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Measuring of Thermal Conductivity and Heat Capacity of Different Papers and Heat-sealing Process of Papers With a Polymer Layer

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MEASURING OF THERMAL CONDUCTIVITY AND HEAT CAPACITY OF DIFFERENT PAPERS AND HEAT-SEALING PROCESS OF PAPERS WITH A POLYMER LAYER

By

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A THESIS

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By Amenah Sabbar Khalaf
Dissertation Advisor: Dr. Douglas W. Bousfield

An Abstract of the Thesis Presented
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Packaging is an important and safe way to protect our food and other things through transportation, but the use of plastic based packaging is causing issues with plastic pollution. To replace plastic packaging with a paper-based option that has a barrier coating, good heat sealing will be needed to be a drop-in replacement. While much as been studied on the heat sealing of polymer layers, little has been reported around the heat sealing of paper that has a polymeric barrier coating.

Experiments were conducted to characterize the thermal conductivity and heat capacity of paper as a function of press pressure and paper thickness which provided parameters for the model for validation. A simple method to measure both the heat capacity and thermal conductivity was developed. Layers of paper were used to slow the response time of a thermal probe to obtain thermal conductivity as a function of press pressure. Values are reported for four different papers. A hot press was used to laminate polyethylene film to paper to determine the time and temperature needed to form a bond between surfaces. In addition, modeling of a time-dependent heat transfer process based on a finite element method was presented that shows the interaction of paper properties with the process parameters as time progresses. A simple expression was developed to predict the time and temperature needed to heat seal paper layers as a function paper properties and the polymer melting point.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

INTRODUCTION

1.1 Motivation

As inhabitance increases, more waste streams are produced. Most of the waste is from plastics. There are thousands of plastic materials thrown in the dump every month without recycling them into useful materials. As all the world is now looking for something more sustainable, degradable, and less harmful to the environment. Most oceans are now filled with unrecyclable plastic material even with all the legislation that the governments have applied to protect the environment and the animals. People usually look for something easy to use which is single-use plastic which also ends up on the ocean or in the dump. As we are looking for alternatives like using paper instead of plastic, which is a sustainable material and has more opportunities to use for multiple things like cups, dishes and bags, and other very useful things. But by using paper it is also a challenge to heat seal it without a plastic layer. Looking for less contamination for beverage and food packaging, especially after Coronavirus people are more conserved about their health and food. Using the paper will cover all the aspects that the customer and industry are looking at.

A 41.9% of the shareholders (more than 206 million shares) of McDonald's corporation supported a proposal filed by As You Sow that asked about providing a list of actions to reduce plastic pollution. The shareholders involved in the study represent more than $51 billion in market value Jaekel (2022).

1.2 Literature Review

The heat sealing process is of great interest for many industrial processes and applications. It requires applying heat to form a bond between the polymer and paper. Many literatures have focused on heat sealing that involved polymer to polymer system but heat sealing of polymer to paper has gained limited attention.

1.2.1 Heat Sealing Properties

Finding the optimum sealing parameters has gained more attention in the literature. Merabtene et al. (2022) used a polyethylene coating in flexible paper-based packaging materials varying grammages and thicknesses as well as using an oriented polypropylene and polyethylene laminate for a comparison between both systems. The results obtained showed that a strong seal was achieved at a temperature of 90 °C for the paper-based material and at 100 °C for the polypropylene and polyethylene laminate, both at a dwell time of 2 s. In addition to that, the paper-based material was able to handle temperatures up to 220 ^{o}C compared to the other system that shrank at 140 °C. For the paper-based material, pressure changes had no effect on the seal strength, the increased thickness however did increase the seal strength. The pressure used in the study was 3 bar.

Sealing conditions were found to highly influence the strength of the end-use products. Hauptmann et al. (2021) studied the effect of sealing conditions such as pressure, temperature, time, jaw pattern, and climate conditions (moisture) on Hot-Tack and Cold-Tack of polypropylene (OPP)-based polyolefin bonded with polyethylene (PE) or cast polypropylene (CPP) as well as two different barrier paper materials. The authors concluded that the temperature was the key parameter for the polyolefins while the sealing pressure, time, and moisture content in paper were the influencing parameters in the sealing of paper materials because heat transfer was improved as time and sealing pressure increased.

Meka et al. (1994) used a finite element analysis and experiments to study the influence of the interfacial temperature on the sealing of polyethylene films as time progresses. The effect of sealing process variables such as maximum temperature, dwell time, and pressure on the seal properties of polyethylene was investigated. The authors summarized that increasing pressure and dwell time at temperatures above the polymer's final melting point will have a negative impact on the seal's appearance because the sealing area would undergo material deformation.

Mihindukulasuriya et al. (2013) used heat transfer simulations to study the interface temperature during the heat sealing of two layers of linear low density polyethylene films (LLDPE) in the presence or absence of a contaminant liquid (water) at the interface. These models were validated with experiments and good agreement was found. The key finding is that in the case of water existing at the interface, the interface temperature decreases affecting the melting of LLDPE films.

Planes et al. (2011) compared the heat sealing properties of a single layer with that of the multilayers composed of one polyethylene (LDPE) layer and one or three polyethylene terephthalate layers coated with aluminum. They concluded that the single films showed poor performances compared to the multilayer films which showed improvement in the mechanical properties.

The relationship between surface properties and heat sealability was investigated using flame-treated low-density polyethylene (LDPE) coating Tuominen et al. (2013). The authors proved that the heat sealing properties of LDPE-coated paper were affected by the flame treatment method. This has been seen in the heat sealing temperature of LDPE coating which was decreased or doubled depending on the change in the equivalence ratio of flame treatment.

McPhail et al. (2003) used linear low-density polyethylene in the barrier film, Escal®, to study the mechanical properties through heat sealing of polymer to polymer layers. Here, the LLDPE layers were fixed in the middle between the two horizontal layers of the barrier film. The research aimed to find the optimum parameters that result in the best heat seal and high strength and integrity. Both tensile test and scanning electron microscopy were used to assess the seal strength. The authors concluded that the optimum temperature is 117 °C which is close to the melting temperature of the polymer of 118 °C. In addition to that, the optimum cooling time was 3.53 s and the maximum load achieved was 17.1 N.

The heat sealing parameters such as platen temperature and dwell time were investigated to look at their effect on a seal of a linear low-density polyethylene (LLDPE) Mueller et al. (1998). The sealing temperature ranged from 100 to 130 °C (polymer melting temperature is 130 °C) and the seal time ranged from 1 to 100,000 s. The T-peel configuration and scanning electron microscope were used to determine the seal strength. The strong and good seal was achieved with temperatures of 115 °C or higher. These results were attributed to the heterogeneous composition of the LLDPE used where at 115 °C an easy diffusion occurs across the interface by the lower molecular weight (more highly branched chains) followed by crystallization upon cooling and connections across the interface.

Theller (1989) investigated the parameters that influence seal strength during the heat sealing of flexible web materials. The sealing variables such as time, pressure, and temperature were studied using the low density polyethylene (LDPE) coextruded film and high density polyethylene/ethylene vinyl acetate-polybutylene (HDPE/EVA-PB) coextruded film. The results indicated that the dwell time and interface temperature were the key parameters in heat seal strength while pressure showed little influence.

Malsen et al. (2008) performed experiments to explore the mechanical properties of the heat seal joints formed from several multi-layered metalized films and aluminum-polymer laminates. The effect of the heat seal parameters such as temperature and time on the seal strength of tested films was examined. For the range of seal times and temperatures used in this study, the results showed that both temperature and time had no significant influence on seal strength.

Najarzadeh et al. (2014) focused on finding the optimum heat sealing process parameters for the monolayer linear low-density polyethylene (LLDPE) film. The effect of seal dwell time, temperature, and pressure on seal strength was studied. The surface roughness of the seal interface (seal microstructure) was looked at using the atomic force microscopy (AFM) technique. The results showed enhancement in seal strength as dwell time increased where there was a linear correlation between the seal strength and the square root of sealing time. The temperature changes showed an impact on seal strength as well. The strong impact of temperature and dwell time on the seal strength depends on the availability of heat in the interface of the films. Both temperature and dwell time followed the mechanism of crystal melting and interdiffusion while pressure affects seal strength through the wetting mechanism.

Merabtene et al. (2023) investigated the heat sealability of thermoplastic film (oriented polypropylene layer/polyethylene), dispersion-coated paper, and PE-coated paper based material. The results indicated that the thermoplastic film showed strong seal strength and leakproof ability compared to paper-based materials. Besides, both sealing temperature and dwell time were the key parameters affecting seal strength for paper-based material. The scanning electron microscopy showed fiber tears and full delamination for the paper-based materials where they increase as the paper grammage, thickness, and sealing temperature increase. Finally, the PE coated papers resulted in twice the seal strength than that of the dispersion coated paper. At a temperature of 130 ^{o}C and a dwell time of 0.5 s, the thermoplastic film showed seal strength that was four times stronger than the PE-coated paper and five times stronger than dispersion-coated paper. This study did not include any modeling or analysis of heat transfer.

1.2.2 Barrier Properties and Package Strength

Ponkamo (1995) discussed methods and techniques to enhance package strength and quality. These techniques included the addition of radial reinforcing glue which was found to increase package strength. These radial glue lines produce a stronger package by supporting the package during transfer and lifting and by preventing the wrapping material from peeling after a minor tear. Here, the two-ply laminate was replaced by the coated kraft wrapping material with a polyethylene barrier. The continuous heat sealing method was used in the new wrapping technique and is called heat-sealed wrapping method. The heat-sealed wrapping method is better than the conventional glue-based wrapping by the high levels of package tightness and appearance achieved.

Leminen et al. (2015) tried to find the best optimal imaging method to analyze the creases that are formed in the package during the press-forming process of polymer coated paperboard trays. Different microscopic imaging methods were used that were Scanning electrode microscopy (SEM), X-ray Microtomography, Optical light microscopy, and Polarized light microscopy. The authors concluded that light microscopic imaging was the most suitable, fastest, and most affordable method that detected creases and leakage.

Triantafillopoulos et al. (2020) viewed recent studies on more environmentally friendly and sustainable foodservice packaging products where, for example, paper-based coffee cups are favorable in replacement to plastic-coated paper cups made of polyethylene liners due to the ease of recyclable, compostable, biodegradable, and low cost. Many countries such as the UK, Australia, Canada, South Africa, China, India, Singapore, and Taiwan had agreed that within a five years timeline single-use fossil-based plastics, including PE-lined OOH paper coffee cups will not be used. Starbucks on the other hand put $10 million into the NextGen Cup Challenge project in 2018 which deals with allowing paper coffee cups to be used around the world. Other companies like McDonalds, Coca Cola, the Yum! Group, Wendy's, and Nestlé have joined the projects as well.

Zhao et al. (2011) reviewed theories about polymer adhesion to various surfaces. They found out that the paper strength can be affected by molecular adhesion and the viscoelasticity of the joints. The article also concluded that the weakest layer between the polymer-paper interaction is the paper itself because of its layered structure, so the adhesion is always limited by the cohesive strength of the paper.

Dudbridge (2009) studied the failure of poor heat seals created in the food packaging process and estimated the food waste that is generated by this failure. The integrity of heat seals was studied using data collected from different production sites generated by different food products and packaging machines. The results came out with around 24% of heat seals at risk of failure. This percentage is responsible for 480,000 tonnes of potential food waste. Failures in heat seals were mostly related to the products that had liquid and crumb components and that resulted in product contamination in the seal area. The author suggested generating a new method to reduce the incidence of inadequate seals and food waste.

Brewis et al. (2003) emphasized that polyolefins and olefinic copolymers are the most widely used materials however they showed bonding difficulties and needed surface pretreatment to increase the polarity.

Cha et al. (2004) collected a review on biopolymer-based antimicrobial packaging. The antimicrobial agents were added to control microbial growth in a food product by allowing an unfirm atmosphere inside the package. The amount of the preservatives used should be low so that little should have possible contact with the food. The review concluded that the film or coating technique is the most effective method despite the complicity associated with its application. Importantly, safety requirements are important to meet when adding these preservative agents to packaging technology.

The effects of filler addition such as clay and neutralizing agent on barrier properties of (barrier-coated paper) carboxylated styrene/butyl acrylate dispersion coatings were studied by Andersson et al. (2002). The impact of these fillers as well as drying conditions on the water vapour permeability, water absorption, oxygen permeability, and heat sealability was investigated. The temperature, time, and pressure were selected carefully to seal the coated strips while the seal strength was assessed by the peak load necessary to break the joints. The tests showed that the failure mode was mainly cohesive with fiber tear occurring as strips peeled away.

Andersson et al. (2002) investigated the heat sealability of barrier-coated paper. The sealability was tested for latex dispersions such as carboxylated applied into the three-ply base paper as well as for neutralizing agents such as NH3 and NaOH. The Y-peel force on sealed material was used to assess the seal strength besides using scanning electron microscopy and electron spectroscopy for chemical analysis methods for interpretation. The carboxylation showed good barrier properties and heat sealability. The addition of surfactant on the coating surface resulted in poor adhesion between sealing surfaces. This work did not include any modeling or analysis of heat transfer.

Poulose et al. (2022) used potato fruit juice (PFJ) as a barrier coating material on the paperboard. The PFJ was first coated on the paperboard, then on the top of the PFJ layer, the paperboard was extrusion-coated with a layer of poly(lactic acid) (PLA) or a blend of PLA and poly(butylene adipate terephthalate) (PBAT). The water vapor transmission rate was reduced for the paperboard coated with a PFJ layer and the extrusion-coated layer compared to the uncoated one. Adding the PFJ layer between the paperboard and the extrusion-coated layer reduced the oxygen permeability. Moreover, there was a significant improvement in the grease resistance of the paperboard with the addition of these multilayer coatings. The combination of PLA- and PFJ-coated samples gave good barrier properties while PFJ with PLA/PBAT layers resulted in better adhesion and heat-sealing properties.

1.2.3 Thermal Conductivity Measurements

The thermal conductivity of a material is important to understand the heat transfer process, especially conductive heat transfer. Guérin et al. (2001) developed a method to measure the thermal conductivity of the fibrous network, polymers, and model coating layers as well as handsheets of different kinds of pulps or blends of pulps. The thermal conductivity was measured in a device in which a sample was laid on a hot plate maintained at a certain temperature and then a mass with a lower temperature was put in contact with the sample. As time progresses, the temperature will transfer into the mass depending on the thermal conductivity of the sample until reaching the temperature of the hot plate. The thermal conductivity was measured using three steps. First, the time of response was calculated for each thickness of the pile (and thus a thermal resistance) based on temperature measurements. Next, a straight line was fitted between the thermal resistances and the thickness of the piles. Finally, the thermal contact resistance was calculated from the intercept of this straight line with the y-axis while the material intrinsic thermal conductivity was calculated from the slope of this regression straight line. The pressure applied to the sample was about 100 kPa and the heated plate temperature was between 40 to 45 °C while the initial temperature of the mass was about 5 °C lower. The results showed weak variations in the thermal conductivity with the thickness of the LWC paper and its uncoated base paper. The limitations of this method are that the thermal conductivity should be less than 1.5 W/m K for valid measurements, the temperature cannot vary on a large range of values, and finally, pressure variations cannot be made using the device.

Sergiy et al. (2012) measured thermal conductivity and heat capacity for several commercially available copy paper samples. The operating temperature was varied to look at temperature’s influence on the properties. Here, steady-state measurements of the temperature difference were used for thermal contact resistance and thermal conductivity measurements while the transient temperatures recorded during heat-up and cool-down periods were used for the specific heat determination. The thermal conductivity was measured in the following steps. The paper sheet was placed on a hot plate. The heat then flowed in the vertical direction through the sample to the cooling unit. The temperatures of both sides of the paper sheet were measured as a function of time. The total resistance for paper stacks of different stack thicknesses was measured and found to be a straight line. The slope of this line represented the thermal conductivity of the sheets while the intercept represented the thermal contact resistance (with the metal) of both sides of the sheet. The time required to reach steady temperature distributions was 3 h. One key finding here is that the thermal conductivity and heat capacity are strongly controlled by the sheet density and less by the filler contents and the nature of the fibers. For example, a lower density sheet is having a lower thermal conductivity and vice versa. Another key finding is that the thermal conductivity increases as the number of sheets (thickness) in the stack increases because the resistance is linearly increased as the thickness increases. Moreover, the thermal conductivity increases as the temperature increases and is best described by a second-degree polynomial in temperature.

Kerr et al. (2009) measured the thermal conductivity of a coated paper substrate consisting of clay and calcium carbonates as its pigments, and styrene–butadiene (SB) latex and starch as its binders. A hot disk system was used to measure temperature increase as a function of time. In this system, a thin film probe is sandwiched between the two paper sheets and the two high-conducting stainless steel blocks where it serves as a heat source and as a temperature sensor. The voltage drop across the probe was monitored where it is directly proportional to the temperature increase of the sensor. The resistance of the probe was calculated from the initial resistance of the probe, the thermal coefficient of resistivity, and time. Finally, a plot of the total temperature increase versus time was used to calculate the thermal conductivity of the paper sheet. It was found that the increase in the coating mass leads to an increase in the thermal conductivity. However, the opposite was seen with the toner gloss and toner density which decreased as the thermal conductivity increased. As a result of that, the thermal conductivity of a paper substrate should be low in order to obtain high printing efficiency.

Kerekes (1979) developed a method to measure the thermal conductivity and contact resistance of paper. The surface temperature of the paper was measured with an infrared backscatter thermometer while in contact with a hot roll for a known time. Some theoretical equations for dynamic heat transfer were used to predict the temperature rise in the paper and as a sequence the thermal conductivity of the paper when all other parameters are well known. The results indicated an increase in the thermal conductivity upon calendering while the contact resistance experienced a decrease. Moreover, the heat transfer to paper was substantially improved because of calendering due to the reduction in the contact resistance of the web. In addition, the reduction in the web thickness and the increment in the thermal conductivity because of web densification both contribute substantially.

Gerstner et al. (2009) simulated the relationships between the thermal conductivity and the coating structure of calcium carbonate coatings. The thermal diffusivity and conductivity were also measured experimentally using a series of heated pigment tablets. The samples used were having coatings in the range of 0–25 pph of styrene acrylate latex and calcium carbonate. The system used to measure the thermal diffusivity had an insulted heating plate, thermocouples on top and bottom of the tablet connected via an extension cable to the interface of the data acquisition system, and a 7 kg weight at the top of the tablet. The thermal diffusivity was then calculated when the numerical solution of the heat diffusion equation matched the experimental measured topside temperature response. The results indicated that the thermal diffusivity and conductivity showed a maximum at binder concentration of 8 wt%. The mercury porosimetry results showed improvement in the connectivity (thermal conductivity) at lower binder levels and showed a reduction in the thermal conductivity at higher binder levels.

Gray-Stuart et al. (2019) measured the overall effective thermal conductivity of papers composed of corrugated fibreboard. The paper thermal conductivities measured in the lab were used in the finite element models (COMSOL Multiphysics 4.4) of heat transfer in corrugated fibreboard for validation purposes. The thermal conductivity was measured as follows. A stack of paper sheets was sandwiched between two polystyrene insulating layers. A 10 kg weight was placed on top of the polystyrene insulating layer to ensure good contact. A transient hot-wire heat source was used to heat the stack with transient temperature being measured with thermocouples connected to a Measurement Computing Data Logger. The authors modified the transient hot-wire method done by Imakoma et al. (2000) to measure the thermal conductivity of paper in all directions. The experimental results showed that the paper was highly anisotropic and the thermal conductivity was much larger in the machine and cross machine directions than in the thickness direction. The modeling results on the other hand showed good agreement with the experimental ones. Finally, it was found that the majority of heat was transferred through the fluted medium and the high thermal conductivity in the machine direction can be used to facilitate this process.

1.2.4 Heat Sealing of Paper and Polymer

Some other limited literature has focused on the polymer to polymer adhesion and paper-to-polymer adhesion. For instance, Aghkand et al. (2018) used COMSOL Multiphysics to simulate heat transfer and look at the effect of different process parameters and material properties on the interface temperature in a heat sealing process of multilayer polymeric films. The authors also performed some experimental works for comparison. The article focused on studying temperature variations and how these change in temperate affected the thermal conductivity, specific heat, and density of polymers. The process of measuring the interface temperature was done using a fine thermocouple. The modeling results were in very good agreement with the experimental data for the very wide range of temperatures used in this study. It was proved that the crystallinity of the sealant layer was the critical parameter in determining the dwell time required to get the desired seal strength. Moreover, increasing the polymer thickness to 130 μm resulted in an increase in the dwell time to 1.8 s. Furthermore, increasing the sealing pressure to 2.7 N/mm^2 showed no impact on the interface temperature evolution.

Hurnanen (2016) studied the heat sealing of paper with polymer film using hot-bar sealing. The relationship between the material properties and the heat sealing process was investigated. For example, the influence of seal time, temperature, and pressure on seal strength and peel characteristics of sealed materials was studied. The one key finding in this thesis is that the temperature was having a large impact on seal strength. The sealing time was having the same effect as the temperature on the seal strength where longer times were required to transfer more heat to the polymer film and melt it. Another key finding is that the increased sealing pressure did not influence the seal strength. It was also found that the increased pressure had an impact on the peel characteristics because more pressure could compact the surface of the paper so that the molten polymer will not penetrate valleys and voids of the paper's surface resulting in a smaller fiber amount index. At low pressure, on the other hand, the heat will not transfer easily to the polymer film due to the materials not being in as close contact. The seal strength was high at the optimum time, temperature, and pressure and any increase in these parameters will not lead to an increase in the seal strength. The sealing time of 3 s, pressure of 0.7 MPa, and temperature of 170 ^{o}C resulted in high seal strength because the penetration of polymer into the paper resulted in mechanical interlocking. The relationship between seal strength and fiber amount index was not straightforward.

1.3 Structure of Thesis

The structure of this thesis is discussed as follows. Chapter 1 presents past works as well as the motivation behind this thesis. Chapter 2 contains step by step on how to experimentally measure paper heat capacity, thermal conductivity as well as thermal diffusivity for different types of paper and layers. This chapter also includes experimental results obtained from pressing a polyethylene film at various temperatures and times against a paper of interest. The mechanical tester is also used to measure the final bond strength. Chapter 3 shows the modeling set-up, results, and discussions for unsteady non-isothermal heat transfer in paper and paper-polymer-paper systems. Chapter 4 concludes the key observations and contains recommendations for future considerations.

EXPERIMENTS

2.1 Introduction

Experiments were conducted first to characterize the thermal conductivity and heat capacity of four different papers as a function of press pressure and paper thickness. Second, low-density polyethylene (LDPE) films from (McMaster-Carr, Princeton, NJ, USA) with a thickness of 100 μm was laminated to paper using a hot press changing paper temperature and press time. The papers used were having thicknesses of 240 μm and 430 μm to cover the whole range of paper thicknesses used in this study. Finally, the bonded samples were taken to the mechanical tester (model 5564, INSTRON, Norwood, MA, USA) to measure the final bond strength.

2.2 Materials and Methods

Some simple tests were developed in the laboratory to measure the thermal properties of different papers. Figure 1 shows the steps followed to calculate the heat capacity, density, thermal diffusivity, and thermal conductivity of paper in the laboratory. The four papers that were selected for this study were classified based on their thicknesses as 240 μm, 308 μm, 383 μm, and 430 μm, respectively. Each one of these papers was tested for thermal properties as a stack of 4 layers, 9 layers, and 14 layers on each side. In the following sections, the processes of measuring heat capacity, density, thermal diffusivity, and thermal conductivity of different types and layers of paper are discussed in detail.

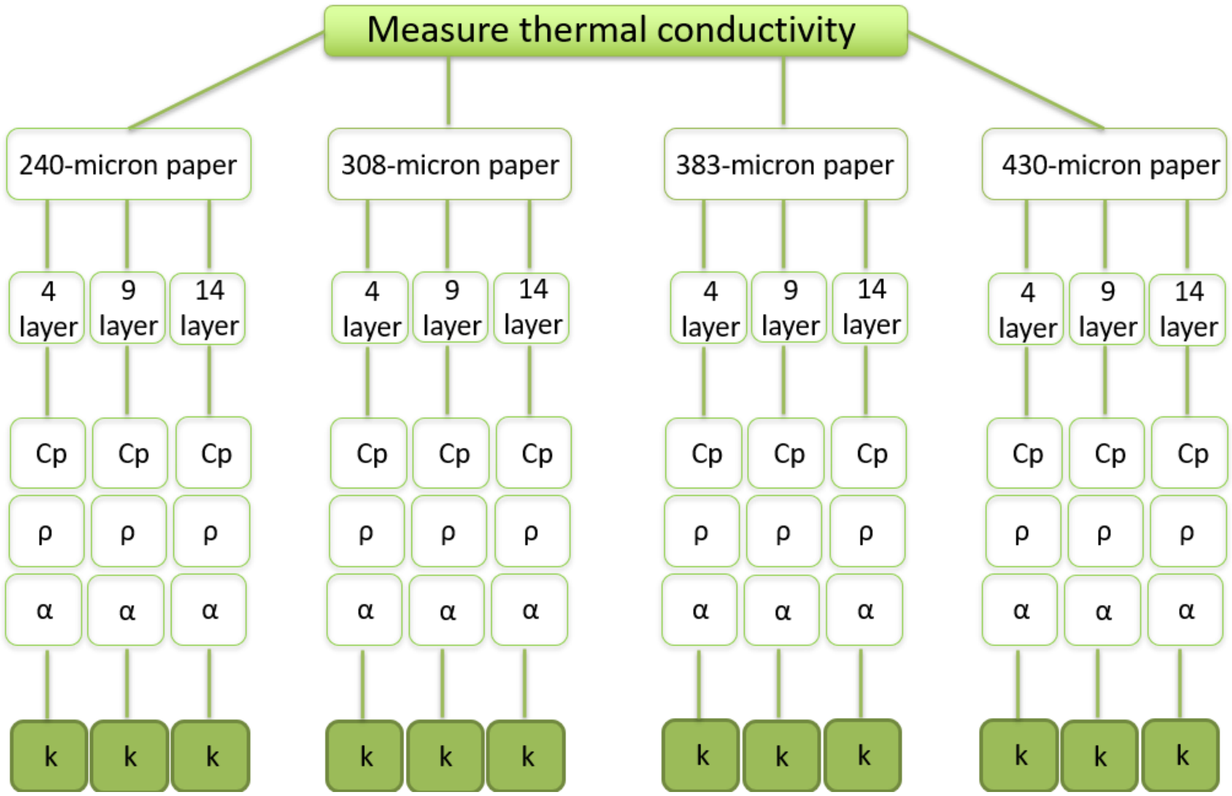

Figure 1. Scheme to measure the thermal properties of different papers.

2.2.1 Measurement of Paper Heat Capacity

One of the key parameters that influence the heat transfer process is the heat capacity of the paper. The method developed here is a simple, low-cost, and easy method to measure the heat capacity of paper. Before measurements, all paper samples were cut into 101.6 × 101.6 mm squares. The weight of each sample was recorded. Besides, a known amount of water was used. In addition, the temperatures of paper and water were recorded. The process of measuring the heat capacity of paper included first heating the paper samples to 150 ºC in the oven for 20 min. After that, the heated papers were soaked in water that was initially near room temperature. Then, the temperature of the water was recorded after contact with the heated papers. Finally, the energy balance equation was used to calculate the heat capacity. Figure 2 illustrates the process while Equation 1 is used to calculate the heat capacities of the four different papers.

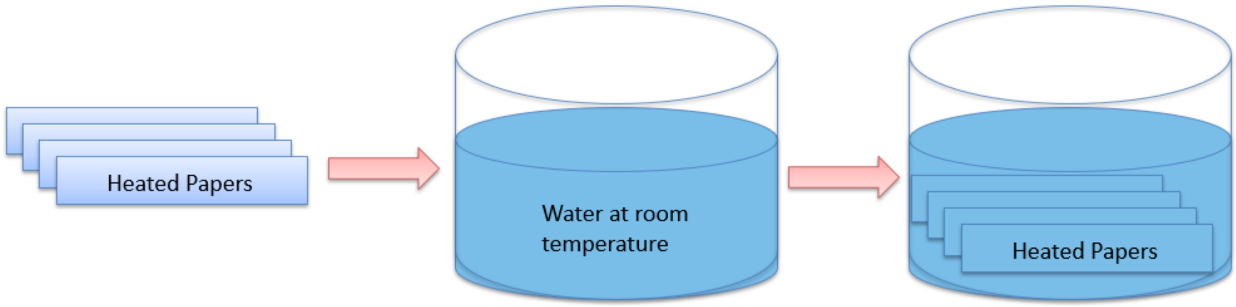

Figure 2. A scheme to capture the whole process.

Equation 1 is based on energy balance between paper and water as

$$CP_{paper} = \frac{W_w \, CP_w (T_{wa} - T_{wb})}{W_p \, (T_p - T_{wb})} \qquad (1)$$

Where W_p is the weight of paper, W_w is the weight of water, CP_w is the heat capacity of water, CP_{paper} is the heat capacity of paper, T_p is the temperature of the heated papers, T_{wb} is the initial temperature of the water, and T_{wa} is the increase in water temperature after contact.

2.2.2 Measurement of Paper Thermal Conductivity

Thermal conductivity is the most important parameter in understanding heat transfer between the paper and the heated source. Here, two different methods were used to measure the thermal conductivity of paper, the hot press method and the hot plate method. In both methods, the increase in the paper temperature as a function of time was recorded.

The carver press consists of an upper fixed platen and a lower moving platen. Both platens can be heated to the desired temperature and pressure can be fixed to the desired value as well. Here, the pressure was fixed to 0.1 MPa and 0.4 MPa while the temperature was fixed to 200 ^{o}C. During measurements, 101.6 × 101.6 mm squares of paper samples were put onto the lower platen with a thermocouple that is installed at the center between the samples. The thermocouple is connected to a thermometer that measures the temperature as a function of time. In this case, the heat will transfer from the hot platens into the papers that were initially at room temperature and the thermocouple will read that as time progresses. Figure 3 captures this method using four papers on each side of the platens as an example. Besides the four papers, nine papers and fourteen papers were tested as well. Lower number of paper layers results in a rapid temperature response that is difficult to analyze.

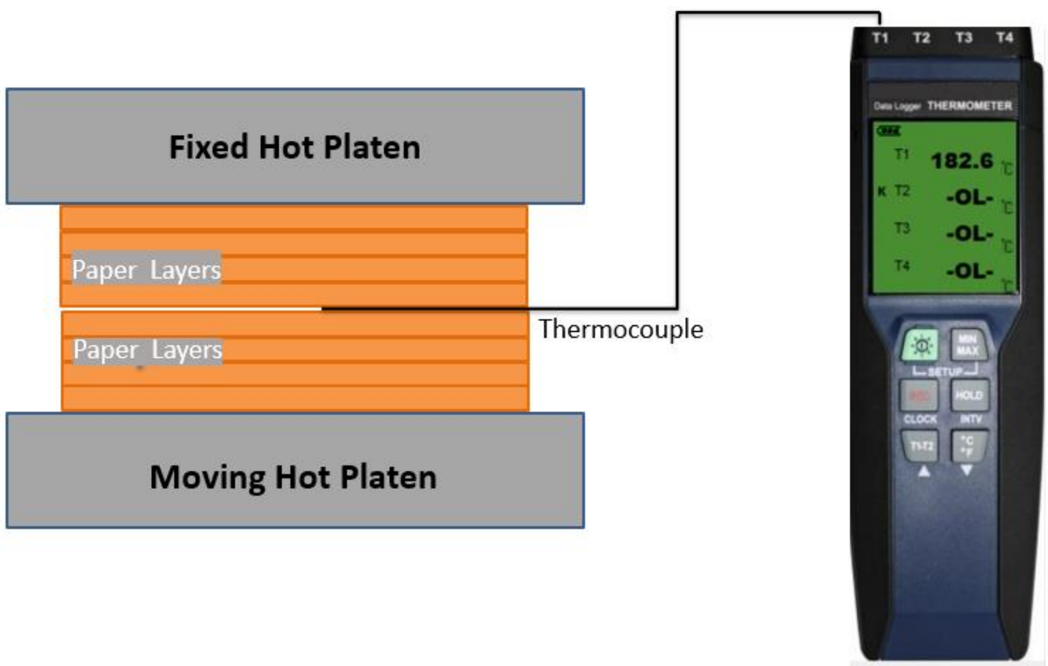

Figure 3. Measuring temperature increase as a function of time in four layers of a paper using the carver press method.

The hot plate method is similar to the carver press method in which both methods involve hot pressing, however, with the hot plate method the temperature was fixed to 150 ^{o}C, and paper samples were all insulated and put onto the heated plate with a one paper layer remained at the top. In addition, this method uses a weight at the top of the layers to generate force and good contact. Figure 4 shows the hot plate method.

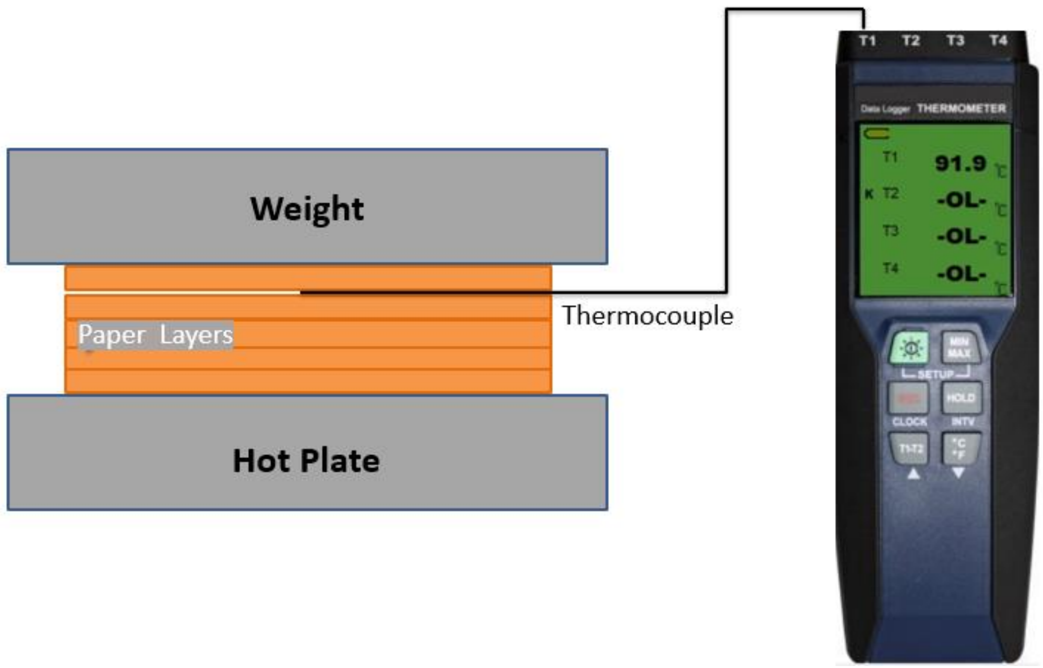

Figure 4. Measuring temperature increase as a function of time in four layers of a paper using the hot plate method.

The density of each paper layer was calculated by dividing the basis weight of the paper by the thickness of the same paper as

$$\rho = \frac{Basis\ weight}{Thickness} \tag{2}$$

The thermal diffusivity of each paper layer was measured for both methods as follows. Six different points were taken from the graph that shows the temperature as a function of time as in Figure 5. These random points should be taken from the curve before steady state temperature reaches.

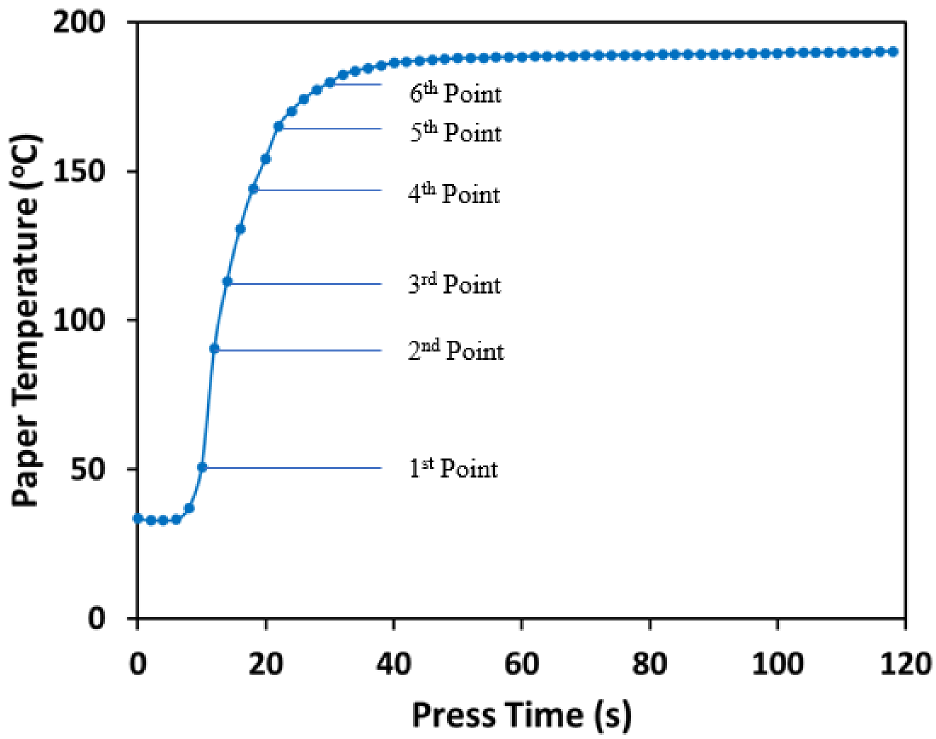

Figure 5. Paper temperature as a function of press time for the four-layers 240 µm paper type pressed at 0.4 MPa in the carver press method.

For each paper type and layer as well as for press and hot plate methods, a similar temperature-time curve was obtained which was used to calculate the thermal diffusivity of paper. The dynamic temperature at the center is given by standard text books such as Bird, Steward, and Lightfoot (1959) as

$$\frac{T_1 - T}{T_1 - T_0} = 2 \sum_{n=0}^{\infty} \left(\frac{(-1)^n}{\left(n + \frac{1}{2}\right) \pi} \, exp\left(-\left(n + \frac{1}{2}\right)^2 \pi^2 \, \alpha \, \frac{t}{b^2} \right) \right) \tag{3}$$

Where T_1 is the temperature of the hot source, T_o is the initial temperature of the paper, T is the temperature change with time, n is the number of terms, α is the thermal diffusivity of paper, t is time, and b is the thickness of the paper stack. Often, good results are obtained with three to five terms.

There is another way to calculate the thermal diffusivity of the paper using temperature profiles for unsteady-state heat conductions in a slab of finite thickness as shown in Figure 6. Figure 6 is the full solution of the dynamic analysis of heat transfer in a slab. In this figure, the y-axis ($T-T_o$ / T_1-T_o) can be calculated for different T_1, then by inserting these values into the y-axis and dragging straight lines towards the right-hand side of the figure, the values of τ are estimated. Finally, the equation located at the top left side of the figure is used to calculate the thermal diffusivity of the paper as

$$\frac{\alpha\, t}{b^2} = \tau \qquad (4)$$

Where τ is the dimensionless time and represents the curves inside the figure. It is important to point out that both Equations 3 and 4 gave similar results.

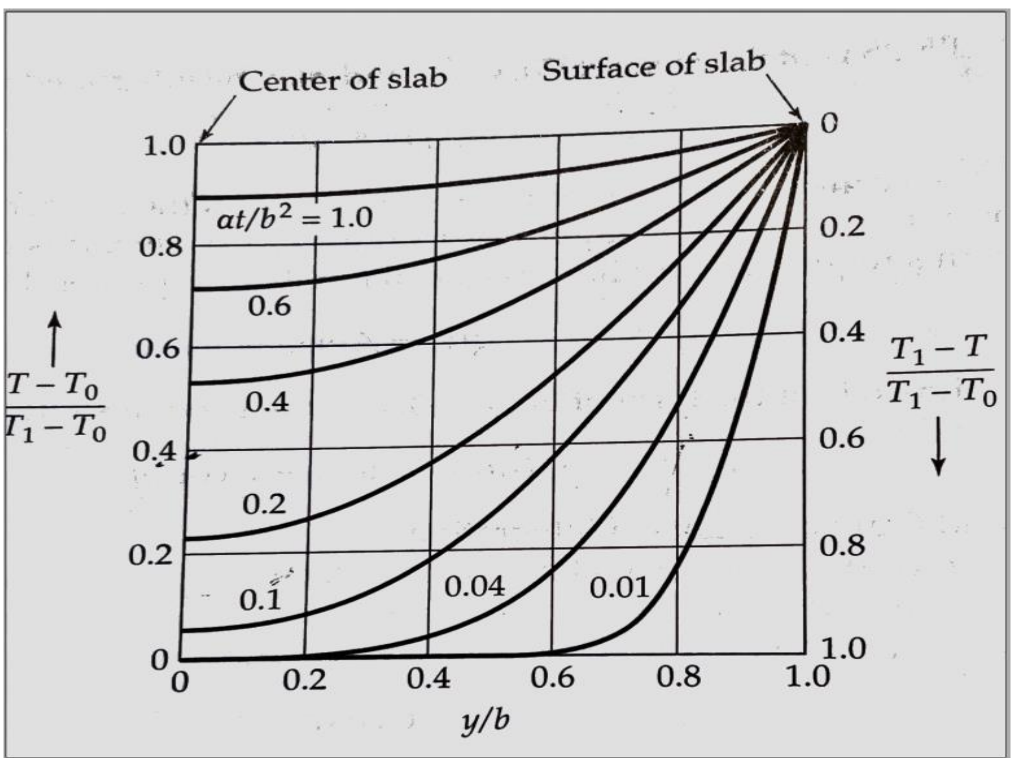

Figure 6. Temperature profiles for unsteady state heat conductions in a slab of finite thickness b. (H. S. Carslaw and J. C. Jaeger, conduction of heat in solids, 2nd edition, Oxford university press 1959, p. 101).

After calculating the necessary parameters, the thermal conductivity of all papers is calculated as an average of six points as

$$k = \rho\, \alpha\, CP_{paper} \qquad (5)$$

2.2.3 Temperature-Time Relationship

Figure 6 above is used to draw a relationship between time and paper temperature. Here, the dimensionless time (τ) is plotted against the dimensionless temperature ($T\text{-}T_o$ / $T_1\text{-}T_o$) as shown in Figure 7 below. The paper temperature increases with time and is best described by a second-degree polynomial in time.

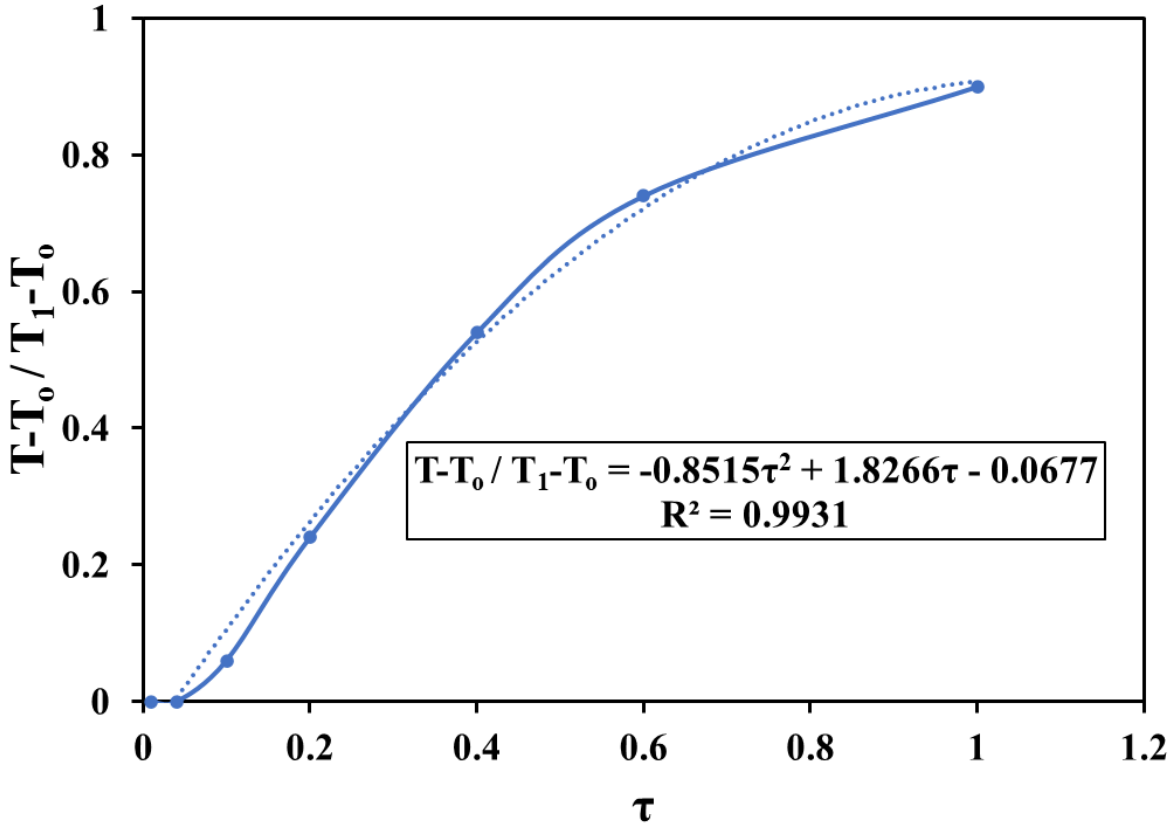

Figure 7. Temperature of the paper as a function of the press time.

By using Mathcad Prime 8 to solve the second-degree polynomial equation and assuming that the paper temperature is reaching half of the platen temperature (0.5) or at the platen temperature itself (0.9), then τ can be calculated for both cases using solve block as in Figure 8 below.

Guess Values

$$\tau_{0.5} := 0.2 \qquad \tau_{0.9} := 0.7$$

Constraints

$$-0.8515 \cdot \tau_{0.5}^{\ 2} + 1.8266 \cdot \tau_{0.5} - 0.0677 = 0.5 \qquad -0.8515 \cdot \tau_{0.9}^{\ 2} + 1.8266 \cdot \tau_{0.9} - 0.0677 = 0.9$$

Solver

$$\tau_{0.5} := \mathbf{find}\left(\tau_{0.5}\right) = 0.377 \qquad \tau_{0.9} := \mathbf{find}\left(\tau_{0.9}\right) = 0.954$$

Figure 8. Mathcad Prime 8 to solve the second-degree polynomial equation.

Now Equation 4 can be used with other parameters known to calculate the time required for the paper to reach half of the platen temperature or the platen temperature itself as in Figure 9 below.

$$\alpha_{240} := 1.821 \cdot 10^{-8} \cdot \frac{m^2}{s} \qquad \alpha_{430} := 3.305 \cdot 10^{-8} \cdot \frac{m^2}{s}$$

$$b_{240} := 4 \cdot 240 \cdot \mu m \qquad b_{430} := 4 \cdot 430 \cdot \mu m$$

$$t_{0.5} := \frac{b_{240}^{\ 2} \cdot \tau_{0.5}}{\alpha_{240}} = 19.084\ s \qquad t_{0.5} := \frac{b_{430}^{\ 2} \cdot \tau_{0.5}}{\alpha_{430}} = 33.754\ s$$

$$t_{0.9} := \frac{b_{240}^{\ 2} \cdot \tau_{0.9}}{\alpha_{240}} = 48.303\ s \qquad t_{0.9} := \frac{b_{430}^{\ 2} \cdot \tau_{0.9}}{\alpha_{430}} = 85.434\ s$$

Figure 9. Time required for two different papers of four layers each to reach half of the platen temperature or the platen temperature itself using 0.4 MPa press pressure in carver press method.

This method is simple and useful where the time required for the paper to reach any temperature can be calculated for any paper and the only parameters that need to be known are both the thickness and the thermal diffusivity. Figure 9 shows that less times are needed to heat the 240 µm paper and higher times are required for the 430 µm paper.

2.2.4 Other Paper Properties

The paper samples were also characterized in terms of porosity, thickness, basis weight, and permeability. The thickness of the sample was measured with a digital micrometer (Marathon) while the weight of the sample was measured with a Mettler Toledo Analytical Balance (AL204).

The Gurley porosity test was used to measure the air permeability of papers applying the TAPPI standard. This test required recording the time for an air volume of 100 cm^3 to flow through a paper area of 645 mm^2 with a pressure drop of 1.2 kPa. The permeability of the paper is calculated from Darcy's law as

$$K = \frac{\mu \, \delta \, V}{\Delta P \, A \, t_g} \qquad (6)$$

Where K is permeability, µ is the viscosity of air, δ is the thickness of paper, V is the volume of air, ΔP is pressure drop, A is the area for flow, and t_g is Gurley time.

The void fraction of a paper was measured with the silicone oil absorption method. Paper samples were soaked in 50 ml of silicone oil (Sigma-Aldrich, 0.1 Pa s) for six hours to be fully saturated. The samples were then removed from the oil and gently wiped to remove any surface oil. Finally, the samples were weighed to determine the mass of oil transferred into the pores. From this mass, the porosity of the paper is measured as

$$\varepsilon = \frac{W_2 - W_1}{A\ \delta\ \rho_{So}} \tag{7}$$

Where W_2 is the paper weight after oil soaking, W_1 is the initial paper weight, A is the paper area, δ is the paper thickness, and ρ_{so} is the silicone oil density which is 0.97 g/ml.

2.2.5 Heat Sealing of Polymer Films into Different Papers

Experimentally, a low-density polyethylene (LDPE) film of 101.6 × 101.6 mm square with a thickness of 100 µm was pressed in a carver (Model C, Carver USA) against two sheets of paper of 101.6 × 101.6 mm square and thicknesses of 240 µm and 430 µm. The pressing process was done at 1, 10, 30, and 60 s press times, 0.4 MPa press pressure, and 150, 200, and 225 °C paper temperatures. The 0.4 MPa pressure was used after accounting for the cross-sectional area of the carver cylinder and the paper sample. The heat sealing process is shown in Figure 10.

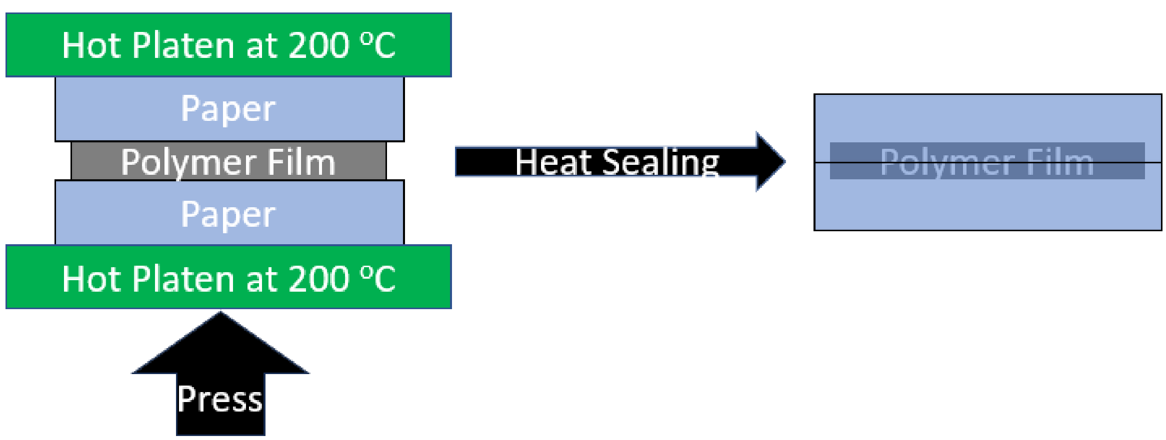

Figure 10. Heat sealing process using carver press method.

2.2.6 Tensile Test

A mechanical tester (model 5564, INSTRON, Norwood, MA, USA) was used to measure the force needed to peel samples apart. Samples peeled were 25.4 mm wide and 101.6 mm long. One side of the sample was clamped into the grip of the mechanical tester while the other side was fixed to a peel wheel with tape. Figure 11 shows an image of the peeling test. The peeling occurs at 90° and at a rate of 50.8 mm/min where both force and displacement are recorded.

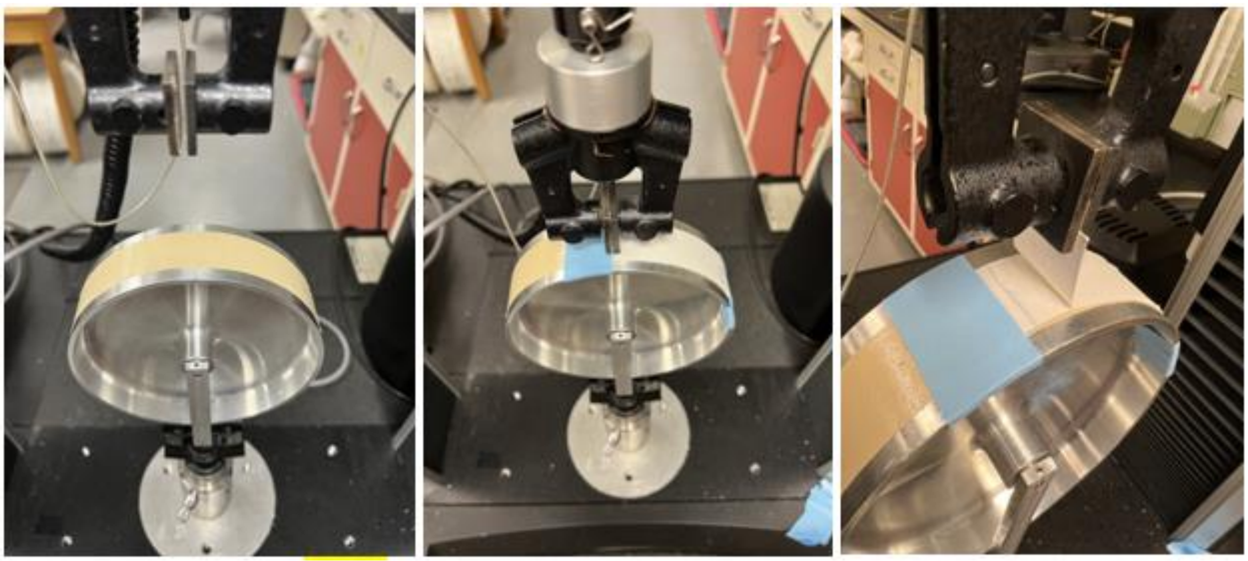

Figure 11. Peeling test to separate two glued samples.

RESULTS AND DISCUSSIONS

3.1 Thermal Properties of Different Papers

The values of thermal diffusivity and thermal conductivity in all tables below were calculated as an average of six different values of temperatures and times taken at the same run. The values of heat capacity and density of papers were reported in the caption of each table. Table 1 shows the results for the 240 μm paper at 4, 9, and 14 layers using the hot plate method and the carver press method at two different press pressures. From the results, it can be seen that as the paper layers increased, the values of thermal conductivity increased as well. This increase was not significant for the hot plate method; however, it showed an impact on the results for the carver press method where the thermal conductivity values ranged from 0.025-0.093 W/m K for the low and high press pressures. It is important to mention that for the 4 layers of paper, the thermal conductivity values were close regardless of press methods and pressures.

Table 1. Thermal diffusivity and thermal conductivity values for the 240 μm paper with different layers and press methods. The heat capacity is 1.974×10^{3} J/Kg K and the density is 825 Kg/m^3.

Press Method	240 μm	α, Thermal Diffusivity m^2/s	k, Thermal Conductivity W/m K
Hot Plate	4 layers	1.250×10^{-8}	0.020
	9 layers	1.927×10^{-8}	0.031
	14 layers	2.299×10^{-8}	0.036
0.1 MPa Carver	4 layers	1.575×10^{-8}	0.025
	9 layers	3.556×10^{-8}	0.055
	14 layers	5.606×10^{-8}	0.085
0.4 MPa Carver	4 layers	1.821×10^{-8}	0.028
	9 layers	3.305×10^{-8}	0.082
	14 layers	5.886×10^{-8}	0.093

Table 2 shows the thermal properties of the 308 μm paper. Similar to Table 1 above, the thermal conductivity values increased as layers increased and were close for the hot plate method while they ranged from 0.023-0.076 W/m K for the low and high press pressures. Here, the values of thermal conductivity were smaller than the 240 μm results for the carver press method for both press pressures. Similar to the 240 μm paper, for the 4 layers paper, the thermal conductivity values were close regardless of press methods and pressures.

Table 2. Thermal diffusivity and thermal conductivity values for the 308 μm paper with different layers and press methods. The heat capacity is 1.374×10^3 J/Kg K and the density is 735 Kg/m^3.

Press Method	308 μm	α, Thermal Diffusivity m^2/s	k, Thermal Conductivity W/m K
Hot Plate	4 layers	2.540×10^{-8}	0.025
	9 layers	3.485×10^{-8}	0.035
	14 layers	3.512×10^{-8}	0.035
0.1 MPa Carver	4 layers	2.373×10^{-8}	0.023
	9 layers	4.608×10^{-8}	0.049
	14 layers	4.965×10^{-8}	0.050
0.4 MPa Carver	4 layers	2.669×10^{-8}	0.026
	9 layers	4.878×10^{-8}	0.052
	14 layers	7.516×10^{-8}	0.076

Table 3 shows the results for the paper that had a thickness of 383 μm. These results were not so far from others. The trend in which as the thickness of paper increased the thermal conductivity increased was not as obvious with this sample. Similar to 240 and 308 papers, the thermal conductivity values were close for the hot plate method and ranged from 0.027-0.098 W/m K for the carver press method for both pressures.

Table 3. Thermal diffusivity and thermal conductivity values for the 383 µm paper with different layers and press methods. The heat capacity is 1.344×10^3 J/Kg K and the density is 748 Kg/m^3.

Press Method	383 µm	α, Thermal Diffusivity m^2/s	k, Thermal Conductivity W/m K
Hot Plate	4 layers	2.525×10^{-8}	0.025
	9 layers	4.240×10^{-8}	0.042
	14 layers	3.243×10^{-8}	0.035
0.1 MPa Carver	4 layers	2.816×10^{-8}	0.027
	9 layers	4.970×10^{-8}	0.066
	14 layers	9.140×10^{-8}	0.098
0.4 MPa Carver	4 layers	3.301×10^{-8}	0.032
	9 layers	7.302×10^{-8}	0.098
	14 layers	8.847×10^{-8}	0.095

Table 4 shows the results for the paper that had a thickness of 430 µm. This paper represents the highest thickness used in this study. Again, this paper agreed with all previous papers in which all 4 layer papers gave close results, the hot plate method resulted in very close values, and values of thermal conductivity increased as thickness increased. Also, the thermal conductivity values ranged from 0.027-0.052 W/m K for the low and high press pressures. It is also important to point out that the values here are low compared to 240 and 383 µm papers and close to the results of 308 µm paper.

Table 4. Thermal diffusivity and thermal conductivity values for the 430 µm paper with different layers and press methods. The heat capacity is 1.065×10^{3} J/Kg K and the density is 765 Kg/m^3.

Press Method	430 µm	α, Thermal Diffusivity m^2/s	k, Thermal Conductivity W/m K
Hot Plate	4 layers	2.941×10^{-8}	0.020
	9 layers	5.641×10^{-8}	0.046
	14 layers	5.369×10^{-8}	0.044
0.1 MPa Carver	4 layers	3.463×10^{-8}	0.027
	9 layers	5.343×10^{-8}	0.041
	14 layers	5.647×10^{-8}	0.046
0.4 MPa Carver	4 layers	3.305×10^{-8}	0.026
	9 layers	5.513×10^{-8}	0.042
	14 layers	6.439×10^{-8}	0.052

The values of heat capacity and density for each paper were reported in the caption of each Table above. The 430 µm paper has the lowest heat capacity followed by 383, then 308, and finally 240 µm papers. The density on the other hand is high for the 240 µm paper followed by 430, then 383, and finally 308 µm papers.

More layers should not have higher k, however, others have reported similar results. For example, Sergiy et al. (2012) reported that as the sheet layers of the copy paper increase to 1, 2, 3, and 4 the thermal conductivity increases to 0.050, 0.065, 0.071, and 0.075 respectively. The reason behind thermal conductivity increases with thickness is not clear but it may be due to the airflow in paper pores promotes heat transfer.

Sergiy et al. (2012) also put on a list of previously measured thermal conductivities for different paper grades as in Table 5. Most of the values in the table are within the range of what measured in this thesis.

Table 5. Thermal conductivities and contact resistances reported for different paper grades Sergiy et al. (2012).

Investigations	Thermal conductivity (W/[mK])	Contact resistance (km^2/W)
Water-saturated blotter paper (solid contents from 0.3 to 0.52)[1]	0.54–1.6	
Uncalendered sheets (density from 600 to 1,000 kg/m^3, chemical pulp)[32]	0.06–0.11	
Uncalendered sheets (density from 400 to 600 kg/m^3, mechanical pulp)[32]	0.04–0.068	
Calendered sheets (density from 1,000 to 1,100 kg/m^3, chemical pulp)[32]	0.105–0.16	
Calendered sheets (density from 625 to 750 kg/m^3, mechanical pulp)[32]	0.05–0.065	
Handsheets (density 420 kg/m^3, bleached sulfite softwood)[10]	0.016–0.02	
Test liner, density from 625 to 875 kg/m^3, unbleached recycled pulp[36]	0.075–0.118	
Uncoated wood free, density* 737 kg/m^3[35]	0.15	0.833×10^{-3}
Coated wood free, density* 872 kg/m^3[35]		0.662×10^{-3}
Uncoated wood containing base, density* 689 kg/m^3[35]		0.606×10^{-3}
Coated wood containing, density* 879 kg/m^3[35]		0.476×10^{-3}
Present work (density from 625 to 1,260 kg/m^3, copy paper, uncoated and coated)	0.08–0.18	0.35×10^{-3} to 1.5×10^{-3}

Gray-Stuart et al. (2019) also reported some values of thermal conductivity of paper in machine, cross machine, and thickness directions as shown in Table 6. The values obtained were high for the machine and cross machine directions however the thickness direction provided close values to the ones measured in this thesis.

Table 6. Geometric and thermophysical properties of paper including the thermal conductivity in the machine, cross machine, and thickness directions Gray-Stuart et al. (2019).

Paper name	Grammage (g/m^2)	Thickness (μm)	k_m (W/mK)	k_{cm} (W/mK)	k_z (W/mK)
140 KTL	140	174	0.56 (0.06)	0.58 (0.06)	0.09 (0.01)
150 RFM	150	213	0.62 (0.03)	0.35 (0.02)	0.07 (0.01)
120 RFL	120	177	0.41 (0.03)	0.29 (0.02)	0.08 (0.01)
205 WTL	205	231	0.72 (0.05)	0.57 (0.03)	0.08 (0.01)
112 RFM	112	172	0.45 (0.03)	0.40 (0.06)	0.08 (0.01)
120 SCM	120	147	0.53 (0.04)	0.36 (0.07)	0.08 (0.01)
Mean	141	186	0.55	0.43	0.08

Bracketed values are standard deviations

KTL Kraft top liner; *RFM* recycled fluting medium; *RFL* recycled liner board; *SCM* semi-chemical medium; *WTL* white top liner

3.2 Heat Sealing of Two Different Papers

The paperboard properties of the uncoated papers are shown in Table 7. All properties are within normal ranges. The permeabilities are calculated from Equation 6 while the void fractions are calculated from Equation 7.

Table 7. Uncoated paperboard properties.

	Basis Weight (g/m^2)	Void Fraction	Air Permeability (m^2)
240 µm Paper	190	0.4	8.8×10^{-15}
430 µm Paper	336	0.5	3.0×10^{-14}

All is an average of more than five repeats.

Tensile test analyses were performed to look at the final bond strength of different samples. Figure 12 shows polyethylene strength (load) versus press time for 240 and 430 µm papers glued at 1, 10, 30, and 60 s press times, 0.4 MPa press pressure, and 150, 200, and 225 °C paper temperatures. For the 430 µm paper, the load needed to peel the samples apart is high, especially for the high press time. The peel force reached around 12 N in some cases. However, the load was low for the 1 s press time which ranged from 0.5 to 3.5 N. In addition, adhesive failure modes were seen for all 430 µm samples. For the 240 µm samples on the other hand, the load was low for all cases which ranged from 0.8 to 4.7 N with higher press times yielded in higher loads. Similar to the 430 µm paper, the failure was adhesive failure mode in almost all 240 µm samples.

Higher loads seen for the 430 µm paper are due to the high permeability and porosity this paper has which help the polymer to penetrate deeper and perform “mechanical interlocking ” between the polymer and the adherends compared to the 240 µm paper which has low permeability and porosity and low penetration.

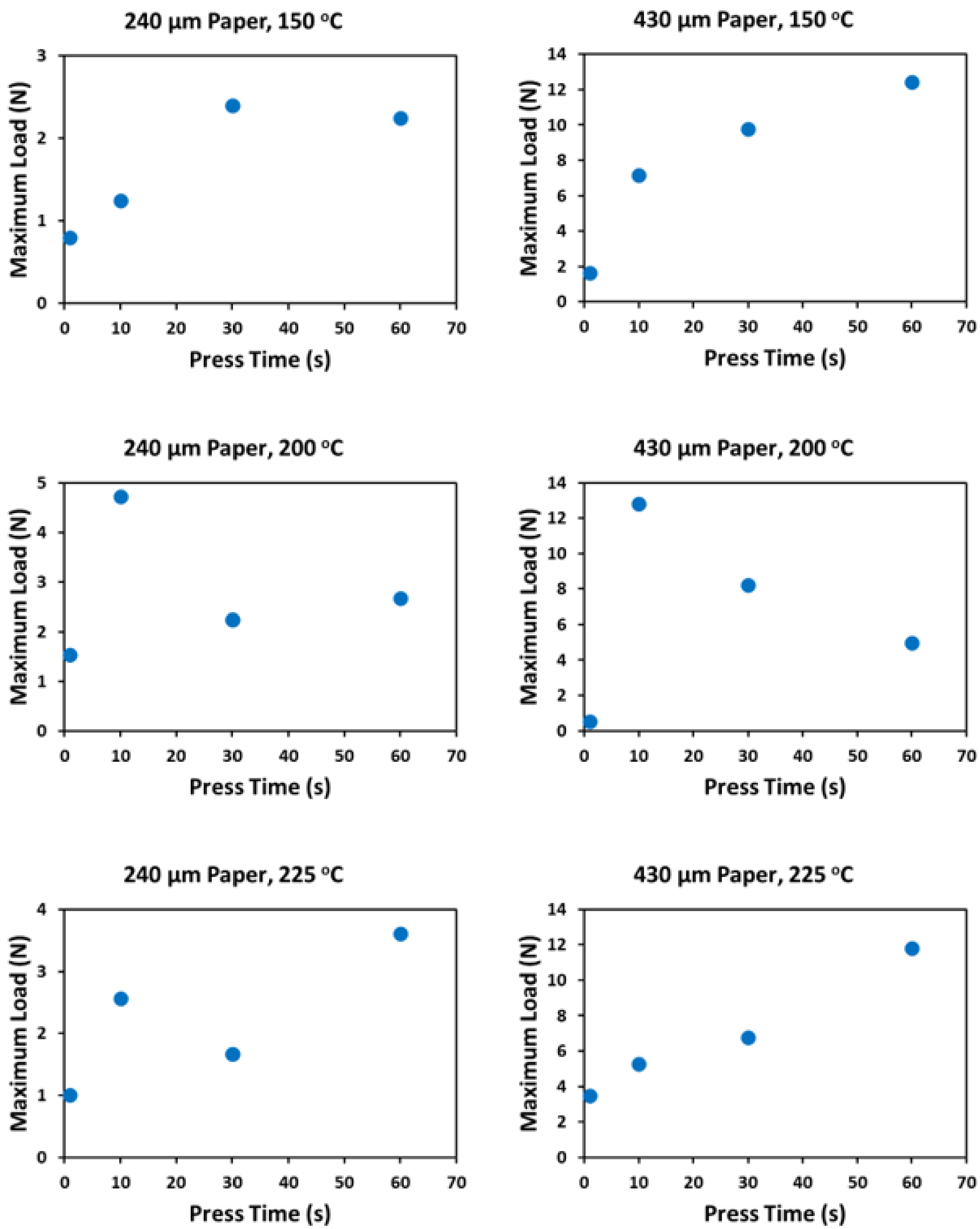

Figure 12. Tensile test results for 240 µm paper (left column) and 430 µm paper (right column) for 0.4 MPa press pressure.

3.3 Conclusions and Summary

- A method with a simple hot plate and another with a carver press were developed to estimate the thermal conductivity of different papers.
- A method was used to obtain the heat capacity of paper that is easy to do in the laboratory.
- A simple equation was developed to predict the time and temperature needed for heat sealing.
- Values of thermal conductivity are in the range of what others have reported.
- Values between papers selected here are similar.
- The method with the hot plate gives similar results as the carver press.
- The influence of press pressure seems minimal for the range tested here.
- More layers cause the calculated value to increase. The reason is not clear. It may be airflow in paper pores promotes heat transfer.
- Adhesions of polyethylene films into the 430 µm papers resulted in higher loads compared to the 240 µm paper which is due to the higher porosity and permeability of this paper.
- As press time increases the load needed to peel the samples apart increases as well.
- Adhesive failure modes were seen for almost all 240 µm and 430 µm samples.

MODELING

4.1 Introduction

The heat sealing of multi-layers of paper applying polymer in between was simulated by using COMSOL Multiphysics to understand the influence of heat transfer parameters. The heat-sealing process can be influenced by many parameters and the properties of the sealant layer. The purpose of using modeling is to find the optimal conditions for heat sealing. The industry finds that the most favorable condition for heat sealing is by trial, and error and the flow of this path is more expensive and wasteful. By using modeling, sealant performance can be achieved without wasting time and money performing and repeating experiments on heat sealing.

Modeling reduces time and effort before laboratory work. It can also point the light on the key parameters that need to be addressed during experiments. Besides, it can be beneficial for the short time scale problems as well as microsystems. However, modeling usually has assumptions that limit the process and need to be listed and documented well.

4.2 Model Set-Up

A commercial finite element code (COMSOL Multiphysics 6) was used to model 2D heat transfer in a porous media. The porous media was paper that consisted of various papers with different physical properties. Two simulations were used in this research: One is for heat transfer in paper while another is for heat transfer in paper and polymer. The physical properties of paper were taken from the experimental results; however, the polymer properties were obtained from the literature.

4.2.1 Modeling of Heat Transfer in Paper

In the software, heat transfer in solid physics was chosen to study heat transfer in different layers of paper (4, 9, and 14 layers). A 2D rectangle geometry was selected to represent paper. The problem was an unsteady-state. The material tree is used to assign physical properties to the geometry hence the paper. The next tree was the physics tree where the boundary and initial conditions are set. The temperature was assigned to the inlet of the heated plate at 150 ºC while the paper was at room temperature initially. Also, no flux conditions were set on the walls. The mesh tree is used to specify the number of elements (besides others) that need to be accounted for in the computational steps. The extremely fine mesh was selected as shown in Figure 13 with a mesh selection window depicted in Figure 14. Figure 15 shows the tree of the model.

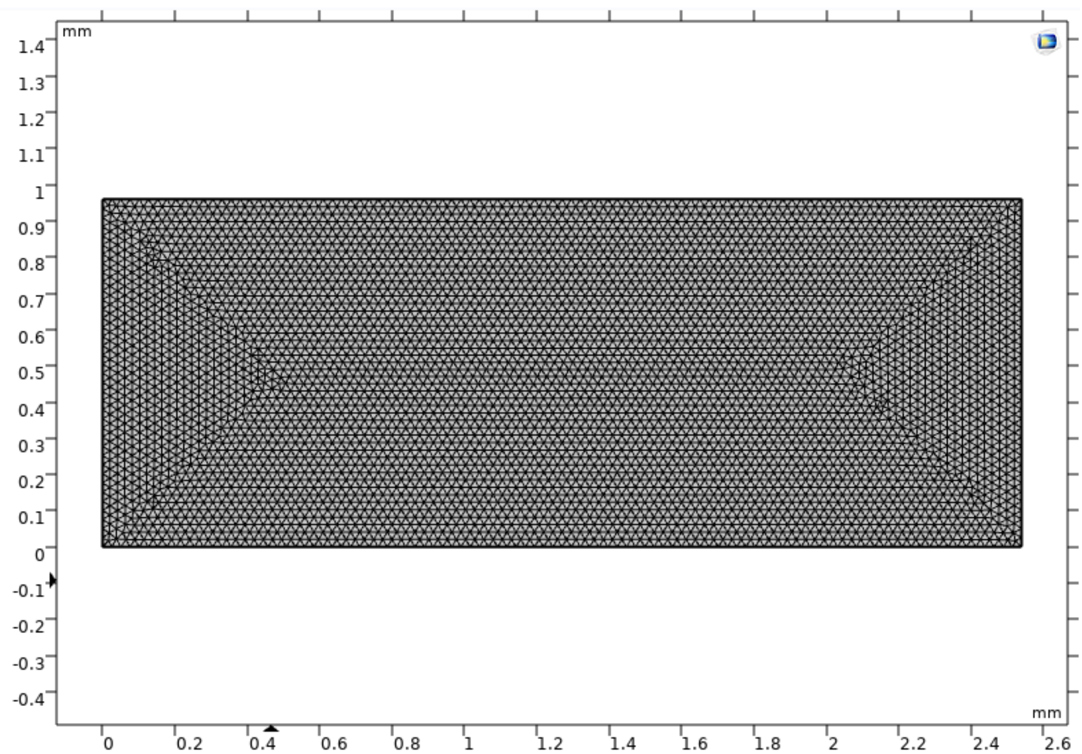

Figure 13. Paper geometry in 2D coordinate meshed system.

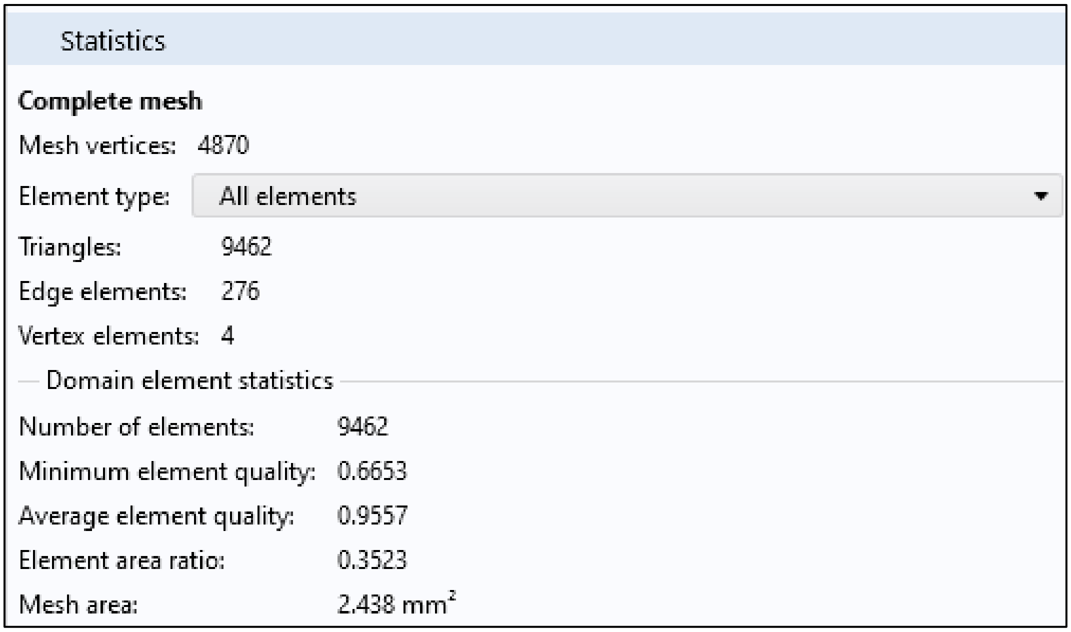

Figure 14. Extremely fine mesh statistic for paper.

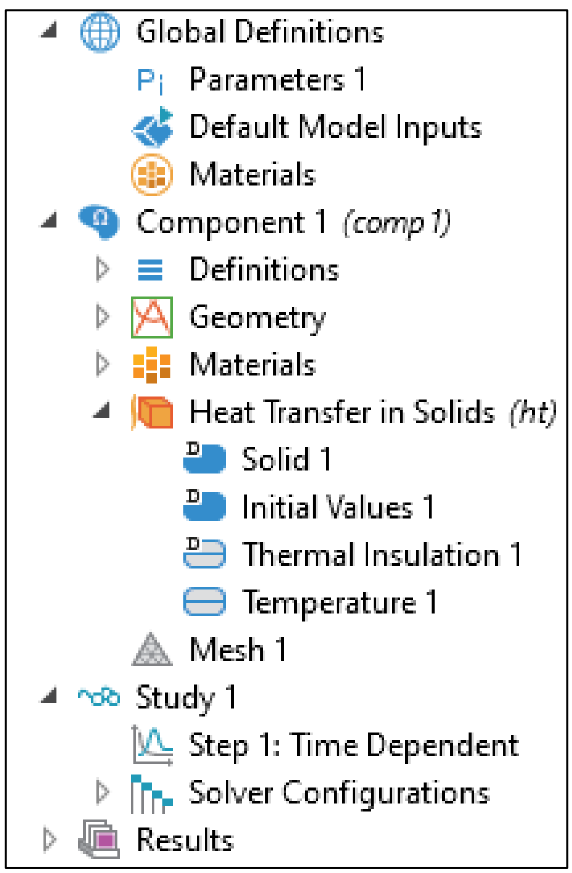

Figure 15. Tree properties for heat transfer in paper.

It is worth mentioning that for the range of papers studied here, the thermal conductivity and heat capacity were measured in the lab using two different techniques, the heated plate method, and the carver press method. In the carver press method, the nip pressure was fixed to 0.1 MPa or 0.4 MPa. Here, the modeling results were only compared to the experimental ones that were obtained from the heated plate method using only four layers. Table 8 presents the paper parameters that were measured in the lab and used inside the model.

Table 8. Model parameters for the experimental heated plate method.

4-layers	Cp, Heat capacity (J/Kg K)	ρ, Density (Kg/m^3)	k, Thermal conductivity (W/m K)
240 µm paper	1.974×10^3	825	0.020
308 µm paper	1.374×10^3	735	0.025
383 µm paper	1.344×10^3	748	0.025
430 µm paper	1.065×10^3	765	0.020

Each value is an average of six runs.

4.2.2 Results and Discussion

The experimental results were compared with the model predictions as shown in the following figures. From the figures, we can see that the lab results agreed well with the model predictions for the 240 µm paper and were close for the 308 and 383 µm papers but underpredicted the model for the 430 µm paper. Moreover, both model and experimental results followed the trend in which the temperature came to a steady state at around 2 minutes. The model always reached the temperature of the heated plate (150 ºC) at the end of the run, however, the experiments varied as the paper thickness varied. For instance, the paper temperature after five minutes of the run was 148 ºC, 130 ºC, 136 ºC, and 107 ºC for the 240 µm, 308 µm, 383 µm, and 430 µm, respectively. Besides thickness changes, these results are also influenced by the thermal conductivity, heat capacity, and density of the selected paper.

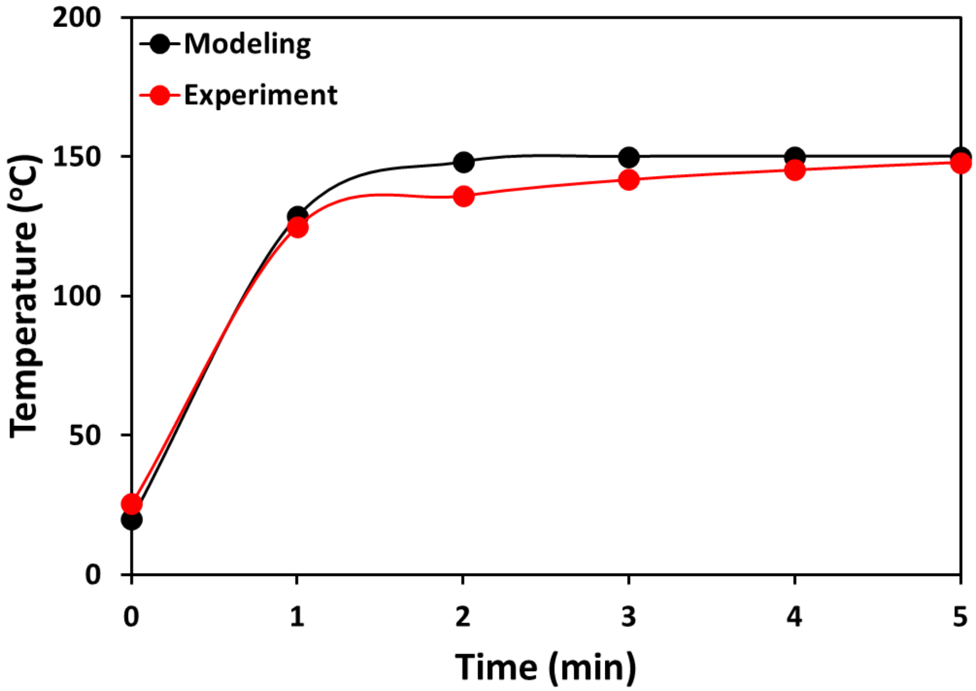

Figure 16. Comparing model predictions to the experiments for the 4-layers of 240 µm paper.

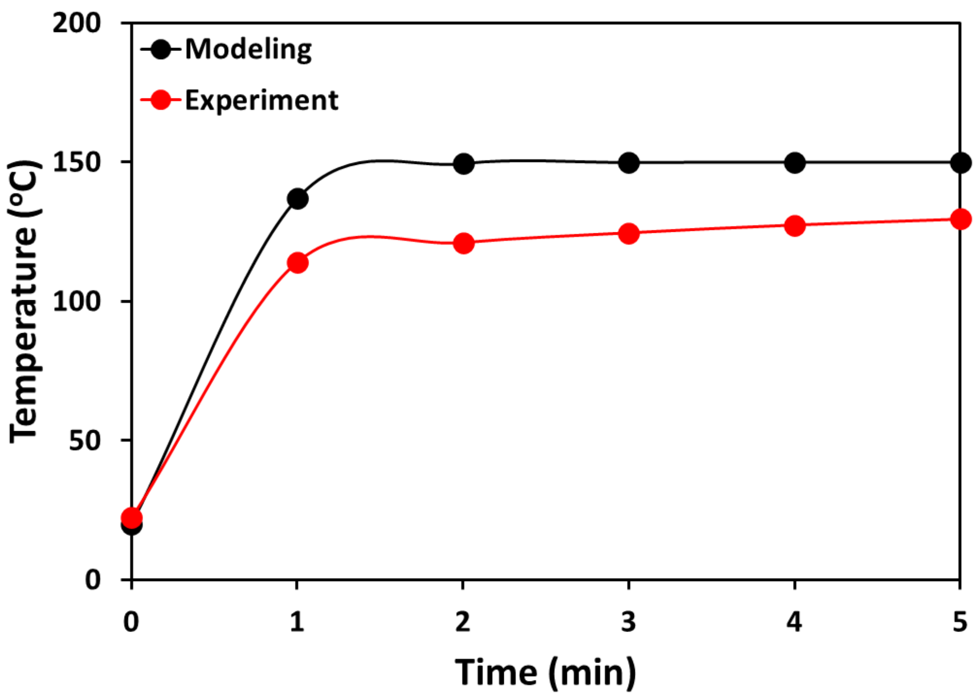

Figure 17. Comparing model predictions to the experiments for the 4-layers of 308 µm paper.

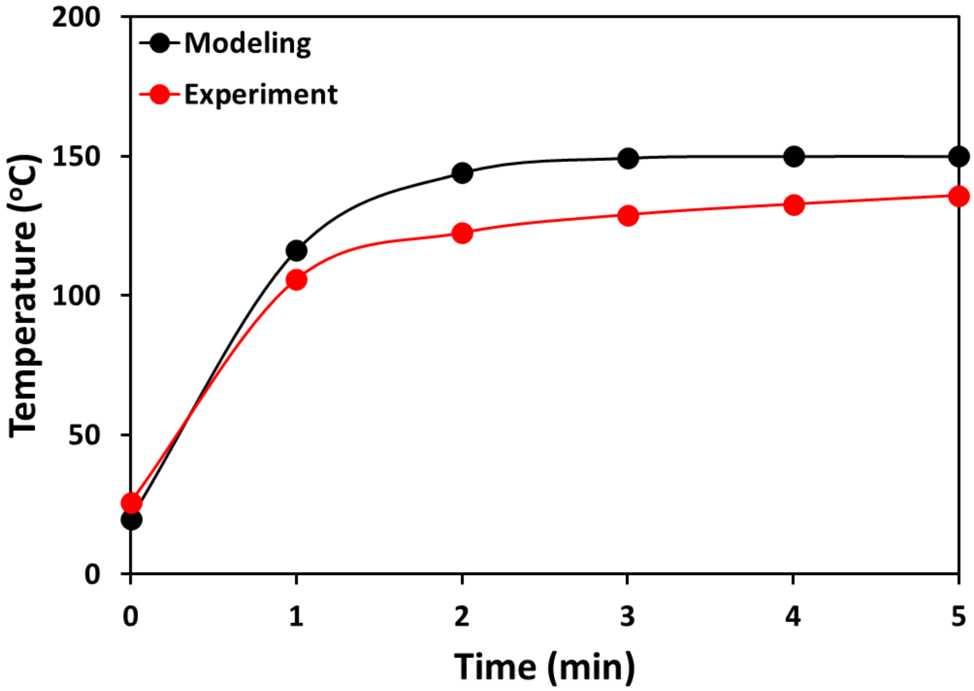

Figure 18. Comparing model predictions to the experiments for the 4-layers of 383 μm paper.

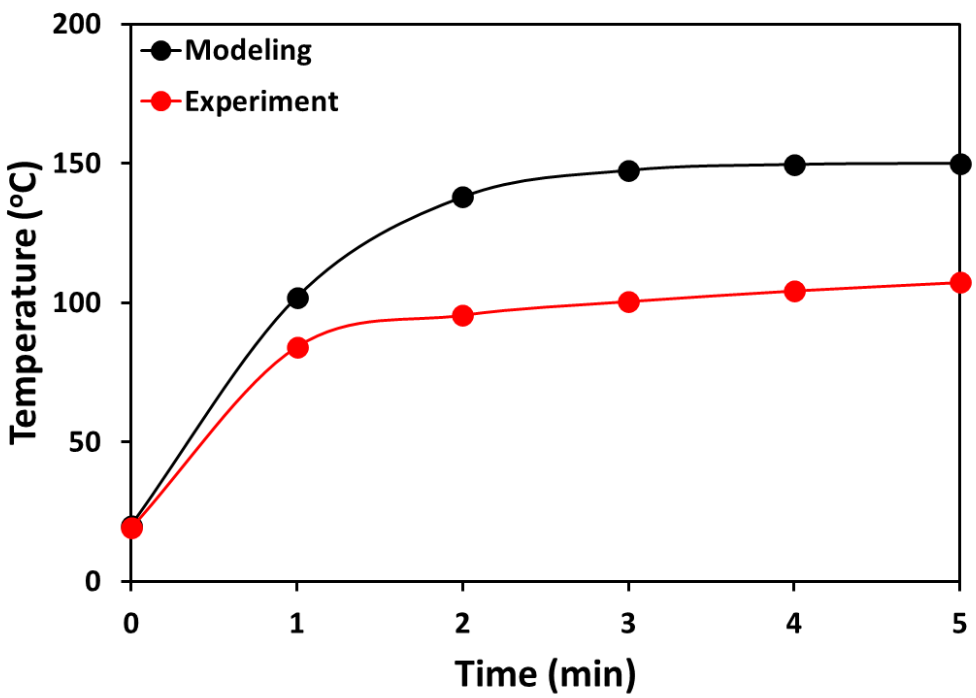

Figure 19. Comparing model predictions to the experiments for the 4-layers of 430 μm paper.

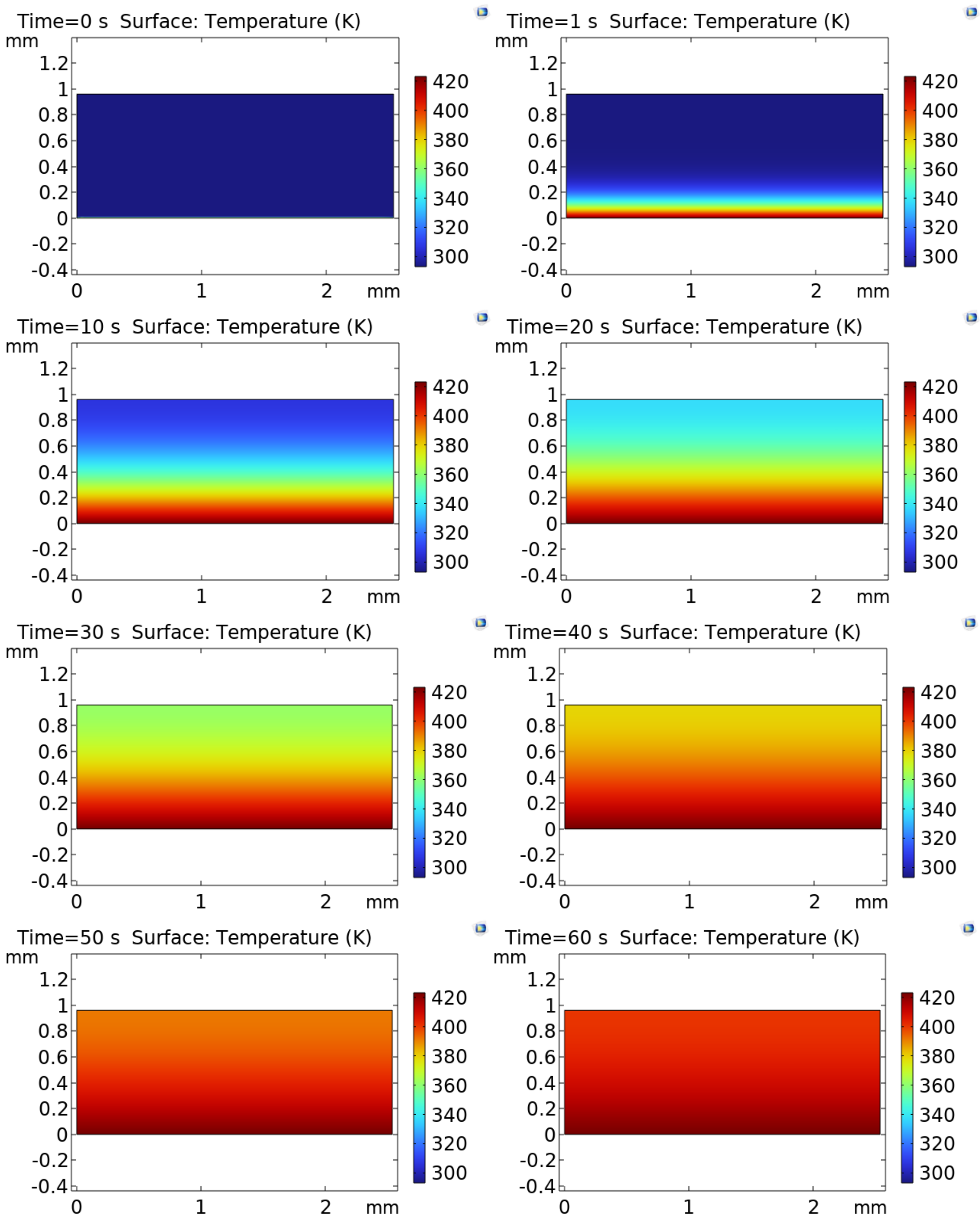

Figure 20. 2D images of temperature as a function of time for 240 μm paper. Red is high temperature and blue is low.

These results are encouraging and motivating to test other layers and compare them with the model as well. This is the first time where the thermal conductivity of paper was measured in the lab and used in the model for validation where it has never been measured using such a process in the literature.

It is worth noting that during the first 60 seconds of the run, there is a big jump in the temperature of the paper then it stayed constant at around 150 ^{o}C for the remaining time. The 2D images from COMSOL modeling for the 4-layers of the 240 μm paper are shown in Figure 20 above. From the 2D images, the temperature of the paper was low at low time steps but it increases as time increases to become at the temperature of the hot surface at 60 seconds time step.

4.2.3 Modeling of Heat Transfer in Paper and Polymer

The second part for modeling is the polymer heat sealing into the paper. Instead of heat transfer in solids, heat transfer in fluids was used here. In addition, the laminar flow physics was selected to show when the polymer penetrates the papers. In the laminar flow physics section after we neglected both gravity and inertia terms, we added two boundaries that were the inlet and the outlet to which we assigned the inlet (0.4 MPa top boundary) and the outlet pressure (0 MPa bottom boundary). This pressure value was selected to be similar to the pressure value used in the experiments. The 2D geometry used was simple where three rectangles were connected: two for the paper and another for the polymer (100 μm thickness). The problem was an unsteady-state. The material tree is used to assign physical properties to the papers and polymer. The next tree was the physics tree where the boundary and initial conditions are set. The temperature was set to a high value (200 ^{o}C) for both sides of the papers and the polymer layer was in between while the papers and polymer were at room temperature initially. Also, no flux conditions were set on the walls. The extremely fine mesh was selected as shown in Figure 21 with a mesh selection window depicted in Figure 22. Figure 23 shows the tree of the model.

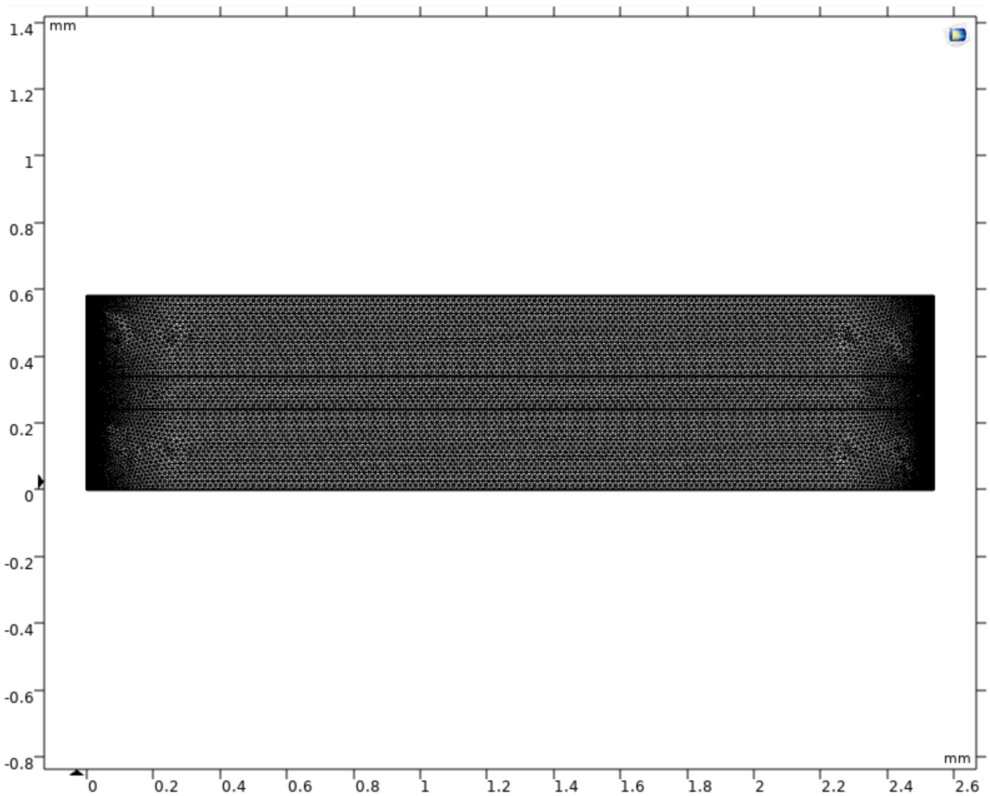

Figure 21. Papers and polymer in between geometry in 2D coordinate meshed system.

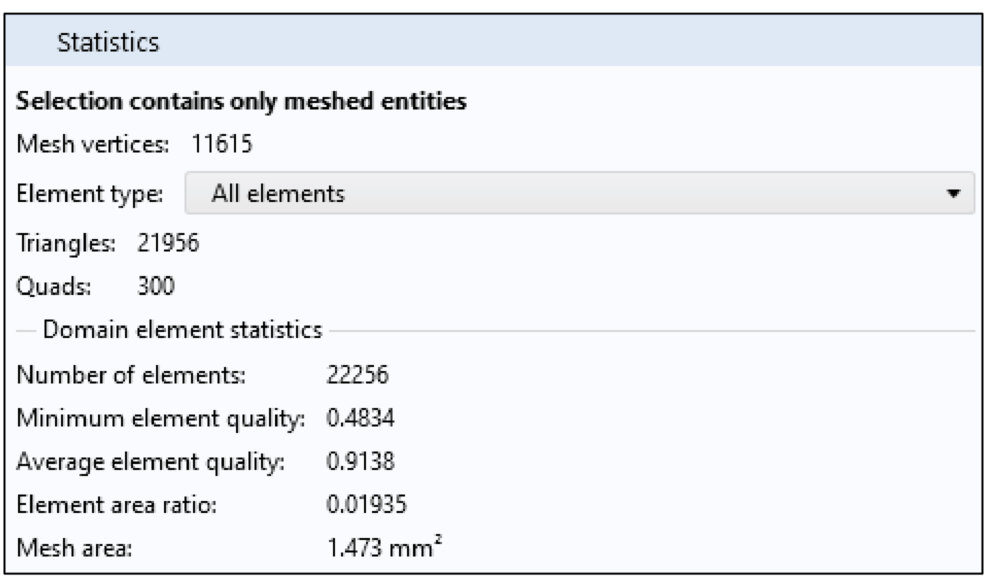

Figure 22. Extremely fine mesh statistic for papers and polymer.

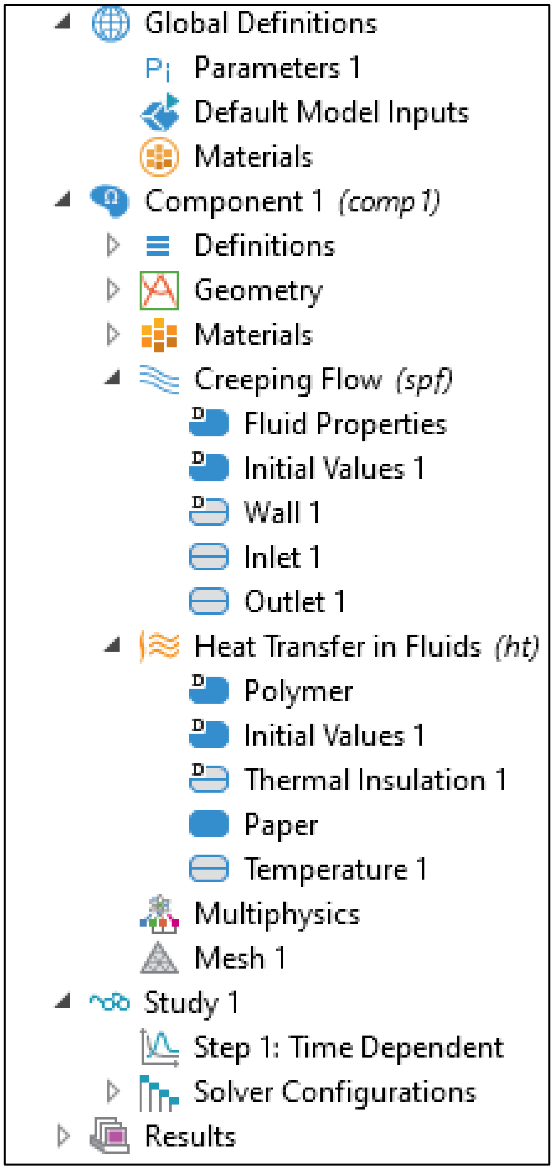

Figure 23. Tree properties for the polymer flow into the paper as well as heat transfer.

Conversely, to heat transfer in the paper above, the modeling results were only compared to the experimental ones that were obtained from the carver press method using only one layer of paper. Table 9 presents the parameters of the paper that were measured in the lab and the polymer properties that were obtained from the literature. It is important to note that the thermal conductivity values listed in the table are an average of the thermal conductivity values calculated for the different layers of paper (4, 9, and 14 layers).

Table 9. Model parameters for the experimental carver press method.

	Cp, Heat capacity (J/Kg K)	ρ, Density (Kg/m^3)	k, Thermal conductivity (W/m K)
240 µm paper	1.974×10^3	825	0.067
308 µm paper	1.374×10^3	735	0.051
383 µm paper	1.344×10^3	748	0.075
430 µm paper	1.065×10^3	765	0.040
Polymer	2.315×10^3	925	0.480

Each value is an average of six runs except polymer parameters are from literature.

4.2.4 Results and Discussion

Figure 24 shows the modeling results of temperature increases as a function of time for the 240 µm paper while Figure 25 shows that for the 430 µm paper. It is obvious that as the paper thickness increases the time required for the polymer layer to reach the temperature of platens increases. This situation is clearly shown in Figures 26 and 27 which show the temperature increases as time progresses. The temperature profile shown in both figures represents the surface temperature of the polymer. The reason for this model is to record the time required for the polymer to reach the temperature of the heated platen so it can flow into the paper and be sealed. It can be seen that two seconds is enough for the polymer to achieve the heated platen temperature for the 240 µm paper. However, more time was needed when the paper thickness increased to 430 µm. Also, changing points on the polymer surface resulted in the same temperature profile meaning that the temperature was uniform along the surface of the polymer.

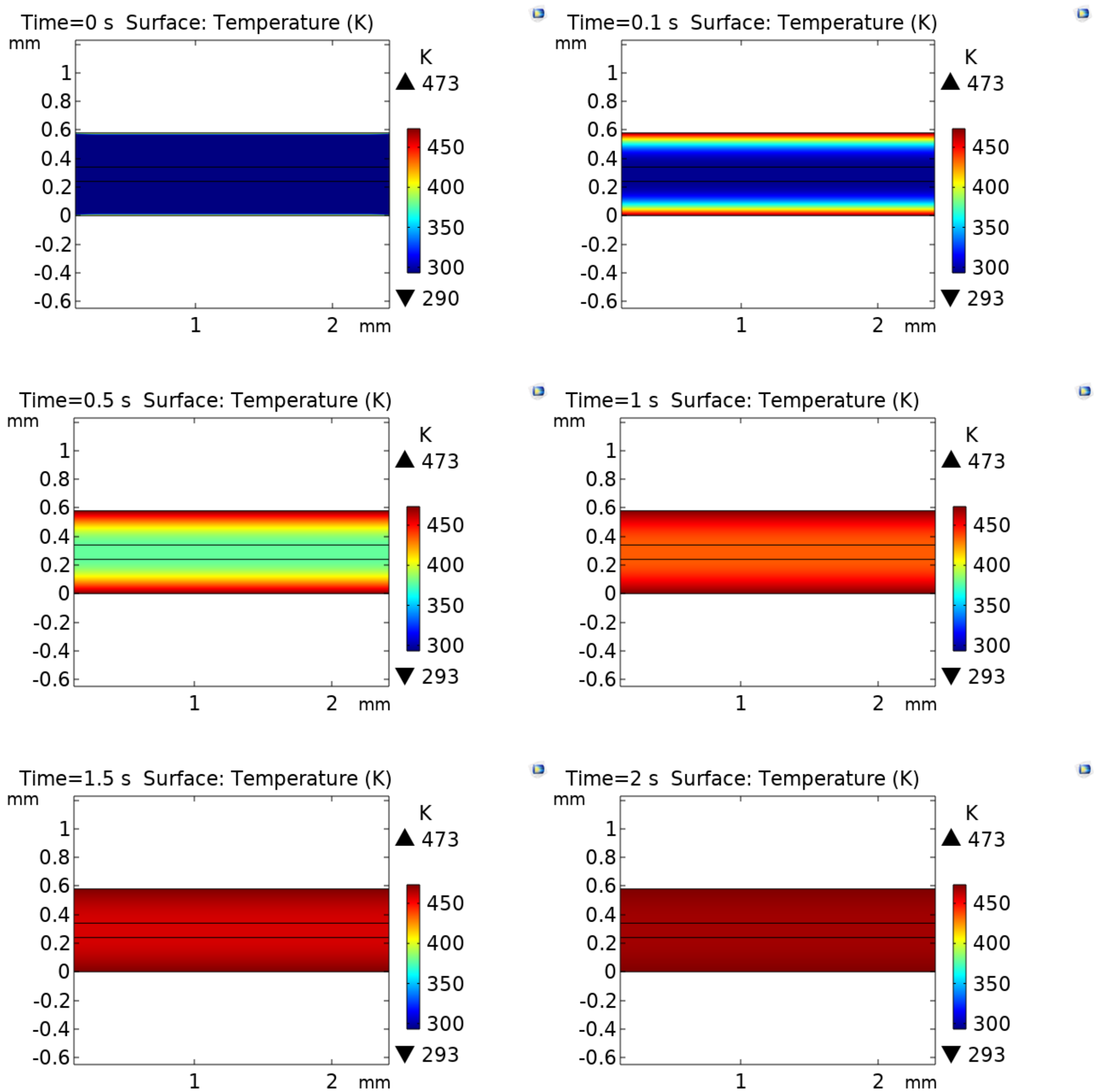

Figure 24. 2D images show how the temperature transferred through 240 µm papers into the polymer layer as a function of time. Red is high temperature and blue is low.

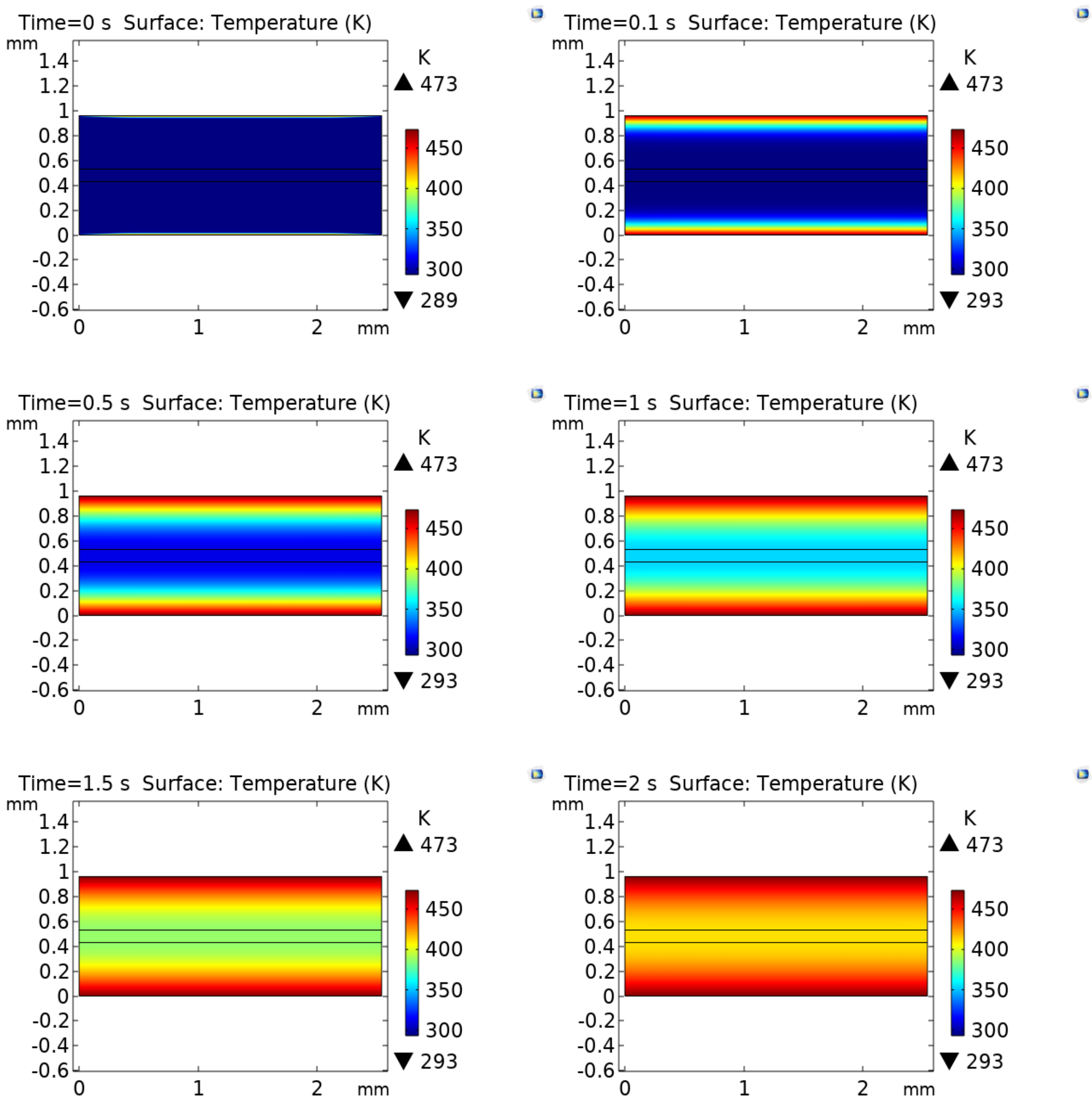

Figure 25. 2D images show how the temperature transferred through 430 µm papers into the polymer layer as a function of time. Red is high temperature and blue is low..

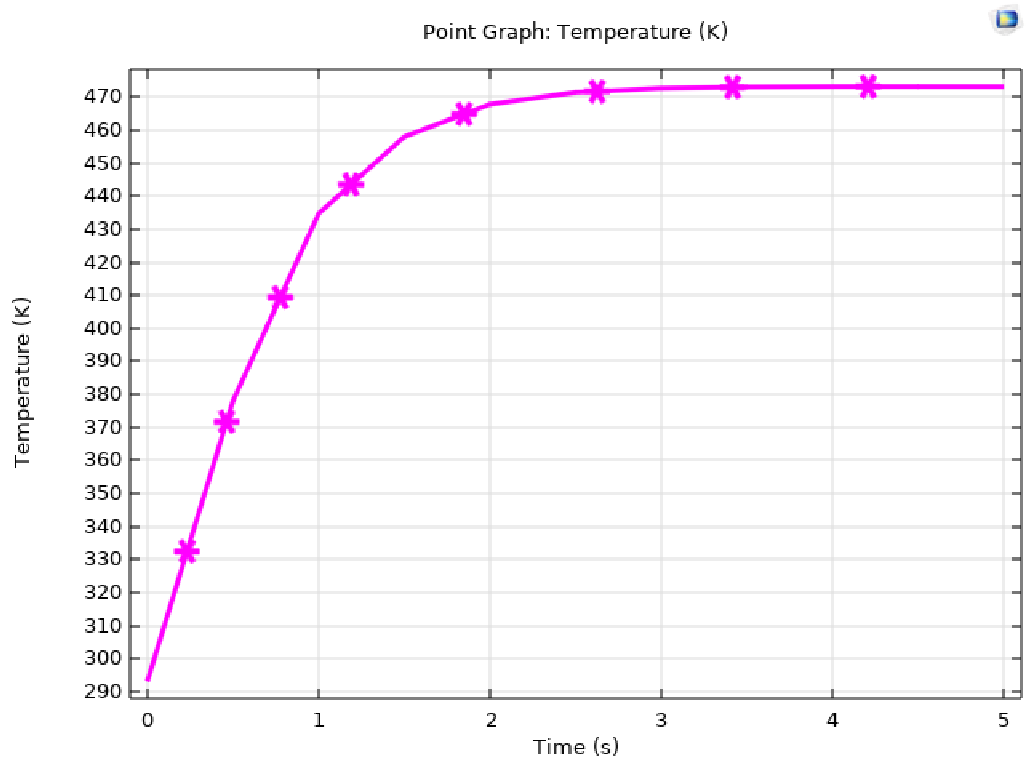

Figure 26. Surface temperature of polymer as a function of time for the 240 µm paper.

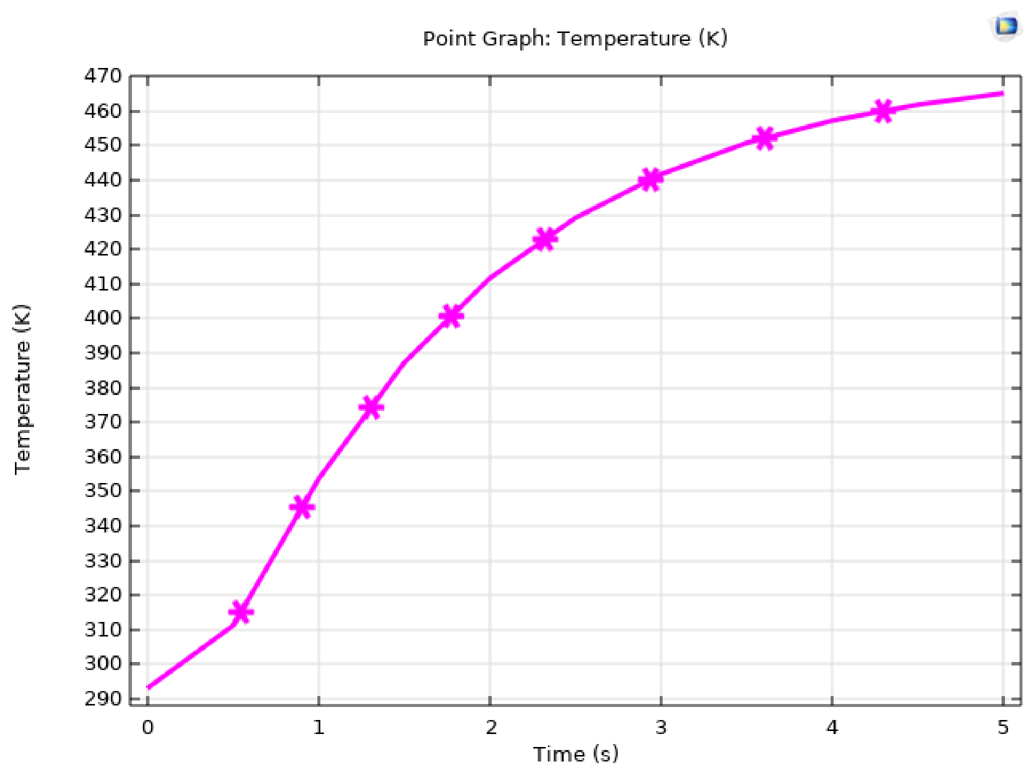

Figure 27. Surface temperature of polymer as a function of time for the 430 µm paper.

4.3 Conclusions and Summary

- This study aims to understand the influence of paper thermal conductivity, heat capacity, and density on the heat sealing process of polymer to paper.
- The model agreed in a reasonable range with the experiments for the heat transfer in the paper.
- For the polymer flow model, as the thickness of the paper increases the polymer needs more time to reach the temperature of the paper and flow.
- Each paper has a different thermal conductivity, heat capacity, and density which affects the heat transfer process. In the current model, both 240 µm and 430 µm papers were tested but others are planned as well.
- It would be beneficial and less costly to test modeling before the experiments. It would save time and effort to predict the temperature and better understand heat transfer during heat sealing.
- By using COMSOL modeling, the optimum time and temperature can be predicted and used for the experimental heat sealing process.
- Modeling a wide range of paper and polymer properties can help in understanding the parameters that are most influencing the process.

CONCLUSIONS AND RECOMMENDATIONS

The three main goals of this thesis are first to experimentally estimate the thermal conductivity and heat capacity of different papers, second to study the influence of these thermal properties on the bond strength of polymer-paper substrates, and third to track the time-temperature relationship between paper-paper and polymer-paper samples using a finite element method investing the properties measured in the laboratory. In addition, a simple equation was developed to predict the time and temperature needed for heat sealing. A mechanical tester was also used to measure the final bond strength of glued samples.

The first part of this thesis involved using the carver press and hot plate methods to measure the thermal conductivity of 240, 308, 383, and 430 μm papers. In both methods, 4, 9, and 14 layers of paper were used. The results for all methods were close and promising novel and simple methods for future usages where the values of thermal conductivity are in the range of what others have reported.

The second part of this thesis involved the sealing of low-density polyethylene (LDPE) film to 240 and 430 μm papers. The carver press method was used with 150, 200, and 225 ^{o}C paper temperatures, 1, 10, 30, and 60 s press times, and 0.4 MPa press pressure. The results for the two papers used here indicated that the permeability and porosity of the paper are the key parameters where higher permeability and porosity helped in getting higher bond strengths. Also, higher press times resulted in higher loads with adhesive failure modes seen in almost all samples for all conditions.

Regarding modeling, the results showed agreement in a reasonable range with the experiments for the heat transfer in the paper. For the polymer model, as the thickness of the paper increases the polymer needs more time to reach the temperature of the paper and flow. Modeling can help in reducing lab efforts, time, and cost and it could be a great tool to predict the temperature and understand heat transfer prior to the experiments.

Although there were many parameters addressed in this thesis, there are still other subjects that need more attention in the future. The first recommendation would be to investigate different polymers and see how that could impact the bond strength. The influence of press pressures on thermal properties measurements and the heat sealing process could be a separate study. Another recommendation would be to further study the penetration of polymer into the paper and what type of adhesion occurs. It would be beneficial also to study the effect of pore size distribution and pore volume on both systems. Another study could look at the influence of latex levels and pigment sizes on the heat sealing process. A great study would be to investigate the thermal properties and heat sealing of papers with lower thicknesses. Finally, it would be interesting to dive more into modeling and look for the integrity of heat sealing which has a very big impact on food waste investing COMSOL modeling.

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