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REDUCTION OF VOLATILE ORGANIC COMPOUNDS EMITTED DURING KILN DRYING OF SOUTHERN YELLOW PINE LUMBER

By

Joseph Dahlen

A Thesis Submitted to the Faculty of Mississippi State University in Partial Fulfillment of the Requirements for the Degree of Master of Science in Forest Products in the Forest Products Department

Mississippi State, Mississippi

December 2006

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Joseph Martin Dahlen

2006

REDUCTION OF VOLATILE ORGANIC COMPOUNDS EMITTED DURING KILN DRYING OF SOUTHERN YELLOW PINE LUMBER

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The objective of the research is to reduce the volatile organic compounds (VOCs) emitted during kiln drying of southern yellow pine (SYP) lumber. Three treatment methods were explored to determine their effect on VOC emissions. The three methods were configured at the air exhaust of a pilot-scale dry kiln.

1) A steel reactor containing copper tubing heated to 100°, 200°, and 240° Celsius was evaluated. Temperature did not statistically affect VOC emissions (p-value $= 0.1674$). The average reduction of VOCs at 240 \degree C was seven percent. 2) The use of hydrogen peroxide and ultra-violet (UV) light did not reduce VOCs emitted during kiln drying. 3) The use of Fenton's reagent, with hydrogen peroxide and ferrous sulfate, also did not reduce VOCs emitted during kiln drying. 4) It is recommended that future studies with copper heated to temperatures above 240° C be performed.

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CHAPTER I INTRODUCTION

Background

Volatile organic compounds (VOCs) are released from southern yellow pine (SYP) lumber during kiln drying. VOCs react with nitrogen oxides (NO_x) in the presence of ultra-violet (UV) light to form ground level ozone (O_3) , a major component of smog. The Environmental Protection Agency (EPA) has regulated VOC emissions more strictly during the past decade due in part to the 1990 amendments to the Clean Air Act (Boswell, 1991).

Objective Statement

The objective of the research was to examine potential methods to reduce the quantity of VOCs emitted during kiln drying of SYP lumber. Current technologies used to reduce VOCs are not compatible with lumber dry kilns due to the high moisture present in kiln exhaust, non-stable air flow, and economics. Three treatment methods were explored to determine their effect on VOC reduction using a pilot-scale dry kiln.

The primary treatment method, heated copper, explored the use of copper as a catalytic oxidizer for VOC reduction. A heated 3.5-inch (8.9 centimeter) diameter; fourfoot (1.2 meter) long steel reactor containing seventy feet (21.6 meter) of 0.625 inch (1.6 centimeters) diameter copper tubing was heated to 100°, 200°, and 240° Celsius (212°, 392°, 464° Fahrenheit). Three runs at each temperature (treatments) and three unheated control runs were conducted using a randomized complete block design. The kiln exhaust was sampled prior to the reactor and after the reactor to determine the effect of the heated reactor on VOC emissions.

A second treatment method, hydrogen peroxide and UV Light, used hydrogen peroxide (H_20_2) in the presence of ultra-violet (UV) light, to attempt to produce hydroxy free radicals (●OH) that may react with and thus reduce VOC emissions. Hydroxy free radicals are the neutral form of the hydroxide ion, a highly reactive compound that can react with VOCs and other compounds in the atmosphere (Waters, 1946). A stainless steel wet scrubber was used as the reactor in combination with a single UV light while the hydrogen peroxide was circulated through the scrubber via a water pump. Twelve and 450 watt UV lights were tested separately to investigate the optimal bulb power needed to reduce VOCs. The research was conducted in collaboration with the Institute for Clean Energy Technology at Mississippi State University (formerly Diagnostic Instrumentation and Analysis Laboratory).

A third treatment method, Fenton's reagent, tested a Fenton's reagent with hydrogen peroxide and ferrous sulfate (FeS0₄), to produce \bullet OH radicals that may react with and thus reduce VOC emissions. This method is similar to the second treatment method. Fenton's reagents can oxidize VOCs and contaminants by reacting hydrogen peroxide with an iron catalyst. A stainless steel wet scrubber was used as the reactor while the hydrogen peroxide and ferrous sulfate was circulated through the scrubber via a water pump. The research was also conducted in collaboration with the Institute for Clean Energy Technology.

CHAPTER II LITERATURE REVIEW

Southern Yellow Pine Lumber

SYP is composed of four major species; loblolly (*Pinus taeda* L.), short-leaf (*Pinus echinata* M.), long-leaf (*Pinus palustris* M.), and slash pine, (*Pinus elliottii* E.) as well as five minor species (Neubrech, 1939). Lumber from these trees is categorized and graded as SYP because of their similar anatomy and physical properties (Forest, 1999). The lumber from these trees is the strongest and densest amongst the commonly used softwoods that grow in the United States (Hoadley, 1993). The specific gravity of SYP typically averages between 0.51 and 0.61 (Hoadley, 1993). SYP is primarily used as a feedstock for paper, structural lumber, and treated wood products.

SYP trees are located primarily in the Southern Eastern United States, i.e. from East Texas to the Atlantic Coast and North to southern Virginia and Kentucky (Wear and Greis, 2002). These states account for fifty-eight percent of the timber production in the United States (Southern Pine 2005), and collectively they produce more forest related products than any single foreign country (Wear and Greis, 2002). SYP is the primary species group harvested from the South, with production totaling about 18 billion board

feet in 2004 (Southern Forest, 2005). SYP accounts for forty-four percent of softwood lumber production in the United States (Howard, 2001).

In 1997, SYP grew on over 62 million acres in the United States on both natural and plantation stands (South and Buckner, 2003). In 1952, the Southern region accounted for approximately forty-one percent of lumber production while the Pacific region accounted for approximately thirty-eight percent of lumber production. In 1997, production in the Pacific region decreased to eighteen percent while production in the southern region increased to fifty-eight percent. (Prestemon and Abt, 2002). But while forest-based production continues to shift to the South, the total amount of SYP acreage decreased by sixteen million acres since 1953, primarily due to urbanization (South and Buckner, 2003). To meet the increased demand for forest products, plantation grown trees now account for thirty-two million acres in 1999 (Prestemon and Abt, 2002).

Lumber Drying

Water typically constitutes roughly half of the weight of freshly sawn, green lumber (Haygreen and Bowyer, 1996). Liquid water and vapor water reside within the cell lumen and the cell wall is saturated with bound water in green lumber. During lumber drying, all of the liquid water is removed from the cell lumen while some water vapor still remains within the cell lumen and some bound water still remains in the cell wall. (Haygreen and Bowyer, 1996). The wood is dried from approximately 100 percent moisture content (dry basis) to below nineteen percent moisture content for softwood lumber, and between six to eight percent for hardwood lumber (Haygreen and Bowyer, 1996). Advantages to drying lumber include reduction of weight for shipping and

handling purposes, avoiding in-service shrinking and swelling, reduction of attacks from mold, fungi, and insects, increased strength properties, ease of application of wood preservatives, and customer satisfaction (Henderson, 1951).

The fastest practical method of drying lumber is through kiln drying. Softwood lumber is typically dried in compartment kilns (Forest, 1991). Compartment kilns often have charges loaded into the kiln via tracks and the charges remain stationary during the drying process. In 1993, there was an estimated 2500 lumber dry kilns used to dry SYP lumber in the United States (Sellers, 1993).

Softwood kilns usually use steam (indirect heat) or combustion gasses (direct heat) to increase the temperature of air to high temperatures where it is passed through the stacked lumber via a series of fans (Forest, 1991). Water is vaporized from the wood surface into the air. As the water evaporates from the surface of the wood into the air, water migrates from the inside of the wood to the surface of the wood. (Henderson, 1951). The rate of drying in the wood is determined by the physical properties of the wood, the wood moisture content, the dimensions of the wood, and the drying conditions. The drying conditions are controlled with the air flow rate of the kiln, the velocity of the fans, the dry and wet bulb temperatures, and the size of the lumber charges (Koch, 1972). As the wet bulb temperature, and hence the moisture in the air increases in the kiln the air is exhausted through vents and new air is circulated in. The physical properties of the wood dictate how fast the water moves from the inside of the wood to the surface. The ideal situation is the removal of water from the surface of the lumber at the same rate at which water moves from the inside to the surface of the lumber (Wilkinson, 1979).

 Different wood species dry at different rates. For example, SYP is a fast drying species while red and white oak dry very slowly due to slow moisture movement from the interior of the wood to the surface. Kiln schedules are used to control the proper amount of drying in wood. The industry categorizes kiln schedules in four types, low, conventional, elevated, and high-temperature (Forest, 1991). A low-temperature dry kiln operates between 70° and 120° Fahrenheit. A conventional-temperature dry kiln operates between 110° and 180° Fahrenheit. An elevated-temperature kiln operates between 110° and 211° Fahrenheit. A high-temperature kiln operates above 212° Fahrenheit, typically ranging from 230° to 280° Fahrenheit (Forest, 1991). The majority of southern yellow pine is dried in high-temperature dry kilns, while the majority of hardwood lumber is dried in the conventional-temperature kilns.

Extractives

A small fraction of the overall wood components is composed of numerous compounds that are soluble in water or organic solvents; these compounds are termed "extractives" (Sjostrom, 1993). Extractives are non structural components that aid in insect and fungi protection, and provide a food source to a living tree (Sjostrom, 1993). SYP contains approximately two to ten percent extractives (Panshin et al., 1950). Extractives include terpenoids, flavonoids, lignans, phenols, waxes, fats, sugars, and resin acids (Obst, 1998). The terpenoids are the largest component of SYP extractives and are composed of isoprene units composed of five carbon and eight hydrogen atoms. (Sjostrom, 1993). Terpenoids can be used in a variety of products including fragrances, insect repellents, medicine, flavoring agents, and waterproofing materials (Obst, 1998).

Figure 2

Terpenoids (Zavarin and Cool, 1991)

 Monoterpenes are composed of two isoprene units and are the primary chemicals volatilized during SYP drying (Koch, 1972). α-Pinene and β-pinene are monoterpenes that respectively account for approximately sixty-four percent and twenty-eight percent of the terpenes in loblolly pine (Drew and Pyland, 1966). SYP lumber averages between three and four pounds of VOCs emitted per 1,000 board feet of lumber during kiln drying (Milota, 2000).

Alpha Pinene

Beta Pinene

α-pinene and β-pinene

Terpenoids volatilize during lumber drying due to their relatively low molecular weight and high vapor pressure. The volatilization of these oils result in air pollution (Obst, 1998). These pollutants are broadly referred to as volatile organic compounds (VOCs) (Emery, 1991). Volatile organic compounds are emitted into the environment both naturally and through human related activities. While many VOCs occur naturally, the rate of emissions into the atmosphere has increased substantially since 1900 due to human activities (Bailey, 2000). VOCs react with nitrogen oxides (NO_x) in the presence of ultra-violet light to form ground level ozone, which can lower plant growth and cause human respiratory problems (Environmental, 1998).

Pollution Control Technology

VOCs emitted during the industrial processes are typically destroyed with up to 99 percent efficiency through regenerative catalytic oxidizers, thermal oxidizers, and regenerative thermal oxidizers (Gay, 1997). Regenerative catalytic oxidizers destroy VOCs by passing the pollutants through a combustion chamber heated to approximately 425° C (800° F) equipped with platinum and/or palladium catalyst packing. Thermal oxidizers destroy VOCs by passing the pollutants through a combustion chamber heated to approximately 730° to 815° C (1,350° to 1,500° F). Regenerative thermal oxidizers destroy VOCs by passing the pollutants through a combustion chamber heated to 815° to approximately 980° C (1,500° to 1,800° F) (Martinson, 2002).

 Combustion of VOCs for the above technologies relies on a stable ratio between air and fuel for optimal burning efficiency and control of VOCs (Bosch, 1993). These methods unfortunately do not typically work well with lumber kilns due to the nonconstant factors associated with lumber drying, such as variable air flow rate rates, high moisture content in the air stream, stopping and starting of the kilns for loading and unloading, kiln exhaust vents, and economics.

Previous Research

Previous research that has been conducted on VOC emissions from kiln drying SYP (Ingram et al., 1993, Punsuvon, 1994, Thompson, 1996, and Shmulsky, 1998) focused on the quantification of drying emissions from SYP, particularly with regard to age, species, heartwood to sapwood ratio, density, and end grain exposure. High and low temperature drying schedules yielded 5.85 and 5.93 pounds per 1,000 board feet of VOC emissions, with lumber free of heartwood and knots having 3.28 pounds per 1,000 board feet of VOC emissions, with primarily heartwood lumber having 7.41 to 16.6 pounds per 1,000 board feet of VOC emissions (Ingram et al., 2000). End grain influence was found to be not statistically significant in regards to VOC emissions (Shmulsky, 2000). Seasonal influence was found to be statistically significant (Shmulsky 2000). Lumber dimensions were not found to be statistically significant in regards to VOC emissions (Shmulsky, 2000). Initial research on VOC reduction (Banerjee, 1996, Shmulsky, 1998) examined alternative techniques to reduce VOCs. Steaming lumber prior to drying was not found to be statistically significant in regards to VOC emissions. Drying the lumber via radio frequencies (RF) was not found to be statistically significant in regards to VOC emissions (Shmulsky, 2000).

CHAPTER III MATERIALS AND METHODS

Overview

The primary research method (heated copper) on reducing VOCs began in February, 2006 using a heated steel reactor containing copper as the catalytic metal. The second research method (hydrogen peroxide and UV light) began earlier in May, 2005 using a wet hydrogen peroxide scrubber with UV light. The third research method (Fenton's reagent) began in August, 2005 using a wet hydrogen peroxide scrubber with ferrous sulfate.

Heated Copper

Pilot Scale Kiln

A pilot-scale kiln measuring thirty-two inches (81.3 centimeters) in length, thirtyfive inches (88.9 centimeters) in height, and twenty-eight inches (71.1 centimeters) in depth was used to kiln dry the SYP green lumber to below nineteen percent moisture content. Oregon State University and the National Council for Air and Stream improvement (NCASI) have shown that the emissions from pilot-scale kilns reasonably estimate the emissions from commercial scale dry kilns (Lavery and Milota, 2000) (NCASI, 2002). The kiln is equipped with a 12-inch (30.48 cm) diameter variable speed

fan. The incoming and exiting air is controlled via computer based on the wet and dry bulb temperature of the kiln, as well as the current time of the run. The kiln is equipped with a single exhaust vent to allow analysis of the VOC emissions.

The dry bulb temperature of the kiln is monitored and regulated via a thermocouple and associated controller. The wet bulb temperature of the kiln is monitored and controlled via a thermocouple with a cotton sleeve. Part of the cotton sleeve is immersed in a bath of water such that the sleeve wicks-up water around the temperature probe to produce a wet bulb reading.

The pilot-scale kiln lumber capacity is fifteen pieces of rough two by six inches (5.1 to 15.2 cm) nominal lumber twenty-three inches (58.4 centimeters) in length, approximately 28.8 board feet. Two metal stickers were placed between each layer of wood to facilitate air flow for uniform drying. The lumber is stacked three wide by five high in the kiln.

Schematic of pilot-scale kiln and lumber setup

Reactor

The VOC reactor was built from a 3.5-inch (8.89 cm) diameter, four foot (121.9 cm) long section of steel pipe. Seventy feet (21.3 m) of 0.625-inch (1.58 cm) diameter copper tube was placed in the steel pipe to act as the catalyst for VOC reduction. To heat the reactor, a twenty-foot (6.1 m) long, two-inch (5.08 cm) wide, medium temperature, silicon rubber heat tape rated to 260° C (500° F) was obtained from Omega Engineering. An Omega T-type thermocouple was used to measure the temperature inside of the reactor. The thermocouple was located in the middle of the length of the pipe. The heat

tape was wrapped around the pipe but was not in direct contact with the thermocouple. A layer of R-13 fiberglass insulation was wrapped around the reactor. A second layer of R-4 Reflectix aluminum insulation was wrapped around the first layer of insulation. An Omega CN9120A temperature controller was used to control the amount of current supplied to the heat tape. An Omega solid state relay, SSRL240DC50, was utilized for current switching between the temperature controller and the heat tape to provide the necessary amperage power needed to control the heat tape. An Omega, FHS-6 heat sink was placed on the solid state relay to provide the necessary cooling for the unit. Black steel piping and fittings were installed throughout the reactor and exhaust piping. Teflon tape was used on fittings to prevent air leaks.

The reactor was tested at four different settings: control $(21^{\circ} C (70^{\circ} F)$ ambient temperature), 100°, 200°, and 240° C settings. Three runs were conducted for each reactor setting. No current was applied to the heat tape in the control runs. In the 100°, 200°, and 240° C runs, electricity was applied to the heat tape prior to the start of the run to heat the reactor. An additional run was conducted with the copper tubing removed from the steel reactor and without any current applied to the heat tape to note any differences in VOC emissions.

Figure 5

Reactor setup

Analytical

The testing procedure was conducted based on the NCASI – Standard Protocol for the VOC Concentration Measurement Method for Use at Small-Scale Kilns. This method is based on the Environmental Protection Agency (EPA) Method 25A – Determination of Total Gaseous Organic Concentration Using A Flame Ionization Analyzer. The modified method was written specifically for small scale kiln exhaust emissions categorizations. Two J.U.M. Engineering VE-7 total hydrocarbon analyzers were used to measure the amount of emitted volatile organic compounds in the air stream. The J.U.M. VE-7 meets the requirements of the EPA Method 25A as well as the NCASI method. The flame ionization detection method is used to determine the concentration of VOCs in the air stream.

Two samples of the air were pulled from the pre- and post-reactor air streams via vacuum to determine the volatile organic compound concentration per million parts of air. The J.U.M. VE-7 can analyze VOCs accurately if the maximum moisture in the air is below twenty percent. To ensure that the maximum moisture content in the air was below twenty percent, a Greenburg-Smith impinger setup immersed in an ice bath was utilized to remove excessive water from the air lines prior to analysis. After the impinger setup, the sample was pulled through a sample line heated to 180° C to prevent condensation of the air stream.

The analyzers were warmed up prior to each run for a minimum of one hour to allow the highly sensitive parts in the analyzer to heat to temperature and function properly. The analyzers were calibrated at the start of each run with gases containing

zero parts of hydrocarbons per million gases for calibrating the zero point of the analyzers, as well as a 3000 parts of hydrocarbons per million gases for calibrating the span. The 3000 parts per million span gases was selected as a reasonable level according to the NCASI standards for determining VOC levels in SYP (NCASI). The calibrated gases were introduced into the sample lines after the impinger setup which allowed for more accurate calibration of the hydrocarbon analyzers.

Analytical setup

Dry Kiln Schedule

A fourteen hour, high temperature schedule often used by industry was selected as the kiln drying schedule. The dry bulb temperature of the kiln was set to 126° C (260° F)

and a heat ramp up schedule was not utilized. The kiln exhausted air at 0.32, 0.74, 1.06, and 1.7 cubic feet per minute based on the wet bulb temperature of the air in the kiln. If the actual wet bulb temperature was below the set wet bulb temperature, minimal amounts of air were exhausted from the kiln. As the wet bulb temperature increased, additional dry makeup air was forced into the kiln (Appendix C).

Southern Yellow Pine Specimens

Green rough SYP lumber measuring two inches by six inches (nominal) was obtained from a local mill in Ackerman, Mississippi, located approximately twenty-five miles from the Forest Products Department at Mississippi State University. Relatively clear lumber was selected off of the green chain after the logs had been sawn to nominal dimension. The length of the green lumber varied between eight and twelve feet. The lumber was wrapped in plastic, and stored in a cold room refrigerated to 1° C (34 $^{\circ}$ F) prior to usage to minimize the amount of moisture lost and prevent insect and fungal attack.

To minimize the effects of the inherit variability that exists within wood, a randomized complete block design was used to build each of the thirteen kiln charges. An eight to twelve foot parent board was cut up into four, twenty-three inch long specimens. The four specimens were divided randomly into the three treatments and control kiln charges. Forty-five parent boards were cut up to build a total of twelve kiln charges for three runs at each treatment level and three control runs. An additional fifteen parent boards were cut up to build the no-copper run, with each parent board contributing one specimen to the kiln charge.

Sample cut of 2 x 6-inch x 8-foot board

The thirteen runs were conducted in a randomized order. Each charge of green lumber averaged 123.5 pounds prior to drying. The target (final) maximum moisture content for each run was 19 percent.

After each run, moisture content samples were cut from each board to determine the overall moisture content of the kiln charge. The samples averaged one inch (2.5 cm) in length and were cut from the center of the boards with a radial arm saw. The samples were weighed collectively to determine the kiln dry sample weight. The samples were placed in a Blue M Electrical Company oven heated to 103° C for approximately twentyfour hours to determine the oven dry weight. The moisture content of the charge was calculated using the kiln dry sample weight and the oven dry sample weight.

Hydrogen Peroxide and UV Light

 The second treatment method (hydrogen peroxide and UV light) was conducted in collaboration with the Institute for Clean Energy Technology at Mississippi State University. The Institute for Clean Energy Technology was primarily responsible for the wet scrubber reactor, UV lamps, and the testing chemicals, while the Forest Products Department was primarily responsible for the SYP specimens, the analytical instrumentation, and the operation of the dry kiln. The research utilized the pilot-scale kiln, analytical instrumentation, and SYP specimens as discussed previously in the primary research materials and methods section. The reactor and kiln schedule differed from the primary research.

Reactor

A wet scrubber was built of stainless steel and Plexiglas for facilitation of the hydrogen peroxide and UV light research. The pre-reactor air sample was taken immediately after the kiln and prior to the exhaust entering the wet scrubber. The kiln exhaust flowed into the wet scrubber at the bottom of the reactor and the kiln exhaust exited the reactor at the top of the unit. The post-reactor air sample was taken after the wet-scrubber.

The wet scrubber had a water and hydrogen peroxide solution at the bottom of the tank circulated via a water pump and sent through a water filter to remove any solid particles in the solution. The solution was then cooled in an ice bath before being recirculated to the top of the wet scrubber and sprayed into the reactor. A UV light was installed in the middle of the reactor. A glass marble and steel wool bed was installed

below the UV light and above the water and hydrogen peroxide solution to increase the surface area for the VOCs to react with the hydrogen peroxide and the UV light. The pH of the hydrogen peroxide solution was adjusted using sodium hydroxide (NaOH) due to its non-reactivity with the free hydroxyl radicals. The hydrogen peroxide was acquired from United States Peroxide. The initial concentration of the solution was thirty-five percent hydrogen peroxide. The solution was then further diluted with de-ionized water to a variety of concentrations to determine the most effective concentration for VOC reduction.

Figure 8

Wet scrubber – Hydrogen peroxide and UV light

Dry Kiln Schedule

A high temperature drying schedule was utilized with a dry bulb temperature of 112° C (235° F) and an initial heat ramp up time of three hours. The research focused primarily on the peak of VOC emissions that occur after the initial heat ramp up period. The wood was not dried for any specific period of time or to any specific moisture content due to the large variety of testing variables that were researched (Appendix D).
Fenton's Reagent

The third treatment method, Fenton's reagent, using hydrogen peroxide and ferrous sulfate, was also conducted in collaboration with the Institute for Clean Energy Technology at Mississippi State University. The wet scrubber, as used in the hydrogen peroxide and UV light research, was used in the Fenton's reagent research but the UV light was removed from the setup. An additional nozzle was installed on the top of the reactor for the addition of the ferrous sulfate solution. The dry kiln schedule is the same as in the hydrogen peroxide and UV light treatment method.

Figure 9

Wet scrubber – Fenton's reagent

Chapter IV

RESULTS AND DISCUSSION

Heated Copper

 Primary research (heated copper) on VOC reduction using the heated copper reactor began in February 2006. VOC emissions were compared for both the pre- and post-reactor air streams to determine the overall reduction in pollutants emitted. A comparison between the treatments was made using a mixed model in a statistical analysis program, SAS version 9.1.2.

Drying Results Overview

The average green weight prior to drying was 123.5 pounds. The average kiln dry weight was calculated as 68.8 pounds. The average run time was fourteen hours and eight minutes. The average green moisture content percent was calculated as 109 percent, with an average final moisture content of sixteen percent.

Table 1

Overall drying results

Complications

 A four-impinger setup per air stream was used in the first four runs, one at each treatment, (control, 100°C, 200°C, and 240°C). The pre- and post-reactor sample lines were not necessarily pulled at the same rate due to possible air leaks within the impinger train which potentially caused inaccuracies in the data. Due to the problems identified with the first four runs, a one impinger setup per air stream was used in the last eight runs as well as the no copper run.

Another complication for data analysis arose at the beginning of each run. The pre-reactor sample, located relatively close to the kiln, showed VOC emissions almost immediately during each run. The post-reactor sample, located relatively far away from

the kiln, did not have any observed VOCs until approximately one hour into the run. This difference suggests that some VOCs likely condensed during the first hour. After approximately one hour, the exhaust piping heated up enough such that the condensation of the VOCs out of the air stream ceased. To correct for the condensed VOCs, an adjustment was made based on the percent of the difference observed in the one impinger control runs. The average difference between the one impinger post-reactor and the prereactor control runs was eleven percent. All of the post-reactor PPM data was multiplied by 1.11 percent to adjust for the imbalance.

Run One – Control

The first control run drying time was fourteen hours and thirty-six minutes. The green weight of the charge was 126.5 pounds while the kiln dry weight was 69 pounds. The oven dry weight of the wood was 58.4 pounds. The final moisture content of the charge was 18.1 percent. The average air flow rate of the kiln was 1.35 cubic feet per minute. The pre-reactor air stream emissions were 3.65 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 4.47 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 4.96 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were thirty-six percent higher than the pre-reactor emissions.

Run One – Control

Run Two – Control

The second control run drying time was fourteen hours and thirty minutes. The starting weight of the charge was 125.5 pounds while the final weight was 70.3 pounds. The oven dry weight of the wood was 62.6 pounds. The final moisture content of the charge was 12.2 percent. The average air flow was 1.3 cubic feet per minute. The prereactor air stream emissions were 6.11 pounds of VOCs per 1,000 board feet. The postreactor average air stream emissions were 5.60 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 6.22 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were one percent higher than the pre-reactor emissions.

Run Two - Control

Run Three – Control

The third control run drying time was fourteen hours and six minutes. The starting weight of the charge was 119 pounds while the final weight was 69 pounds. The oven dry weight of the wood was 59.1 pounds. The final moisture content of the charge was 16.7 percent. The average air flow was 1.14 cubic feet per minute. The pre-reactor air stream emissions were 4.40 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 3.93 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 4.36 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were one percent lower than the prereactor emissions.

Run Three - Control

Figure 12

Run Four - 100° C

The forth run's drying time was fourteen hours. The starting weight of the charge was 127.5 pounds while the final weight was 68.5 pounds. The oven dry weight of the wood was 58.0 pounds. The final moisture content of the charge was 18.1 percent. The average air flow was 1.41 cubic feet per minute. The pre-reactor air stream emissions were 4.63 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 4.98 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 5.53 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were nineteen percent higher than the pre-reactor emissions.

Run Four - 100°

Run Four - 100° C

Run Five - 100° C

The fifth run's drying time was fourteen hours and twenty-eight minutes. The starting weight of the charge was 123.8 pounds while the final weight was 73.0 pounds. The oven dry weight of the wood was 63.0 pounds. The final moisture content of the charge was 15.9 percent. The average air flow was 1.26 cubic feet per minute. The prereactor air stream emissions were 5.11 pounds of VOCs per 1,000 board feet. The postreactor average air stream emissions were 5.17 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 5.74 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were twelve percent higher than the pre-reactor emissions.

Run Five -100° C

Run Six - 100° C

The sixth run's drying time was thirteen hours and fifty-nine minutes. The starting weight of the charge was 118.0 pounds while the final weight was 67.0 pounds. The oven dry weight of the wood was 56.8 pounds. The final moisture content of the charge was 17.9 percent. The average air flow was 1.32 cubic feet per minute. The prereactor air stream emissions were 5.69 pounds of VOCs per 1,000 board feet. The postreactor average air stream emissions were 5.49 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 6.09 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were seven percent higher than the pre-reactor emissions.

Run Six – 100° C

Run Seven - 200° C

The seventh run's drying time was fourteen hours and two minutes. The starting weight of the charge was 129.5 pounds while the final weight was 69.5 pounds. The oven dry weight of the wood was 58.9 pounds. The final moisture content of the charge was 17.9 percent. The average air flow was 1.37 cubic feet per minute. The pre-reactor air stream emissions were 3.69 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 3.81 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 4.23 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were fifteen percent higher than the prereactor emissions.

Run Seven – 200° C

Run Eight - 200° C

The eight run's drying time was fourteen hours and two minutes. The starting weight of the charge was 123.8 pounds while the final weight was 70.5 pounds. The oven dry weight of the wood was 61.0 pounds. The final moisture content of the charge was 15.5 percent. The average air flow was 1.39 cubic feet per minute. The pre-reactor air stream emissions were 4.60 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 3.81 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 4.23 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were eight percent lower than the prereactor emissions.

Run Eight – 200° C

Run Nine - 200° C

The ninth run's drying time was fourteen hours and one minute. The starting weight of the charge was 119.3 pounds while the final weight was 59.0 pounds. The oven dry weight of the wood was 51.0 pounds. The final moisture content of the charge was 15.7 percent. The average air flow was 1.48 cubic feet per minute. The pre-reactor air stream emissions were 6.06 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 5.51 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 6.12 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were one percent higher than pre-reactor emissions.

Run Nine – 200° C

<u>Run Ten – 240 $^{\circ}$ C</u>

The tenth run's drying time was fourteen hours and four minutes. The starting weight of the charge was 128.3 pounds while the final weight was 69.8 pounds. The oven dry weight of the wood was 59.0 pounds. The final moisture content of the charge was 18.3 percent. The average air flow was 1.3 cubic feet per minute. The pre-reactor air stream emissions were 3.60 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 4.17 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 4.63 pounds of VOCs per 1,000 board feet. The post-reactor emissions were twenty-nine percent higher than the prereactor emissions.

Figure 19

Run Ten -240° C

Run Eleven - 240° C

The eleventh run's drying time was thirteen hours and fifty-nine minutes. The starting weight of the charge was 123.0 pounds while the final weight was 70.0 pounds. The oven dry weight of the wood was 51.6 pounds. The final moisture content of the charge was 13.6 percent. The average air flow was 1.45 cubic feet per minute. The prereactor air stream emissions were 4.30 pounds of VOCs per 1,000 board feet. The postreactor average air stream emissions were 3.56 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 3.95 pounds of VOCs per 1,000 board feet. The post-reactor emissions were eight percent lower than the prereactor emissions.

<u>Run Twelve - 240 $^{\circ}$ C</u>

The twelfth run's drying time was thirteen hours and fifty-nine minutes. The starting weight of the charge was 121.0 pounds while the final weight was 70.0 pounds. The oven dry weight of the wood was 51.0 pounds. The final moisture content of the charge was 16.5 percent. The average air flow was 1.39 cubic feet per minute. The prereactor air stream emissions were 6.33 pounds of VOCs per 1,000 board feet. The postreactor average air stream emissions were 5.40 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 5.99 pounds of VOCs per 1,000 board feet. The adjusted post-reactor emissions were five percent lower than the pre-reactor emissions.

Run Twelve – 240° C

Run Thirteen – No Copper

The thirteenth run's drying time was fourteen hours and ten minutes. The starting weight of the charge was 120.0 pounds while the final weight was 68.5 pounds. The oven dry weight of the wood was 58.5 pounds. The final moisture content of the charge was 17.0 percent. The average air flow was 1.59 cubic feet per minute. The pre-reactor air stream emissions were 7.56 pounds of VOCs per 1,000 board feet. The post-reactor average air stream emissions were 6.92 pounds of VOCs per 1,000 board feet. The adjusted post-reactor average air stream emissions were 7.68 pounds of VOCs per 1,000 board feet. The post-reactor emissions were two percent higher than the pre-reactor emissions.

Figure 22

Run Thirteen – No Copper

Statistical Analysis – All Runs

Statistical analysis was performed on the data collected in all of the runs using both the four-impinger and one-impinger setups. An analysis of variance (ANOVA) model comparing the treatment means was conducted using the mixed procedure in SAS using type III sums of squares. The impinger setup and the treatment method were included in the model as class variables. The blocking factor was included as a random effect. The response variable was the percent reduction observed after adjusting the postreactor hydrocarbon analyzer compared to the pre-reactor analyzer. The interaction between impinger levels and treatments was not statistically significant, p -value = 0.9176 was calculated. Temperature was not statistically significant, p -value = 0.7558. The impinger setup was not statistically significant, p -value = 0.08.

Table 2

PROC MIXED - ANOVA summary

Statistical Analysis – One-Impinger Runs

 Statistical analysis was performed on the data collecting in the runs using only the one-impinger setups. An ANOVA model comparing the treatment means was conducted using the mixed model procedure in SAS. The treatment method was included in the model as a class variable. The blocking factor was included as a random effect. The response variable was the percent reduction observed after adjusting the post-reactor

hydrocarbon analyzer compared to the pre-reactor analyzer. The treatment methods were not statistically significant in regards to VOC emissions, p-value = 0.1674. The temperature of the reactor did not statistically affect VOC emissions at the five percent significant level.

Table 3

PROC MIXED - One-impinger ANOVA summary

Analysis of Treatment Means

 The analysis of treatment means was completed with the one-impinger setup comparing the post and pre-reactor VOC emissions. The post-reactor VOC emissions were similar to the pre-reactor VOC emissions in the control runs. In the 100^o C runs, the post-reactor VOC emissions were ten percent higher than pre-reactor VOC emissions. In the 200° C runs, the post-reactor VOC emissions were four percent lower than prereactor VOC emissions. In the 240° C runs, the post-reactor VOC emissions were seven percent lower than pre-reactor VOC emissions. There was no statistical difference between treatment means.

Table 4

Average treatment reduction

Visual Analysis on Copper Tubing

 Following the completion of the runs, the copper tubing used in the reactor was examined to note any physical and chemical changes that may have occurred during the tests. A black powdery material, likely copper oxide or graphite, formed on the surface of the tubing. The powdery material was present on the copper tubing approximately one foot from the top of the reactor and continued to the bottom of the reactor.

Air Dilution

More accurate data would likely have been obtained if the sample line was diluted prior to J.U.M. VE7 analyzers. Without dilution, the moisture in the air of the samples lines exceeds the twenty percent allowed by the J.U.M. VE7 analyzers. The saturation of air in the sample lines requires the use of the impingers before analysis, which introduced problems with air leaks and different flow rates. Diluting the air would result in less moisture going into hydrocarbon analyzers which would eliminate the impinger setup. Moisture has been shown to reduce the readings on the VOC analyzers with up to twentyone percent decrease in VOC readings if the sample line has high moisture in the air (Milota and Lavery 1998).

Comparison between Control and 240° C Runs After Peak VOC Release

A comparison between the control and the 240° C runs after the peak concentrations of VOCs illustrates some of the effectiveness that the heated reactor is having at the 240° runs. The pre- and post-reactor PPM values are similar in the control runs, while the post-reactor PPM values are slightly lower than the pre-reactor PPM values to the end of the run. This difference may account for the average reduction of seven percent that was observed in the post-reactor emissions in the 240° C runs compared to the pre-reactor emissions.

Run Three - Control After Peak

Figure 23

Run Three – Control after peak

Run Twelve - 240° C After Peak

Run Twelve – 240° C after peak

Hydrogen Peroxide and UV Light

The second treatment method, (hydrogen peroxide and UV Light) began in May 2005 in collaboration with the Institute for Clean Energy Technology utilizing a variety of concentrations of hydrogen peroxide solution in combination with either a 12 or a 450 watt UV bulb.

Table 5

Hydrogen peroxide and UV light summary

 Numerous variables were tested with the hydrogen peroxide and UV light without control runs so no statistical analysis was done on the data. A planned experiment was not conducted due to the collaboration with the Institute for Clean Energy Technology. The data show a variable amount of reduction for each run with the maximum reduction being sixteen percent while the lowest reduction was an increase in VOCs by twenty percent. The average reduction in VOCs was one percent. This data show no clear trend in VOC reduction over the multitude of variables tested.

Fenton's Reagent

The third treatment method using Fenton's reagent began in September 2005 using hydrogen peroxide and ferrous sulfate based on the initial non-success found in the hydrogen peroxide and UV lamp results. A variety of concentrations of hydrogen peroxide and ferrous sulfate were tested to determine the optimal amount for VOC reduction. The maximum reduction observed was twenty-five percent while the minimum reduction observed was an increase in VOCs by eight percent. The average reduction in VOCs was five percent.

Table 6

			Pre-Reactor	Post-Reactor	%
Run	H_2O_2	FeSO ₄	PPM	PPM	Reduction
	5%	1%	2141	1801	16%
2	5%	1%	2100	2276	$-8%$
3	5%	1%	1523	1143	25%
4	5%	1%	2148	2260	$-5%$
5	10%	2%	1116	1168	$-5%$
Average					5%

Fenton's reagent summary

The data shows that a measurable reduction occurred in two of the runs while no reduction occurred in three runs. Other bench top runs, conducted at the Forest Products Laboratory by David Strobel, Research Associate III, however indicated that when ferrous sulfate is initially added to the hydrogen peroxide solution a large amount of oxygen gas is created which seems to prevent any air flow through the emissions stream into the hydrocarbon analyzer. This air flow restriction results in an erroneous reading of approximately zero parts per million on the hydrocarbon analyzers.

Chapter V

CONCLUSIONS

Heated Copper

 The heated copper treatment method involved the use of a steel reactor containing copper tubing heated to 100º, 200º and 240º C to determine VOC differences amongst air samples taken pre- and post-reactor. A total of twelve runs were conducted and analyzed. Four runs used a four-impinger setup and nine runs used a one-impinger setup. An additional runs was conducted without any copper in the steel reactor, this run was similar to the control runs. Temperature did not statistically affect VOC emissions in either the four impinger or one impinger runs. The average reduction of VOCs at the 240° C treatment method during the one impinger runs was seven percent.

Table 7

Average treatment reduction

 \parallel

A variable to explore would be using fresh copper for each run to ensure the VOCs have a clean surface of copper to react to. The majority of the copper tubing was coated with a back, powdery material after being analyzed after the final run, this may have limited the amount of reduction that was observed in the latter runs.

Further experiments conducted at elevated temperatures may result in further reduction of VOCs in the post-reactor air streams which may result in a statistical difference between the treatment means and the control runs. Moving the post-reactor air sample closer to the reactor exit would likely result in more accurate data and would likely prevent condensation of VOCs before sampling.

The average VOC emissions found in the drying runs was 5.8 and 6.1 pounds of VOCs per 1,000 board feet of VOC emissions for the pre-reactor and adjusted postreactor air streams. The average VOC emissions are slightly higher than the common emission values of SYP lumber, typically between 3 and 4 pounds of VOCs per 1,000 board feet.

Hydrogen Peroxide and UV Light

 The use of liquid hydrogen peroxide and UV light does not show promise for use with reducing VOC emissions emitted during lumber drying. The treatment method has worked successfully in treating water pollution but it did not work well with the air pollutants released during kiln lumber drying. A potential solution may be to vaporize the hydrogen peroxide prior to injecting it into the reactor. This would resolve the phase differences between the liquid hydrogen peroxide solution and the gaseous air stream.

If vaporization of the hydrogen peroxide is researched in the future, worker safety must be taken into account before any new technologies be implemented. Hydrogen peroxide, at high concentrations is fairly hazardous, particularly when heated up and vaporized. The high concentrations can occur due to the higher boiling point of hydrogen peroxide compared to water.

A steady supply of hydrogen peroxide must exist at the plant, which would typically involve the use of an on-site hydrogen peroxide reactor. This factor along with the energy needed to run the reactor along with any preventative maintenance that must be performed will add to the price of the lumber.

Fenton's Reagent

The use of Fenton's reagent, specifically hydrogen peroxide and ferrous sulfate, does not show promise for reducing the VOC emissions from lumber kilns. The reaction that occurred between the hydrogen peroxide and the ferrous sulfate generated a large amount of off gas that restricted the flow of the kiln exhaust gas but did not reduce VOCs.

A steady supply of hydrogen peroxide must exist at the plant, which would typically involve the use of an on-site hydrogen peroxide reactor. This factor along with the energy needed to run the reactor along with any preventative maintenance that must be performed will add to the price of the lumber. The Fenton's reagent chemicals used in the study were relatively expensive compared to the price of the lumber, which may make it non-feasible for industrial use.

Overall

One issue that must be resolved before any treatment solution could be implemented into the forest products industry is the overall cost of the technology and any safety hazards that may exist within the operation. The treatment of VOCs must be economically feasible due to lumber being a commodity product as well as relatively easy to use. Developing an expensive treatment method and having the government regulate VOC emissions more strictly may result in the price of the lumber increasing enough so that it is non-competitive with other sources of lumber, mainly imports from Canada. The primary research with the heated copper is relatively simple and inexpensive compared to the hydrogen peroxide and UV light, as well as the Fenton's reagent treatment methods, and could potentially be further improved upon for industrial application.

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APPENDIX A

DRY KILN SCHEDULE:

HEATED COPPER
Kiln Schedule A

If Time $<$ 21600 Seconds, $t = a$ If Time \ge 21600 Seconds, t = b

When $t = a$ If Wet Bulb $>= 198$ then air flow rate $= 1.7$ CFM If $198 >$ Wet Bulb \ge 197 then air flow rate = 1.06 CFM If $197 >$ Wet Bulb $>= 196$ then air flow rate $= .74$ CFM If Wet Bulb \leq 196 then air flow rate = .32 CFM When $t = b$

If Wet Bulb $>= 180$ then air flow rate $= 1.7$ CFM If $180 >$ Wet Bulb $>= 179$ then air flow rate $= 1.06$ CFM If 179 > Wet Bulb >= 178 then air flow rate = .74 CFM If Wet Bulb \leq 178 then air flow rate = .32 CFM

APPENDIX B

DRY KILN SCHEDULE:

HYDROGEN PEROXIDE AND UV LIGHT, FENTON'S REAGENT

Kiln Schedule B

- If (Wet Bulb $>= 180$) then air flow rate $= 1.6$ CFM;
- If (Wet Bulb = 179) then air flow rate = 1.2 CFM;
- If (Wet Bulb = 178) then air flow rate = 1.1 CFM;
- If (178 > Wet Bulb > = 175) then air flow rate = 1.0 CFM;
- If (Wet Bulb = 174) then air flow rate = $.8$ CFM;
- If (174 > Wet Bulb >= 169) then air flow rate = .7 CFM;
- If (169 > Wet Bulb >= 166) then air flow rate = .6 CFM;
- If (Wet Bulb \le 165) then air flow rate = .5 CFM;

APPENDIX C

FORMULAE

Moisture Content % = (kiln dry weight – oven dry weight) / oven dry weight $*$ 100

Wood Weight = Post Drying Weight / (1 + Moisture Content % / 100)

Weight of Water = Green Weight – Wood Weight

% Reduction = $1 - ($ Post Reactor # VOC Emissions / Pre Reactor # VOC Emissions $)*$

100

APPENDIX D

PPM CONVERSION TO VOC POUNDS PER 1,000 BOARD FEET

APPENDIX ${\bf E}$

SAS CODE

OPTIONS PS=**55** LS=**85** NODATE; **Data** VOC1;

Input Run trt impinger block adjred; Cards;

proc mixed;

model adjred = trt impinger trt*impinger; random block; **run**;

Data VOC2;

Input Run trt block adjred; Cards[:]

proc mixed;

model adjred $=$ trt; random block; **run**;