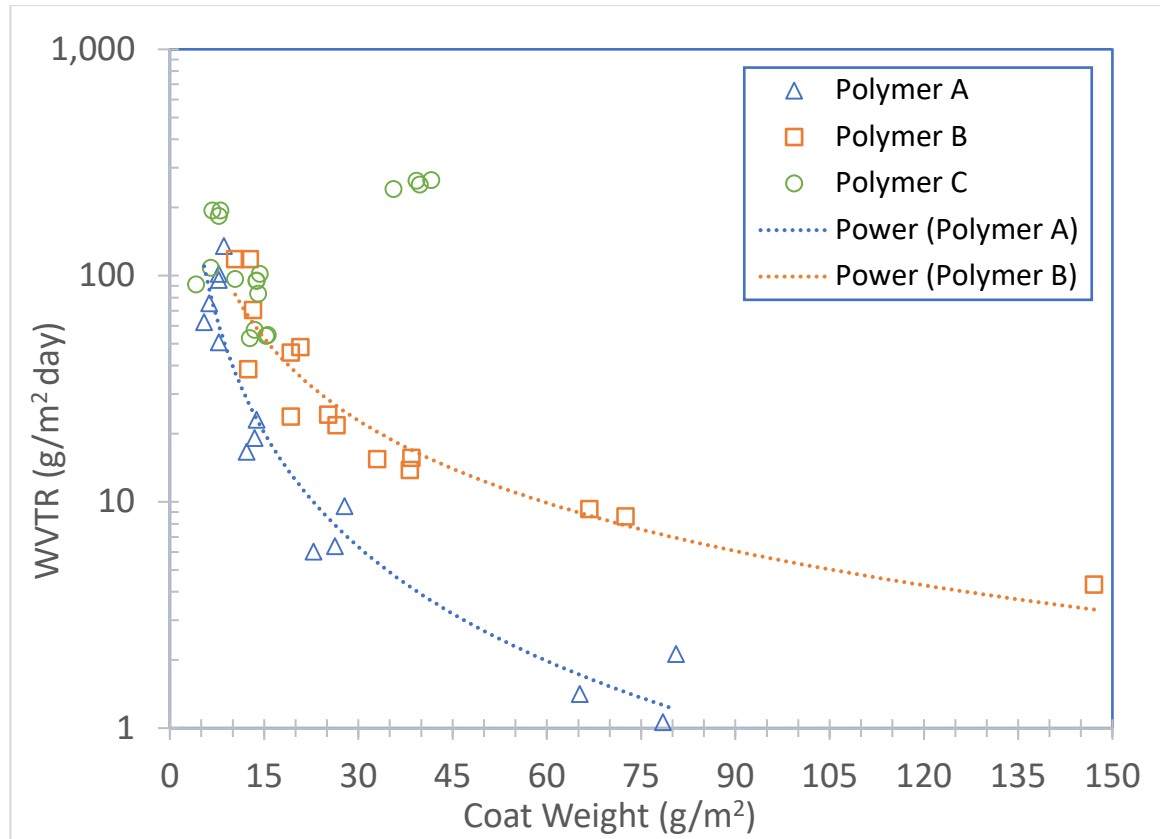


Figure 11. Data comparing polymers A, B, and C coated on Paper A (5% internal CNF).

As shown by Figure 11, at coat weights below 25 g/m², Polymer C's WVTR decreased with slightly increases in coat weight; however, at higher coat weight the WVTR increased. Log scale was used to better show results at low WVTRs, expanding the region between 1 and 10 06 $\left[\frac{g}{m^2 \cdot day}\right]$. Power fit was performed on all three polymers, but it did not give a good visual fit for Polymer C. As mentioned before, power fit was used to simply show the performance in an easier way, it is used for visual purposes only. The power fit is not a perfect representation, as the data has some variance due to the

difficulty of getting a completely smooth surface with a rod coater, but it is a fair visual representation of the trend or performance of the polymers. Polymers A and B performed in similar manners; WVTR decreased with increase in coat weight, which was expected. As coat weight increased, the polymer started to not only fill the porous in the base-paper, but it also formed a uniform layer on top, creating a moisture barrier. As seen by the blue power fit-line and the actual data (blue triangles), Polymer A had lower WVTR values for the same coat weights as Polymer B. This suggests that Polymer A performed slightly better. The lowest WVTR obtained was $1.06 \left[\frac{g}{m^2 \cdot day} \right]$ using Polymer A at a coat weight of 78.5 g/m^2 . The uncoated base-paper had a WVTR of roughly $400 \left[\frac{g}{m^2 \cdot day} \right]$. This means that Polymer A decreased the water vapor transmission by 99.73%, relative to the base-paper alone.

To conclude the test between the different polymers, Polymer A and B were both about equally good, but Polymer A was slightly better. Polymer B was harder to use, because: it dried at a slightly slower rate, was extremely sticky (cleaning the Mayer Rods and rod coater took more time and scrubbing), and when it dried the coating still had some stickiness to it and that could cause problems in mass production and rolling the paper. One benefit of Polymer B, is that it makes the paper slightly more flexible than Polymer A, which is desired in packaging. Though, in the end, Polymer A seemed like the better performer and it was easier to use. Since the results obtained with a single layer of Polymer A were only ten times the desired WVTR value, we tried double coating of Polymer A on Paper A, on the same side, but it was not successful, see Figure 12 below. The first coating was applied and dried successfully, but adding a second layer on top

was not possible. The polymer beaded up, which suggested that Polymer A made a smooth, resistant surface that was liquid-proof.



Figure 12. Double coating of Polymer A on Paper A on the same side.

Similarly, we attempted double coating of Polymer C on Paper A on the same side, we were trying to see if the second layer would cure with the first layer and fill the gaps that were left from the first coating. The coating was also unsuccessful, see Figure 13 below. The first coating was applied and dried in a similar way to the single layer discussed in Figure 11. The second layer was coated successful, but did not dry correctly. While drying, the second layer of coating flaked and became like crystals.

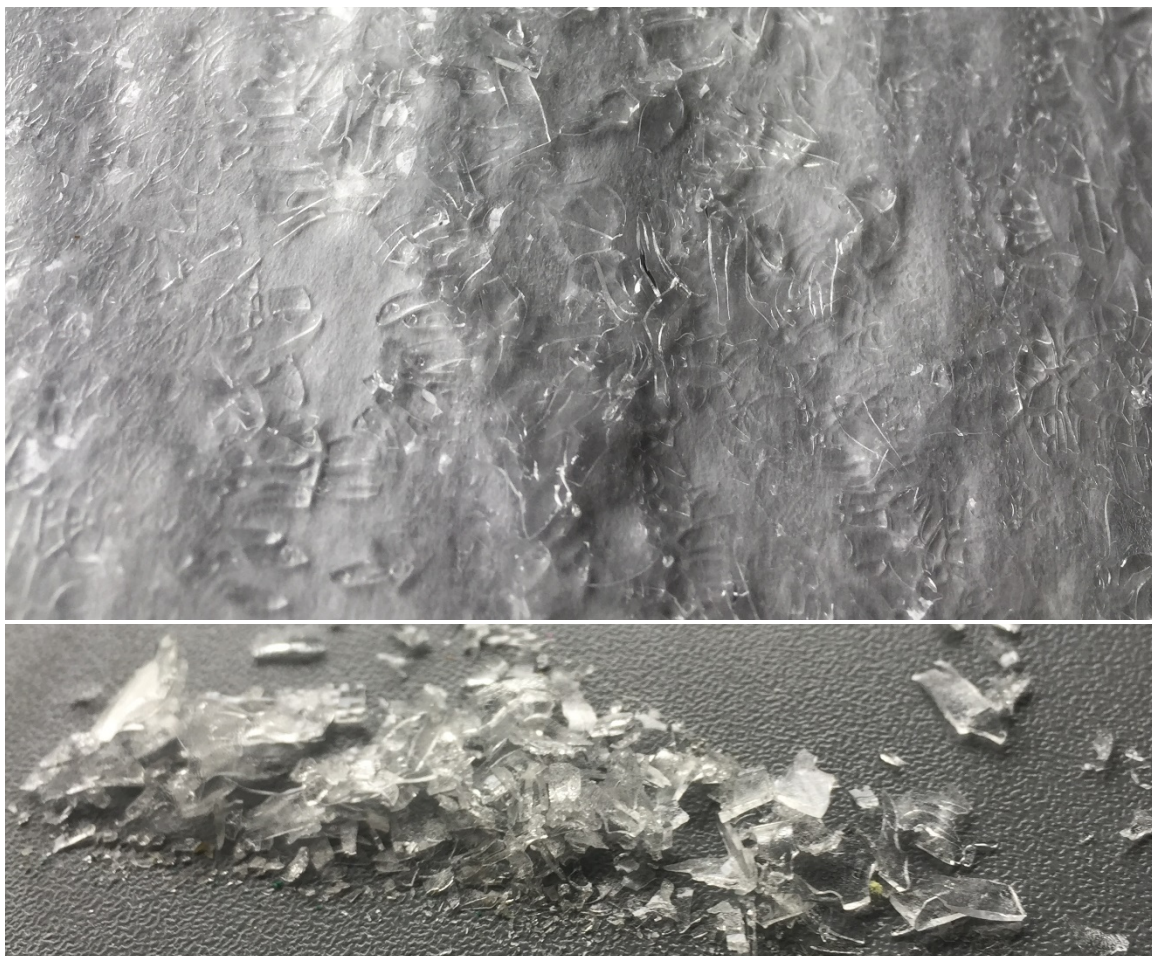


Figure 13. (Top picture) Double coating of Polymer C on Paper A on the same side. (Bottom picture) Crystal flakes that fell off the paper.

Tests examining the different base paper types were conducted next. Figure 14 shows the results obtained for different papers, using Polymer A. The y-axis is given in logarithmic scale to present the data more clearly. Similar to the previous set of experiments, the humidity in the room was about 25% RH. We used the same correction factor given in Equation 8 to correct for the change.

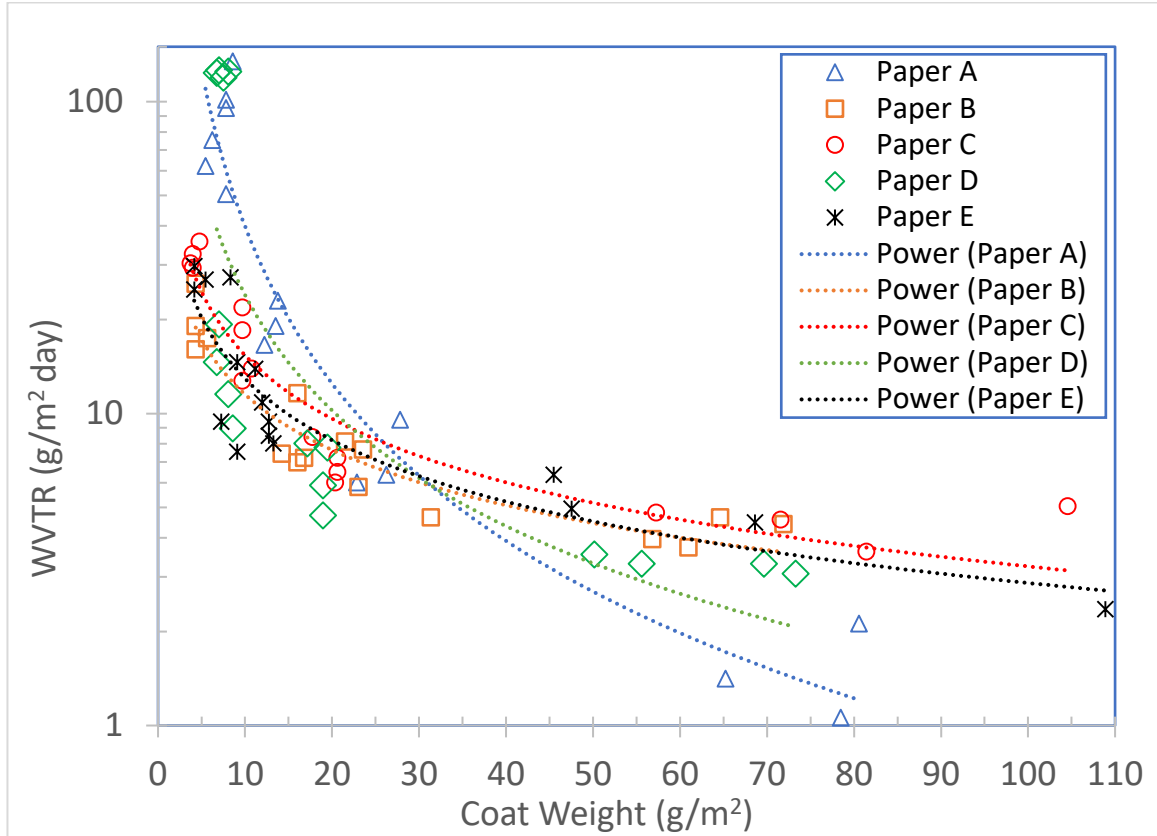


Figure 14. Data comparing Paper A, B, C, D, and E coated with Polymer A.

Power fit was applied to the data to simply show the performance in an easier way, it was used for visual purposes only. The data obtained for the different papers had a lot of variance as seen by the scatter of the points in Figure 14. Power fit was the best visual fit, but it was still not very accurate. At coat weight below 30 g/m², it was hard to distinguish a significant difference in performance as all the papers decreased WVTR about equally with slight increases in coat weight. Paper A gave some of the worst results at a coat weight of about 10 g/m². Since the goal was to get a WVTR below 0.1 $\left[\frac{g}{m^2 \cdot day}\right]$, higher coat weights were targeted. Upon close examination of the trend line and the scattered data points, at a coat weight higher than 30 g/m², Paper A performed the best, even though it had a variance of about 1 $\left[\frac{g}{m^2 \cdot day}\right]$. Paper D (laser print paper) was better than Paper A at coat weight below 30 g/m², but slightly worse above that coat weight.

Papers B, C, and E gave roughly the same results for all the high coat weights; this may be seen by the close power fit line and the overlaying data points. Part of the reason of the poor performance of Papers C may be because the paper was old (made in 2011). When coating Paper C with Polymer A, the base-paper ripped at the edges. The results of Papers A-E coated with Polymer A are contained in Tables 4-8 in the Appendix. The tables also contain detailed air permeability data and water vapor permeability for each sample.

As stated earlier, the goal was to get a WVTR below $0.1 \left[\frac{g}{m^2 * day} \right]$, higher coat weights were targeted; therefore, Paper A gave the best results for higher coat weights with low WVTR values. All experiments from that point on were conducted using Paper A, mainly due to the performance at high coat weight, but also because we had the most supplies of Paper A (enough to run trials in the future as well). The controlled humidity in the room was also fixed after this set of experiments, so all the proceeding results were done at TAPPI Standard Conditions: at 23°C (73°F) and 50% RH.

After determining the best polymer and paper type, water barrier mixtures were tested. Typically, water barrier coatings consist of a polymer and an added pigment (clay) to lower the cost. Pigments are usually cheap and perform well when mixed at lower concentrations with a polymer. The polymer to pigment ratio used was 75:45, respectively, in terms of dry weight. The ratio was recommended by a PhD student who was testing the strength of coating relative to the pigment amount in the same P3Nano research project. All mixtures had pigment weight percent of 37.5%. Pigment weight

percent was calculated by dividing the dry weight of pigment (w_{clay}) by the sum of the dry weight of pigment and polymer:

$$\text{Weight Percent } [\%] = \frac{w_{clay}}{w_{clay} + w_{polymer}} \quad [9]$$

The added pigment made the coating brittle; at high coat weight the coating broke, which destroyed some samples. The highest coat weight of the mixture of Polymer A and Pigment A is shown in Figure 15 below, which was destroyed due to the brittleness of the coating.



Figure 15. Highest coat weight of Polymer A + Pigment A mixture.

One of the challenges to target in the future would be finding the right amount of clay where the cost will be low enough and the product is flexible enough. Results of the four trials that compared mixing Polymers A and C with Pigment/Clays A and B are given in Figure 16 below. Detailed results are also given in Tables 9-12 in the Appendix.

The tables also contain detailed air permeability data and water vapor permeability for each sample.

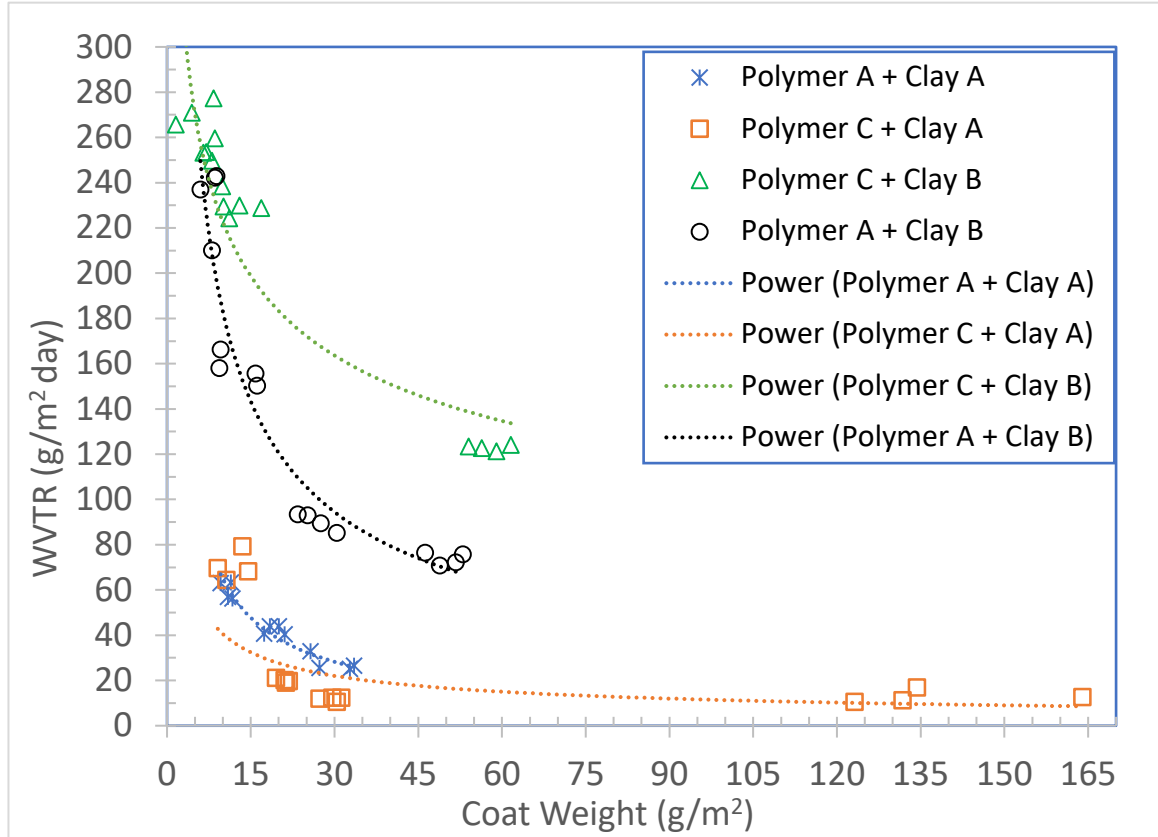


Figure 16. Data comparing mixtures of Polymers and Clays coated on Paper A.

Pigment (clay) B decreased the performance of Polymers A and C significantly; WVTR were above 60 $\left[\frac{g}{m^2 \cdot day}\right]$, even at high coat weight. The mixture of Polymer C and Pigment A gave the best results, detailed data given in Table 10 in the Appendix. The performance of Polymer C improved when it was mixed with Pigment A, but it was still worse than Polymer A alone, as seen in Figure 16. The best WVTR was 10.6 $\left[\frac{g}{m^2 \cdot day}\right]$, using the mixture of Polymer C and Pigment A at a coat weight of 123 g/m², but it was worse than coating the paper with Polymer A alone, which gave 1.06 $\left[\frac{g}{m^2 \cdot day}\right]$. Mixing Polymer A with Pigment A worsened the results, compared to just coating the paper with

Polymer A alone. At a coat weight of 30 g/m², Polymer A alone gave a WVTR of $9 \left[\frac{g}{m^2 \cdot day} \right]$, while the mixture of Polymer A and Pigment A gave a WVTR of about $25 \left[\frac{g}{m^2 \cdot day} \right]$. Complete data of WVTR, air permeability, and WVP are given in Tables 9-12 in the Appendix.

To look at the results above from a different perspective, Figure 18 compares the modification of Paper A and Polymer A with different clays, CNF layer, and double coating. After testing combinations of pigments with Polymer A, testing the addition of a CNF layer was desired. At first, we attempted coating CNF on top of a cured layer of Polymer A on Paper A (same side), but it was not successful, see Figure 17 below. The first coating of Polymer A was applied and dried successfully, but adding a CNF layer on top was not possible. Polymer A made a smooth, resistant surface that did not allow CNF to be coated on top of it.



Figure 17. Coating CNF on top of a dried layer of Polymer A on Paper A

Instead, we coated Paper A with a layer of Polymer A on one side and a layer of CNF on the other side. To avoid wrinkling of the paper, Polymer A was coated first, dried overnight, and cured in an oven for 15 minutes. After that it was coated with a layer of CNF on the other side. Even coating on a different side was not easy; when coating with Polymer A, the paper absorbed some of the polymer and displayed a little of its liquid resistance properties on the other side. This further supports that the polymer/later is not only surface coating, but also filling the pores in the base-paper. Once everything dried, the CNF layer was easy to peel off. When testing for air permeability, the region that was tested was a little damaged (sunburn-like peeling). The results of Paper A coated with a layer of Polymer A on one side and a layer of coated CNF on the other side are

contained in Table 17 in the Appendix. The results were promising, but more future work is needed. The best results were obtained around a total coat weight of 57 g/m², which gave a WVTR of about 2.2 [$\frac{g}{m^2 \cdot day}$]. A single layer of Polymer A alone gave better results than coating with an additional CNF layer, as seen by the comparison in Figure 18 below. Coating the CNF first and de-wrinkling the paper might be a better way to re-test this combination in the future.

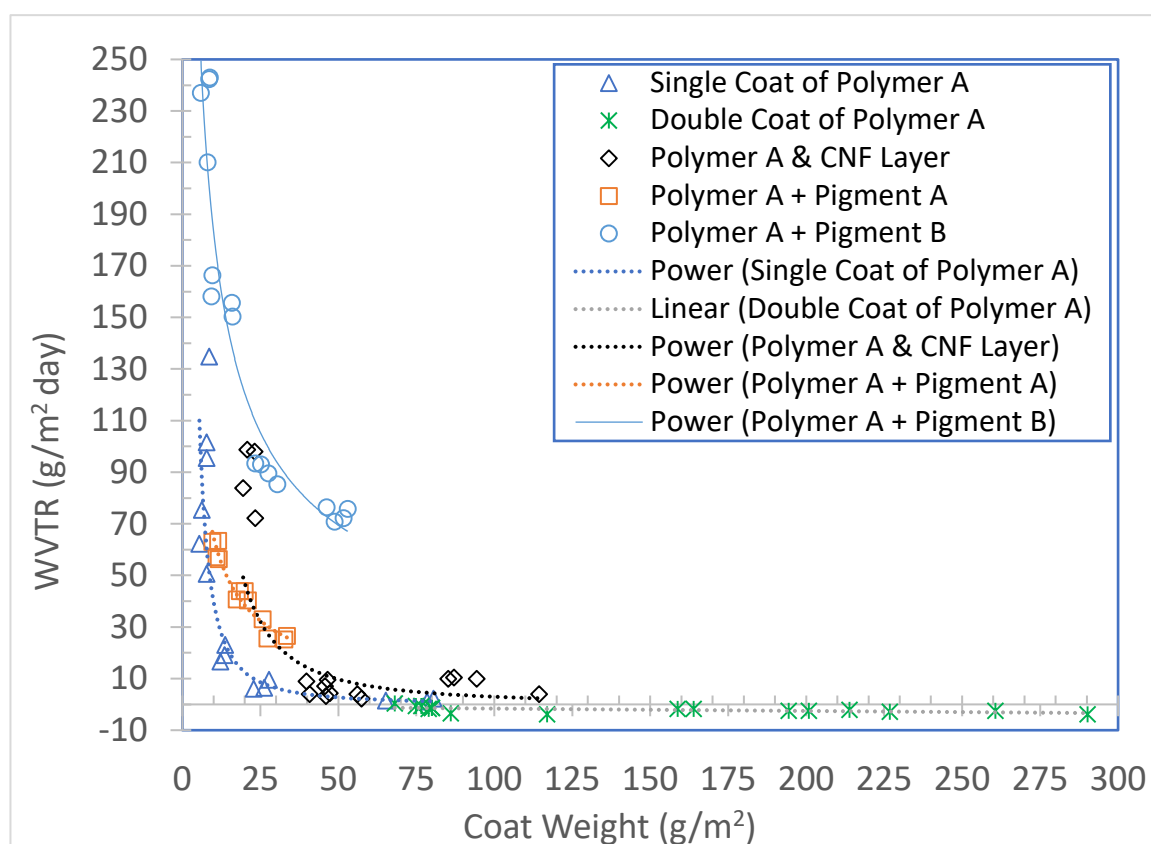


Figure 18. Data comparing mixtures of Single and Double Layer of Polymer A, Polymer A mixed with Pigments A and B, and Polymer A and a layer of CNF: all coated on Paper A.

In the final product, a heat sealable layer will be needed on both sides of the product. Therefore, a series of tests were done coating Paper A with a layer of Polymer A on each side, results are contained in Table 13 in the Appendix. This gave the most promising and interesting results. The second coating did not have as smooth of a surface

as the first one. The total coat weights were between 75 and 290 g/m². Small negative WVTR values were obtained for all but one sample, green points in Figure 18. The result was hard to explain at first, because the jar had gained weight over the 24 hours. This result must come from the adsorption of moisture to the sample on the room-side surface of the coated paper, which increased the weight of the jar. These coatings must block moisture from leaving to such a small level that small gains of weight of the layer can be seen. Even after leaving the samples for several days, no weight loss was measured for a number of these samples.

Looking back at these results, it would be best to condition the samples in the room for a few days before doing the WVTR test. This would allow the samples to come to some equilibrium water content. This method should eliminate any negative values of WVTR.

A small experiment examining heavy coating similar to double coating was conducted to see how the results would look over several days, shown in Figure 19 below. The results of this experiment are contained in Tables 14-16 in the Appendix. As it may be seen, over the days the data goes from negative WVTRs to positive WVTRs. On the first day, the data looks similar to that of the double coating of Polymer A, negative WVTR values. Also, as coat weight increased, the values of WVTR were negative for longer periods of time. As time went by, water vapor started to go up the layers of coating and paper and eventually pushed the moisture on top and left the jar, decreasing the weight (giving positive WVTRs). This double layering or heavy coating of Polymer A should be tested again with conditioning in the future over several days to see if weight loss occurs over longer period of time.

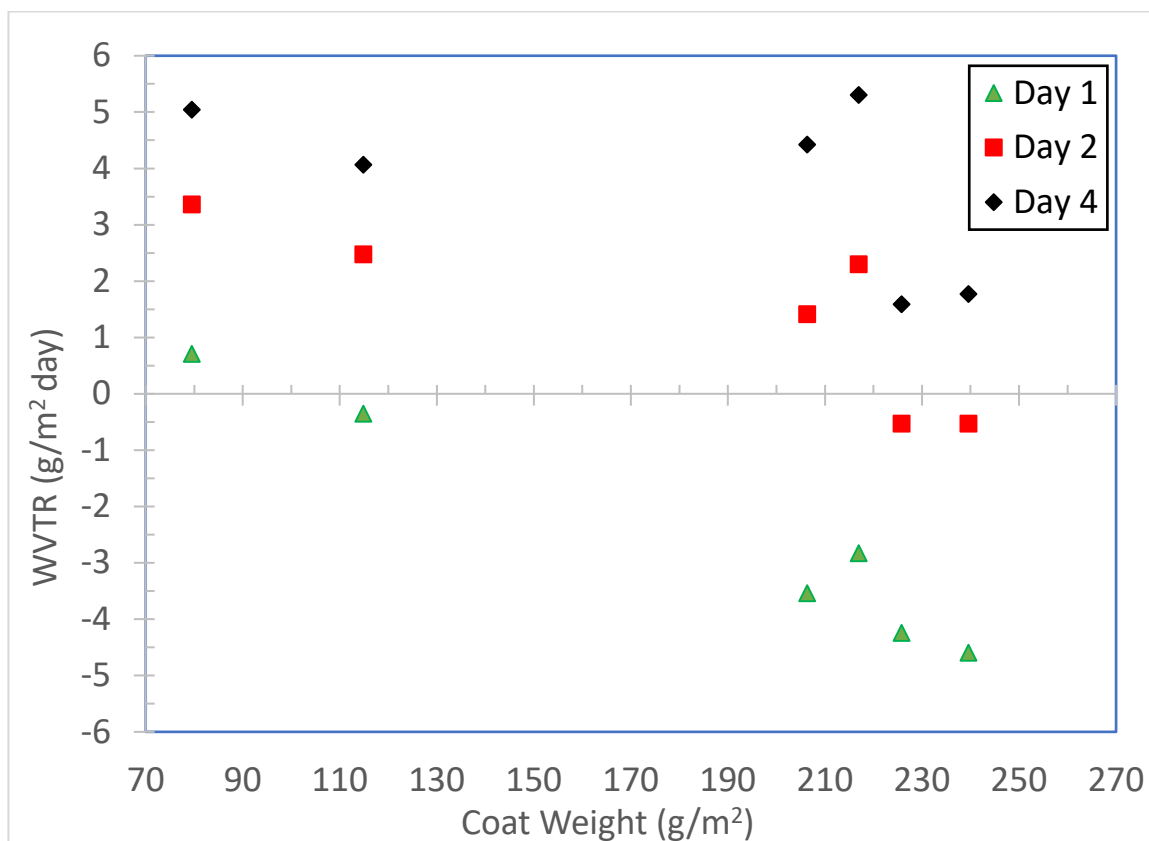


Figure 19. Paper A coated with heavy coat weight of Polymer A.

To conclude on the tests done on Polymer A so far, with the results shown in Figure 18, mixing Polymer A with Pigment B worsened the results, dramatically, compared to just coating the paper with Polymer A alone. Mixing Polymer A with Pigment A also worsened the results, compared to just coating the paper with Polymer A alone, but it was not as bad as Pigment B. New mixtures of lower Pigment A weight percent might give desirable WVTR, while lowering the coat weight of the water barrier coating. Adding a CNF layer also gave worse results than just a single layer of Polymer A, but investigating the addition of the CNF layer before the polymer layer might be something worth doing. Assuming the interpretation given for double coating is accurate, double coating Paper A with two layers of Polymer A (one on each side) or super high

coat weight of a single layer of Polymer A might give us the best desired results of a WVTR of $0.1 \left[\frac{g}{m^2 \cdot day} \right]$.

Similar to Figure 18, Figure 20 compares the coating of Paper A and Polymer C with the different clays mentioned earlier, and Polymer C mixed with CNF. Polymer C alone is very liquid-like, so adding the pigment thickened the mixture and improved performance. This is very clear in how most modification/mixes of Polymer C with pigment and CNF improved WVTR. As mentioned before, the performance of Polymer C improved when it was mixed with Pigment A, the best WVTR was $11 \left[\frac{g}{m^2 \cdot day} \right]$. As indicated before, the performance of Pigment B worsened the WVTR when mixed with Polymer C (also when mixed with Polymer A).

The comparison of Polymer C with pigment and CNF is shown in Figure 20. As coat weight increased, WVTR decreased, with the best WVTR being $13.7 \left[\frac{g}{m^2 \cdot day} \right]$ at a coat weight of 165 g/m^2 . The best results were obtained when Polymer C was mixed with Pigment A, though, performance was ten times worse than coating a paper with Polymer A alone.

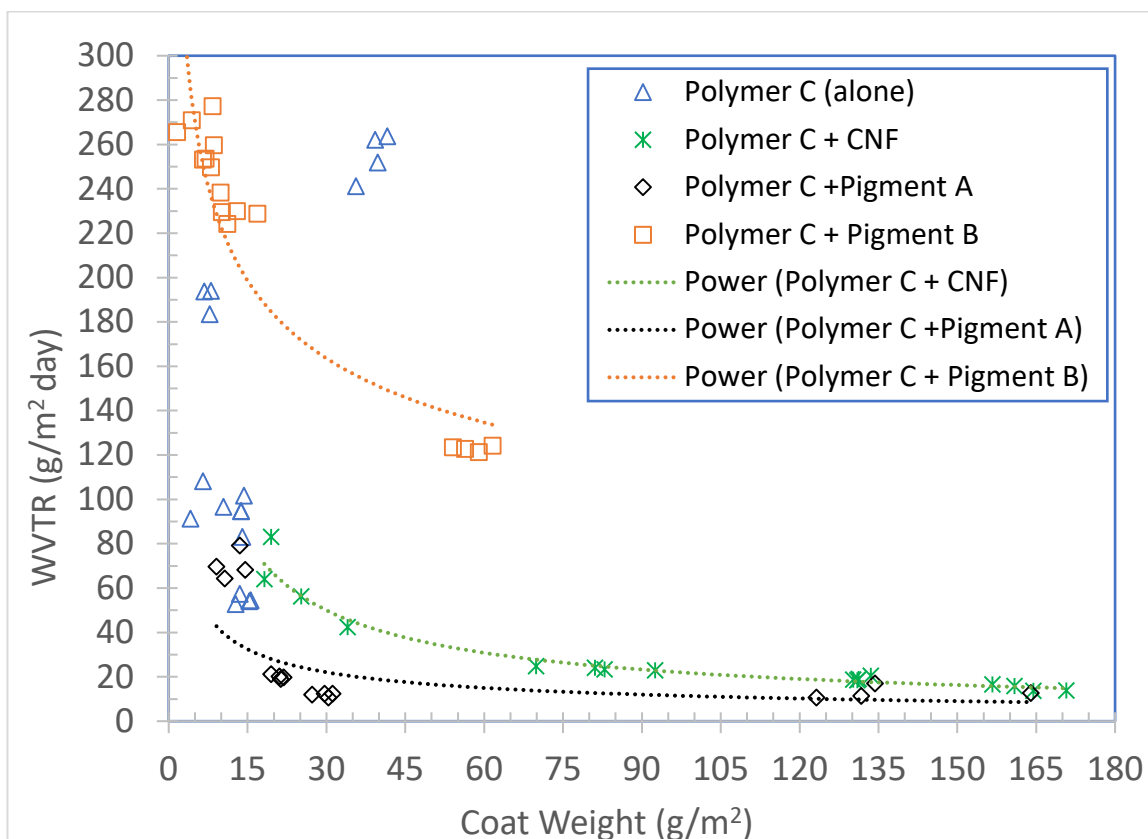


Figure 20. Data comparing mixtures of Single Layer of Polymer C, Polymer C mixed with Pigments A and B, and Polymer C mixed with CNF: all coated on Paper A.

When Paper A was coated with a mixture of CNF and Polymer C, the paper became transparent after it dried. Detailed WVTRs, WVPs, and air permeability results for Paper A coated with a mixture of CNF and Polymer C are given in Table 18 in the Appendix. When coated with the CNF mixture, the mixture bled through the paper to the other side. The first time this was attempted, the paper got glued to the drying surface overnight and became extremely hard to remove. The second time this was attempted, the paper was dried in an oven for 10 minutes, then left overnight on the dry surface to air-dry, and finally cured in an oven for another 10 minutes. A picture of the paper is shown in Figure 21 below. The oven marks can be seen on the paper, and it became transparent and extremely strong. The paper became like plastic, and it was hard to cut through it.

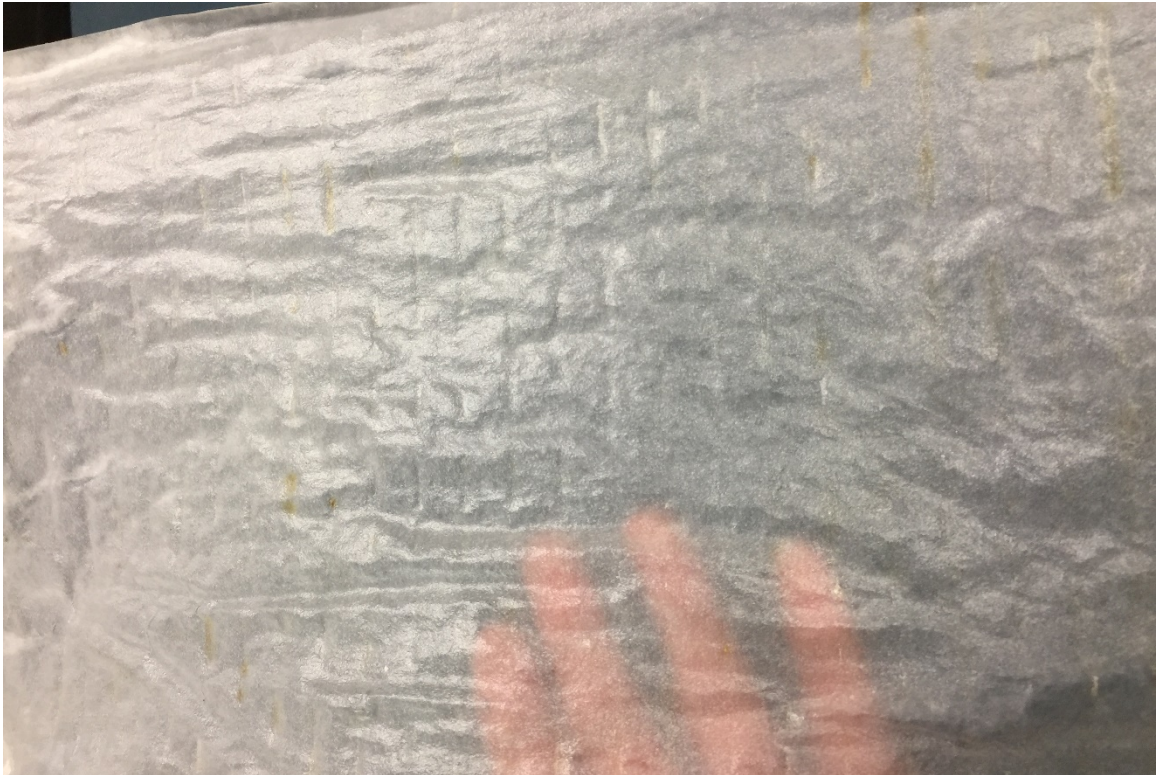


Figure 21. Paper A coated with one layer of a mixture of Polymer C and CNF.

To wrap up the experiments, I conducted a test by covering CNF films with Polymer A (dipping the film in a bowl of Polymer A) then flattened them on Paper A and covered them with a heavy layer of Polymer A. Detailed results are contained in Tables 19-21 in the Appendix. The films were not completely flat on the paper, so they formed air gaps in between them and the paper. Figure 22 below shows the WVTR results over a four-day period. As it may be observed, over the days the data goes from negative WVTR values to positive WVTR values. On the first day, the data looked similar to that of the double coating of Polymer A, negative WVTR values.

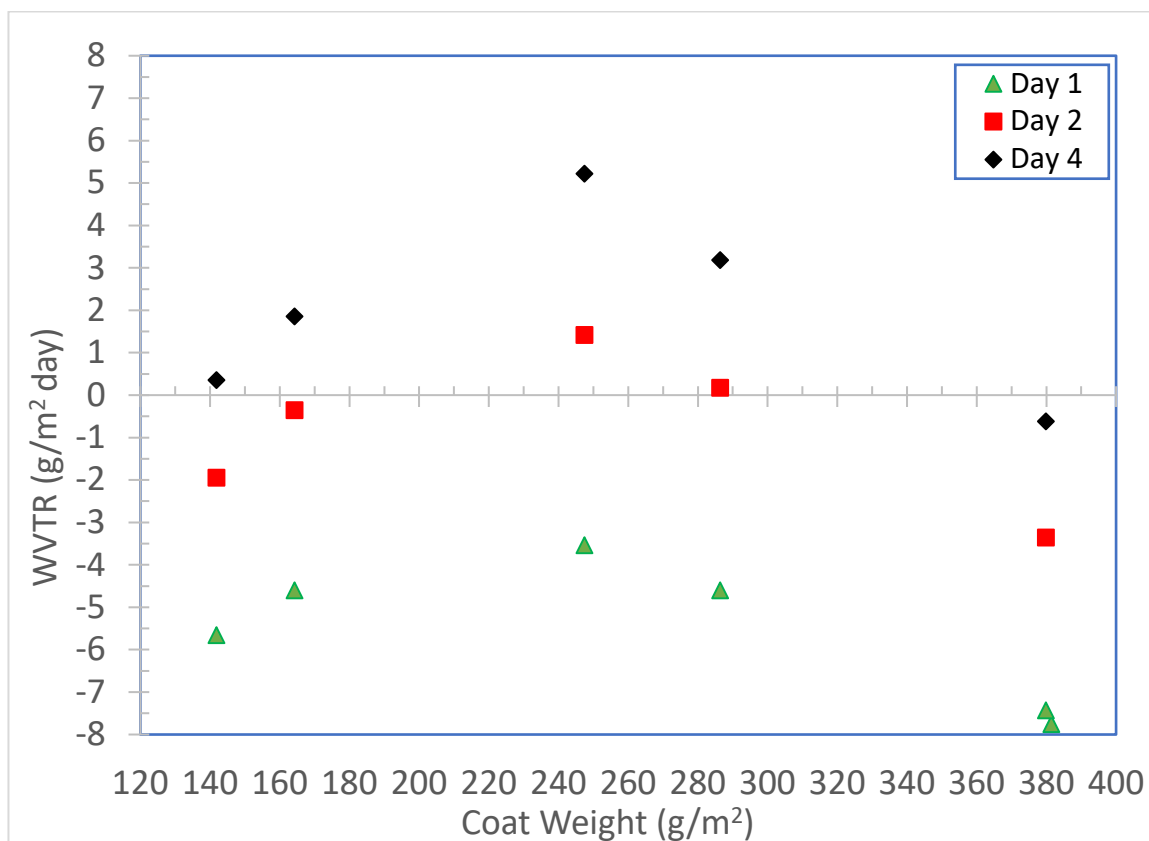


Figure 22. CNF Film covered with Polymer A on Paper A.

This result must come have come from the adsorption of moisture to the sample on the room-side surface of the coated paper, which increased the weight of the jar. Once again, these coatings must block moisture from leaving to such a small level that small gains of weight of the layer can be seen. As time went by, water vapor must have started to go up the layers of coating and paper and eventually pushed the moisture on top and left the jar, decreasing the weight (giving positive WVTRs). It is also important to notice that at the highest coat weight, even after four days, the WVTR was still negative, which meant that moisture was setting on the outside at a rate higher than the water vapor leaving. Compared to the data of latex only presented in Figure 18, the films only improved WVTRs very little, and they actually decreased air permeability, because of the air gaps that formed between the paper and the CNF film, results may be seen in Tables

19-21 in the Appendix. If laying the film completely flat on the paper is possible, the goal of a WVTR of $0.1 \left[\frac{g}{m^2 \cdot day} \right]$ might be reached with this combination as well.

WVTR test on a Sun Chips potato bag was also conducted over four days to see the performance. Detailed results are given in Tables 22-23 in the Appendix. WVTRs were excellent and meet the standards set by PepsiCo of $0.1 \left[\frac{g}{m^2 \cdot day} \right]$. Water vapor permeability (WVP) was calculated for all samples from the WVTR values as described in the procedure. Complete numerical comparison between WVTRs, WVPs, and air permeabilities are given in tables in the Appendix. WVP should be constant at different coat weights, because it is a property of the material. However, due to pin holes or other defects that occur at low coat weight, these values vary with coat weight as well.

CONCLUSION

After testing the different polymers, papers, pigments, and some combination, we have a better understanding of the material available. To summarize everything in terms of water vapor transmission rate (WVTR): Polymer A gave the best values of all the polymers. Paper A gave the best results at coat weights higher than 30 g/m². Mixing pigments with polymer made the paper extremely brittle, but improved the performance of Polymer C, even though it was still not as good as Polymer A alone. High coat weights of Polymer A, double layering of Polymer A, and CNF films covered in Polymer A give the most competitive results. Small negative WVTRs were obtained with these combinations, which most likely suggests that WVTR was quite small. This conclusion may be drawn based on the idea that the adsorption of moisture to the sample on the room-side surface of the coated paper, which increased the weight of the jar. Indicating that these coatings must block moisture from leaving to such a small level that small gains of weight of the layer can be seen. Conditioning the samples in the room for a few days before doing the WVTR test should allow the samples to come to some equilibrium water content. Further investigation of these negative values is recommended, but results so far are very promising.

With these promising results, we can have some hope of changing the world of snack packaging with these renewable, recyclable green materials.

PATH FORWARD

As mentioned before, this research project part of a three-year grant by the U.S. Endowment for Forestry and Communities (P3Nano). The presented work is part of year one of the project, which is mostly the bench scale experiments that include: coating CNF layers on papers, coating water barrier layers, and exploring different paper types. Since I am graduating, a PhD student, Mohammed Al-Gharrawi, will be continuing what is left of year one and future years of project.

Some of the things that need to be investigated in more depth include finding the right pigment to polymer ratio to produce a water barrier layer that is effective and cheap. Another thing would be finding the correct order of layering to prevent wrinkling and peeling. I believe it would be worth investigating using CNF films instead of paper as the base for coating. CNF gives excellent air permeability and replacing paper with CNF films could also reduce thickness drastically. Polymer B should also be tested a few more times, and its stickiness may be useful for sealing purposes on the edges. The project also has a potential of exploring not only potato chips bags, but all sort of snack packaging and paper cups.

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APPENDIX

WVTR = Water Vapor Transmission Rate

WVP = Water Vapor Permeability (of the coating, not accounting for the base paper)

VH# = Very high coat weight #

H# = High coat weight #

M# = Medium coat weight #

L# = Low coat weight #

C# = Control # (base-paper with no coating)

- Trials testing different polymers:

Table 1. Polymer A coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	78.473	Too Dense	1.061	7.97E-10
VH2	80.552	Too Dense	2.122	1.64E-09
VH3	65.221	Too Dense	1.415	8.84E-10
H1	27.803	Too Dense	9.549	2.54E-09
H2	22.866	Too Dense	6.013	1.32E-09
H3	26.244	Too Dense	6.366	1.60E-09
M1	12.213	Too Dense	16.623	1.94E-09
M2	13.772	Too Dense	22.989	3.03E-09
M3	13.512	Too Dense	19.099	2.47E-09
L1	7.795	Too Dense	50.576	3.78E-09
L2	7.795	Too Dense	95.493	7.13E-09
L3	8.575	Too Dense	134.751	1.11E-08
L4	7.795	Too Dense	101.505	7.58E-09
L5	6.236	Too Dense	75.333	4.50E-09
L6	5.457	Too Dense	62.247	3.25E-09
C1	0.000	280.1	400.363	0.00E+00

Table 2. Polymer B coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	147.072	Too Dense	4.308	6.25E-09
VH2	72.497	Too Dense	8.615	6.16E-09
VH3	66.780	Too Dense	9.295	6.13E-09
H1	38.457	Too Dense	15.643	5.94E-09
H2	38.197	Too Dense	13.830	5.21E-09
H3	33.000	Too Dense	15.417	5.02E-09
M1	26.504	Too Dense	21.765	5.69E-09
M2	19.229	Too Dense	23.805	4.52E-09
M3	25.205	Too Dense	24.259	6.03E-09
L1	19.229	Too Dense	45.570	8.65E-09
L2	20.788	Too Dense	48.291	9.91E-09
L3	12.473	Too Dense	38.542	4.74E-09
L4	10.394	28611.2	118.119	1.21E-08
L5	12.732	27058.3	118.119	1.48E-08
L6	13.252	Too Dense	70.282	9.19E-09
C1	0	240.5	424.867	0
C2	0	231.1	444.364	0
C3	0	282.1	444.591	0

Table 3. Polymer C coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	41.575	133.1	263.721	1.10E-07
VH2	35.599	1339.4	241.167	8.65E-08
VH3	39.756	460.6	251.931	1.01E-07
VH4	39.237	421.1	262.183	1.04E-07
H1	14.032	9631.8	83.037	1.17E-08
H2	14.291	9292.7	101.746	1.46E-08
H3	13.772	8749.6	94.570	1.31E-08
H4	13.772	7281.4	94.827	1.32E-08
M1	15.331	18006.8	54.077	8.35E-09
M2	12.732	22357.0	52.795	6.77E-09
M3	13.512	23510.4	57.409	7.81E-09
M4	15.591	14672.6	54.589	8.57E-09
L1	6.496	13486.9	108.154	7.08E-09
L2	10.394	17308.1	96.621	1.01E-08
L3	4.158	14888.4	91.239	3.82E-09
L4	8.055	8178.6	194.010	1.57E-08
L5	6.756	8428.8	193.754	1.32E-08
L6	7.795	8318.3	183.502	1.44E-08
C1	0.000	208.8	416.725	0
C2	0.000	217.6	427.232	0
C3	0.000	278.1	437.484	0

- Trials testing different paper types:

Table 4. Polymer A coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	78.473	Too Dense	1.061	7.97E-10
VH2	80.552	Too Dense	2.122	1.64E-09
VH3	65.221	Too Dense	1.415	8.84E-10
H1	27.803	Too Dense	9.549	2.54E-09
H2	22.866	Too Dense	6.013	1.32E-09
H3	26.244	Too Dense	6.366	1.60E-09
M1	12.213	Too Dense	16.623	1.94E-09
M2	13.772	Too Dense	22.989	3.03E-09
M3	13.512	Too Dense	19.099	2.47E-09
L1	7.795	Too Dense	50.576	3.78E-09
L2	7.795	Too Dense	95.493	7.13E-09
L3	8.575	Too Dense	134.751	1.11E-08
VL1	7.795	Too Dense	101.505	7.58E-09
VL2	6.236	Too Dense	75.333	4.50E-09
VL3	5.457	Too Dense	62.247	3.25E-09
C1	0.000	280.1	400.363	0.00E+00

Table 5. Polymer A coated on Paper B.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	71.890	Too Dense	4.421	3.04E-09
VH2	56.819	Too Dense	3.956	2.15E-09
VH3	64.615	Too Dense	4.654	2.88E-09
VH4	60.977	Too Dense	3.723	2.17E-09
H1	23.559	Too Dense	7.679	1.73E-09
H2	31.355	Too Dense	4.654	1.39E-09
H3	23.040	Too Dense	5.817	1.28E-09
H4	21.481	Too Dense	8.144	1.68E-09
M1	16.803	Too Dense	7.213	1.16E-09
M2	16.024	Too Dense	6.980	1.07E-09
M3	14.205	Too Dense	7.446	1.01E-09
M4	16.024	Too Dense	11.634	1.78E-09
L1	5.630	Too Dense	17.451	9.41E-10
L2	4.331	Too Dense	26.060	1.08E-09
L3	4.331	Too Dense	19.080	7.91E-10
L4	4.331	Too Dense	16.055	6.65E-10
C1	0.000	7909..1	364.381	0
C2	0.000	5094.2	371.827	0
C3	0.000	7353.5	369.733	0

Table 6. Polymer A coated on Paper C.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	71.544	Too Dense	4.571	3.13E-09
VH2	81.418	Too Dense	3.609	2.81E-09
VH3	57.252	Too Dense	4.812	2.64E-09
VH4	104.544	Too Dense	5.053	5.06E-09
H1	20.355	Too Dense	6.015	1.17E-09
H2	20.614	Too Dense	6.496	1.28E-09
H3	20.614	Too Dense	7.218	1.42E-09
H4	17.756	Too Dense	8.421	1.43E-09
M1	10.740	Too Dense	13.955	1.44E-09
M2	9.701	Too Dense	21.894	2.03E-09
M3	9.701	Too Dense	12.752	1.18E-09
M4	9.701	Too Dense	18.526	1.72E-09
L1	4.764	538.5	35.608	1.62E-09
L2	3.984	496.6	32.481	1.24E-09
L3	3.724	480.2	30.315	1.08E-09
L4	3.984	474.5	29.353	1.12E-09
C1	0.000	17.9	494.908	0
C2	0.000	17.9	515.599	0
C3	0.000	17.1	510.306	0

Table 7. Polymer A coated on Paper D.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	73.276	Too Dense	3.065	2.15E-09
VH2	55.607	Too Dense	3.301	1.76E-09
VH3	69.638	Too Dense	3.301	2.20E-09
VH4	50.150	Too Dense	3.537	1.70E-09
H1	17.150	Too Dense	8.017	1.32E-09
H2	18.969	Too Dense	5.895	1.07E-09
H3	18.969	Too Dense	4.716	8.57E-10
H4	19.488	Too Dense	7.781	1.45E-09
M1	6.756	Too Dense	14.619	9.46E-10
M2	8.575	Too Dense	8.960	7.36E-10
M3	8.055	Too Dense	11.553	8.91E-10
M4	7.016	Too Dense	19.334	1.30E-09
L1	8.055	538.5	124.730	9.62E-09
L2	6.756	496.6	123.787	8.01E-09
L3	7.016	480.2	126.145	8.48E-09
L4	7.535	474.5	119.307	8.61E-09
C1	0.000	17.9	555.510	0
C2	0.000	17.9	588.520	0
C3	0.000	17.1	622.237	0

Table 8. Polymer A coated on Paper E.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	Too Dense	45.473	6.366	2.77E-09
VH2	Too Dense	47.552	4.951	2.25E-09
VH3	Too Dense	68.599	4.480	2.94E-09
VH4	Too Dense	108.875	2.358	2.46E-09
H1	Too Dense	13.252	8.017	1.02E-09
H2	Too Dense	12.732	9.431	1.15E-09
H3	Too Dense	12.732	8.488	1.04E-09
H4	Too Dense	11.953	10.846	1.24E-09
M1	Too Dense	9.095	14.619	1.27E-09
M2	Too Dense	9.095	7.545	6.57E-10
M3	Too Dense	7.276	9.431	6.57E-10
M4	Too Dense	11.173	13.911	1.49E-09
L1	39803.4	8.315	27.351	2.18E-09
L2	Too Dense	4.158	24.993	9.95E-10
L3	36421.4	5.457	26.880	1.40E-09
L4	31242	4.158	29.709	1.18E-09
C1	20.7	0.000	433.373	0
C2	18.6	0.000	462.375	0

- Trials testing polymer and pigment mixture on Paper A:

Table 9. Polymer A mixed with Pigment A coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
H1	27.284	Too Dense	25.465	6.65E-09
H2	32.740	Too Dense	25.111	7.87E-09
H3	33.520	Too Dense	26.526	8.52E-09
H4	25.725	Too Dense	32.892	8.10E-09
M1	18.449	Too Dense	43.856	7.75E-09
M2	17.410	Too Dense	40.673	6.78E-09
M3	21.047	Too Dense	40.319	8.13E-09
M4	20.008	Too Dense	43.856	8.40E-09
L1	9.614	Too Dense	62.955	5.80E-09
L2	11.693	Too Dense	56.235	6.30E-09
L3	10.913	Too Dense	56.942	5.95E-09
L4	11.433	Too Dense	63.308	6.93E-09
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

Table 10. Polymer C mixed with Pigment A coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	123.166	Too Dense	10.610	1.25E-08
VH2	131.741	Too Dense	11.318	1.43E-08
VH3	163.962	Too Dense	12.732	2.00E-08
VH4	134.340	Too Dense	16.977	2.18E-08
H1	29.622	Too Dense	12.379	3.51E-09
H2	31.181	Too Dense	12.379	3.70E-09
H3	30.402	Too Dense	10.610	3.09E-09
H4	27.284	Too Dense	12.025	3.14E-09
M1	21.827	Too Dense	19.806	4.14E-09
M2	19.488	Too Dense	21.221	3.96E-09
M3	21.307	Too Dense	19.099	3.90E-09
M4	21.047	Too Dense	20.160	4.06E-09
L1	10.654	Too Dense	64.369	6.57E-09
L2	14.551	Too Dense	68.260	9.51E-09
L3	9.095	Too Dense	69.674	6.07E-09
L4	13.512	Too Dense	79.224	1.03E-08
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

Table 11. Polymer A mixed with Pigment A coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	53.008	Too Dense	75.687	3.84E-08
VH2	48.851	Too Dense	70.736	3.31E-08
VH3	51.709	Too Dense	72.150	3.57E-08
VH4	46.252	Too Dense	76.394	3.38E-08
H1	23.386	Too Dense	93.371	2.09E-08
H2	25.205	Too Dense	93.017	2.25E-08
H3	27.544	Too Dense	89.480	2.36E-08
H4	30.402	Too Dense	85.236	2.48E-08
M1	16.110	Too Dense	150.313	2.32E-08
M2	15.851	Too Dense	155.618	2.36E-08
M3	9.354	Too Dense	158.094	1.42E-08
M4	9.614	Too Dense	166.228	1.53E-08
L1	8.055	Too Dense	210.085	1.62E-08
L2	5.976	Too Dense	236.964	1.36E-08
L3	8.835	Too Dense	242.977	2.06E-08
L4	8.575	Too Dense	242.269	1.99E-08
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

Table 12. Polymer C mixed with Pigment B coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	56.386	Too Dense	122.726	6.63E-08
VH2	54.048	Too Dense	123.434	6.39E-08
VH3	58.985	Too Dense	121.311	6.85E-08
VH4	61.583	Too Dense	124.141	7.32E-08
H1	16.890	Too Dense	228.829	3.70E-08
H2	11.173	Too Dense	224.232	2.40E-08
H3	12.992	Too Dense	229.890	2.86E-08
H4	10.134	Too Dense	229.537	2.23E-08
M1	9.874	Too Dense	238.379	2.25E-08
M2	8.575	Too Dense	259.599	2.13E-08
M3	8.055	Too Dense	249.696	1.93E-08
M4	7.016	Too Dense	253.587	1.70E-08
L1	6.496	Too Dense	253.233	1.58E-08
L2	1.559	Too Dense	265.612	3.97E-09
L3	4.417	Too Dense	270.917	1.15E-08
L4	8.315	Too Dense	277.283	2.21E-08
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

- Trials testing double layers on Paper A:

Table 13. Double coating of Polymer A on Paper A (one layer on each side).

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	226.845	2763.2	-2.953	-6.41E-09
VH2	260.624	Too Dense	-2.476	-6.18E-09
VH3	290.247	Too Dense	-3.890	-1.08E-08
VH4	213.852	360.9	-2.122	-4.35E-09
H1	163.962	Too Dense	-1.768	-2.78E-09
H2	158.765	Too Dense	-1.768	-2.69E-09
H3	194.364	552.2	-2.476	-4.61E-09
H4	200.860	Too Dense	-2.476	-4.76E-09
M1	116.930	Too Dense	-3.890	-4.36E-09
M2	86.009	Too Dense	-3.537	-2.91E-09
M3	80.032	Too Dense	-1.415	-1.08E-09
M4	78.993	Too Dense	-1.768	-1.34E-09
L1	68.079	Too Dense	0.354	2.31E-10
L2	76.654	Too Dense	-0.707	-5.19E-10
L3	80.032	Too Dense	-1.415	-1.08E-09
L4	74.835	Too Dense	-0.707	-5.07E-10
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

Table 14. Heavy coating of Polymer A on Paper A (over 4 days) - Day 1.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	225.805	Too Dense	-4.244	-9.18E-09
VH2	239.577	Too Dense	-4.598	-1.05E-08
VH3	79.513	Too Dense	0.707	5.39E-10
VH4	114.851	Too Dense	-0.354	-3.89E-10
VH5	206.317	Too Dense	-3.537	-6.99E-09
VH6	216.970	Too Dense	-2.829	-5.88E-09

Table 15. Heavy coating of Polymer A on Paper A (over 4 days) - Day 2.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	225.805	Too Dense	-0.531	-1.14E-09
VH2	239.577	Too Dense	-0.531	-1.21E-09
VH3	79.513	Too Dense	3.360	2.56E-09
VH4	114.851	Too Dense	2.476	2.72E-10
VH5	206.317	Too Dense	1.415	2.80E-09
VH6	216.970	Too Dense	2.299	4.78E-09

Table 16. Heavy coating of Polymer A on Paper A (over 4 days) - Day 4.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	225.805	Too Dense	1.592	3.44E-09
VH2	239.577	Too Dense	1.768	4.06E-09
VH3	79.513	Too Dense	5.040	3.84E-09
VH4	114.851	Too Dense	4.067	4.47E-09
VH5	206.317	Too Dense	4.421	8.74E-09
VH6	216.970	Too Dense	5.305	1.10E-08

Table 17. Polymer A and CNF layers on Paper A (one layer on each side).

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	87.048	4453.4	10.334	8.62E-09
VH2	114.332	4664.5	3.890	4.26E-09
VH3	85.229	17156.6	9.903	8.08E-09
VH4	94.324	426.3	9.903	8.95E-09
H1	47.292	Too Dense	4.244	1.92E-09
H2	45.993	Too Dense	3.183	1.40E-09
H3	45.733	Too Dense	7.074	3.10E-09
H4	56.126	12950	3.890	2.09E-09
M1	39.756	Too Dense	8.842	3.37E-09
M2	57.426	30688	2.122	1.17E-09
M3	40.796	Too Dense	3.890	1.52E-09
M4	46.512	12008.4	9.549	4.25E-09
L1	19.488	Too Dense	83.822	1.56E-08
L2	23.126	Too Dense	97.969	2.17E-08
L3	20.788	Too Dense	98.676	1.96E-08
L4	23.386	Too Dense	72.150	1.62E-08
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

Table 18. Polymer C mixed with CNF coated on Paper A.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
VH1	164.482	4453.4	13.656	2.15E-08
VH2	170.718	4664.5	13.793	2.26E-08
VH3	156.686	17156.6	16.623	2.49E-08
VH4	160.844	426.3	15.915	2.45E-08
H1	130.962	Too Dense	18.391	2.31E-08
H2	130.182	Too Dense	18.745	2.34E-08
H3	131.222	Too Dense	19.099	2.40E-08
H4	133.560	12950	20.513	2.62E-08
M1	92.505	Too Dense	22.989	2.04E-08
M2	69.898	30688	24.757	1.66E-08
M3	81.072	Too Dense	24.050	1.87E-08
M4	82.890	12008.4	23.343	1.85E-08
L1	19.488	Too Dense	83.114	1.55E-08
L2	34.040	Too Dense	42.441	1.38E-08
L3	25.205	Too Dense	56.235	1.36E-08
L4	18.189	Too Dense	64.016	1.12E-08
C1	0.000	204.1	357.922	0
C2	0.000	214.2	371.715	0
C3	0.000	204	385.862	0
C3	0.000	163	374.545	0

Table 19. CNF film covered in Polymer A coated on Paper A (over 4 days) - Day 1.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
Film 1	381.452	2778.6	-7.750	-2.83E-08
Film 2	379.893	53	-7.427	-2.70E-08
Film 3	164.222	47.8	-4.598	-7.23E-09
Film 4	141.875	8371.5	-5.659	-7.69E-09
Film 5	247.476	896.8	-3.537	-8.38E-09
Film 6	286.349	15.4	-4.598	-1.26E-08

Table 20. CNF film covered in Polymer A coated on Paper A (over 4 days) - Day 2.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
Film 1	381.452	2778.6	-	-
Film 2	379.893	53	-3.360	-1.22E-08
Film 3	164.222	47.8	-0.354	-5.56E-10
Film 4	141.875	8371.5	-1.945	-2.64E-09
Film 5	247.476	896.8	1.415	3.35E-09
Film 6	286.349	15.4	0.177	4.85E-10

Table 21. CNF film covered in Polymer A coated on Paper A (over 4 days) - Day 4.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
Film 1	381.452	2778.6	-	-
Film 2	379.893	53	-0.619	-2.3E-09
Film 3	164.222	47.8	1.857	2.92E-09
Film 4	141.875	8371.5	0.354	4.81E-10
Film 5	247.476	896.8	5.217	1.24E-08
Film 6	286.349	15.4	3.183	8.73E-09

Table 22. Sun Chips Original Bag - Day 1.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
Chip 1	46.772	Too Dense	0.000	-
Chip 2	46.772	Too Dense	0.354	1.58E-10
Chip 3	47.032	Too Dense	0.000	-
Chip 4	47.552	Too Dense	0.707	3.22E-10
Chip 5	48.071	Too Dense	0.000	-

Table 23. Sun Chips Original Bag - Day 4.

Type	Coat Weight (g/m ²)	Air Permeability (Gurley sec)	WVTR (g/day*m ²)	WVP (g/m*day*Pa)
Chip 1	46.772	Too Dense	0.177	7.92E-11
Chip 2	46.772	Too Dense	0.265	1.19E-10
Chip 3	47.032	Too Dense	0.177	7.97E-11
Chip 4	47.552	Too Dense	0.354	1.61E-10
Chip 5	48.071	Too Dense	0.088	4.07E-11

AUTHOR'S BIOGRAPHY

Thoalfakar Saheb Alsaady was born in Baghdad, Iraq on February 13, 1996. He lived in Iraq for the first ten years of his life, through the war of 2003. In 2006, he moved to Egypt and lived there for three years. In Egypt, he lived in the island of Port Fouad, which is located in Asia, but he attended middle school in Africa, across the Suez Canal. He has been living in the United States since 2009, and became a United States citizen in 2014. He attended Deering High School in Portland, Maine. In his high school years, he led a team to a Novice Division Chess State Championship and graduated among the top 4% of his class. He is a recipient of Senator George J. Mitchell Scholarship and Fellowship. Thoalfakar is in the Honors College at the University of Maine and majoring in Chemical Engineering with a minor in Pre-Medical Studies. He is expecting to graduate from the University of Maine on May 12, 2018 with Magna cum laude. He is also pursuing a Six Sigma Green Belt Certification through a credited program at the University of Maine. Upon graduation, Thoalfakar plans on working in his field of study, Chemical Engineering. He is also considering pursuing a Master in Business Administration in the future after working in the field for few years.