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A Review of Double-Diffusion Wood Preservation Suitable for Alaska

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Abstract

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Currently, all treated lumber used in Alaska is imported from the 48 contiguous states and Canada because there are no wood-treating facilities in Alaska. This report explores conventional and alternative wood-treating methods and reviews previous studies and laboratory tests on treated wood. In investigating wood treatment as a possible processing option for Alaska forest products manufacturers, the double-diffusion method of using sodium fluoride followed by a copper sulfate appeared to be the most advantageous approach. This method of treating wood was identified because it can be used to treat freshly cut or green wood. This was an important factor to consider, owing to the limited drying capacity in Alaska. Little information was available as to the chemical retention after treating and its resistance to leaching.

Keywords: Wood preservation, double-diffusion, Alaska, nonpressured treatment, lumber, copper sulfate.

Contents

Introduction

For decades, the wood industry in Alaska focused on producing cants and chips for export to Asia (USDA FS 1999). Because exporting rules and the Asian market have changed drastically, producers in Alaska are looking into other uses for their wood. Currently, all treated lumber used in Alaska is imported from the 48 contiguous states and Canada because there are no wood-treating facilities in Alaska. Over 10 million board feet (mmbf) of treated lumber are imported every year (McDowell Group 1998).

There are many methods for treating wood. The conventional method for treating wood uses combinations of vacuum and pressure to force chemical into the cell lumens (Zabel and Morrell 1992). Alternative wood-treating methods are nonpressure processes that include brushing, spraying, dipping, and many variations of soaking (Hunt and Garratt 1967). Each method has its strengths and its weaknesses, and has different equipment requirements and chemicals. Treating schedules have been fully developed for some methods and chemical combinations, but some processes have been less thoroughly examined.

The double-diffusion method of treating wood was identified because it can be used to treat freshly cut or green wood.

The double-diffusion method of treating wood was identified because it can be used to treat freshly cut or green wood. Owing to the limited drying capacity in Alaska, this was an important factor to consider. The double-diffusion method is based on sequentially treating green wood in two aqueous chemical solutions that react within the wood matrix to form a precipitate that is highly resistant to leaching and toxic to fungi. Sodium fluoride and copper sulfate are potential components for this process because each chemical could be shipped in crystalline form to producers, and neither is labeled as a restricted-use pesticide. The literature advocates the use of sodium fluoride (Baechler 1963) in the double-diffusion process; however, this use is not included on the sodium fluoride label currently registered with the U.S. Environmental Protection Agency (EPA). The copper sulfate label shows that EPA allows it to be used in wood treatment and requires a first solution of sodium salt or sodium chromate (Griffin 1997). The label indicates that the wood is to be sequentially soaked in each solution for up to 3 days, without regard to wood species or retention.

The lack of information on double-diffusion treatment by using sodium fluoride and copper sulfate led to investigating the effect on chemical retention of treatment times of up to 3 days in each solution. Because chemicals that diffuse into the wood matrix could leach out during service, the extent of such leaching was also investigated. During this work, the potential for selective chemical absorption from the solution to the wood, the migration of chemicals after a 30-day diffusion period, and the location of chemicals after leaching were also examined. Results of this investigation are part of a collective effort to bring useable technical information to Alaska forest product manufacturers about a wood treating method that will successfully treat locally grown species.

Literature Review

Wood Preservation

Wood preservation has existed for millennia. Wooden ships needed protection from marine borers and decay fungi. Initially, shipbuilders used wood that had natural durability against biotic attack. As the availability of those species lessened, shipbuilders looked for treatments that could preserve the wood or at least extend the service life of a ship until it reached its destination. Today, wood preservation plays an important role in our lives. Treated wood is used in foundations, decks, playgrounds, fences, utility poles, railroad ties, and a host of other applications (Zabel and Morrell 1992).

The amount of treatment depends on the level of protection needed. Decay risk, length of service life, cost of treatment, and end-of-life disposal are all considered when determining which treatment method and chemicals to use. There are short- and long-term levels of protection. Short-term protection, such as dipping in a chemical, is used to minimize sapstain damage on fresh-cut lumber. Long-term protection is used to extend the service life of wood used as an end product. Long-term protection is further divided into aboveground or ground-contact levels of protection. Wood in contact with the ground requires more treatment because the decay risk is higher. There are a variety of methods for delivering chemicals into the wood (Zabel and Morrell 1992).

Treated Wood in Alaska

Market—

The McDowell Group (1998) estimated that the market for treated lumber in Alaska was 10 to 15 mmbf per year. Demand for treated dimensional lumber was heaviest in southeast Alaska, which represented 25 to 30 percent of that estimate. The Alaska Railroad (2003) also uses an additional 2 to 3 mmbf of treated railroad ties every year. Since 1996, they have replaced 520,000 ties, and there are plans to add track to the system.

All the treated lumber used in Alaska is imported from the 48 contiguous states and Canada because there are no wood-treating facilities in Alaska. For decades, the wood industry had focused on producing cants and chips for export to Asia (USDA FS 1999). Because exporting rules and the Asian market have changed drastically, producers in Alaska are looking into other uses for their wood. One of those other uses could be treated wood products.

Raw material—

Much of Alaska is publicly owned. Therefore, mills are dependent upon public lands, mainly the Tongass National Forest, for their timber supply. In fiscal year 2003, 115 mmbf were offered for sale.[1] The amount available each year is subject to change owing to legislative and political issues. The uncertainty of the supply from year to year limits the amount of credit banks are willing to extend to mills. Thus, mills trying to adapt to changing markets are hindered by the lack of capital available for investing in manufacturing equipment.

Alaska has four commercial softwood species: Sitka spruce (*Picea sitchensis* (Bong.) Carr.), white spruce (*Picea glauca* (Moench) Voss), western hemlock (*Tsuga heterophylla* (Raf.) Sarg.), and yellow-cedar (*Chamaecyparis nootkatensis* (D. Don) Spach). Spruces and hemlocks are used to make dimension lumber. They have very little natural resistance to decay, and therefore, would have to be treated to withstand the moderate decay risk typical of southeast Alaska and the low decay risk in the interior region (Hunt and Garratt 1967, Scheffer 1971). Yellow-cedar is primarily used for decking and other exterior uses because it is naturally resistant to decay.

Sitka spruce and western hemlock are most abundant in southeast Alaska, whereas white spruce is abundant in interior Alaska. Western hemlock is considered moderately difficult to treat, whereas Sitka and white spruce are considered difficult to treat (USDA FS 1999). Therefore, it is possible that a preservation process successfully used to treat Alaska-grown Sitka spruce would also be able to treat white spruce.

Conventional Wood Treating

The conventional method for treating wood uses combinations of vacuum and pressure to force chemicals into the cell lumens. This process produces a deep,

[1] Brink, S. 2003. Personal correspondence. Former deputy regional forester, natural resources, U.S. Department of Agriculture, Forest Service, Alaska Region, 709 W 9th Street, P.O. Box 21628, Juneau, AK 99802.

uniform penetration of chemicals in wood for applications requiring a long, reliable service life in regions with high decay hazards (Zabel and Morrell 1992).

Equipment—

Conventional treating requires treatment cylinders that are typically 2 to 3 m in diameter, and built to handle pressures around 1034 kPa. The length of the cylinder is based on product requirements, but can extend up to 55 m long. These cylinders are supported by pumps, chemical tanks, thermometers, gauges, controllers, piping, valves, a boiler, and wood transporting systems (Hunt and Garratt 1967). Tracks on both the infeed and outfeed of the treating cylinder allow lumber on trams to be rolled in and out of the cylinder. The outfeed area must capture any liquid coming off the treated wood to avoid environmental contamination. In addition, this area is usually covered to avoid rainwater contamination, as all water runoff must be captured and cleaned. The capital investment for this equipment can easily exceed $1 million (Reader 2000).

Chemical combinations—

Conventional treating methods can use a host of waterborne or oilborne chemical combinations for treating. Many of these chemicals are listed as restricted-use pesticides by the EPA, meaning they can only be applied by certified pesticide applicators (Zabel and Morrell 1992).

Presently, the waterborne chemicals that are commercially used include chromated copper arsenate (CCA), ammoniacal copper quaternary (ACQ), copper azole (CA), and ammoniacal copper zinc arsenate (ACZA). Copper and arsenic are both excellent fungicides, and arsenic also protects wood from insects and marine borers (Zabel and Morrell 1992). Chromium bonds with the lignin inside the wood matrix as well as forming a complex with the copper and arsenic, thereby limiting the leaching of chemicals while the wood is in service (Hartford 1986). Some waterborne chemicals can be shipped dry and mixed onsite by using a local water supply, reducing transportation costs, whereas others are shipped as concentrates and diluted onsite. Treating wood with waterborne chemicals leaves the wood surfaces clean and paintable.

Oilborne chemicals, like creosote, have existed the longest and have proven to be very reliable preservatives. Creosote is a byproduct made during the manufacturing of coke that is used for steel production. Other major oilborne wood preservatives include pentachlorophenol, copper naphthenate, and copper-8-quinolinolate.

Most oilborne chemicals are transported to treating facilities as concentrates or, in the case of pentachlorophenol, in solid blocks.

Treatment processes—

Conventional pressure treating is divided into two processes, the full-cell and the empty-cell. The full-cell, or Bethell process, is used when maximum retention is paramount. A vacuum is first used to remove some air from the wood, and then preservative is added while increasing pressure. The empty-cell process is used when limited preservative retentions are needed. This process does not use a vacuum, but pressure is introduced either before the preservative or immediately after the preservative is added. Variations in the pressure applied have been further named as either the Rueping or Lowry empty-cell processes. Both full- and empty-cell processes require dried wood, unless some form of conditioning can be performed in the cylinder prior to treating (Hunt and Garratt 1967).

Lumber or poles can be treated in less than 20 hours depending on specifications and wood species. Because the wood is secured in the cylinder, the environment is controllable. This allows treaters the option to adjust retention and penetration in order to meet end-user specifications.

Pretreatments—

Over the years, there have been many mechanical innovations used to aid preservative penetration including incising, radial drilling, through-boring, and kerfing. Although they have been proven effective in many ground line applications for posts and timber, they are destructive, and care must be taken to maintain required mechanical strength properties. Incising also reduces the aesthetic quality of lumber if a smooth surface is desired.

Refractory issues—

Some commercially important refractory species, such as Douglas-fir (*Pseudotsuga menziesii* (Mirb.) Franco) and spruces, have been excluded from certain end uses because of the inability to attain the required preservative penetration, despite attaining the recommended chemical retention (Baines and Saur 1985). Lebow and Morrell (1993) had mixed results pressure treating Sitka spruce. None of the charges that used CCA achieved the American Wood Preservers' Association (AWPA) specifications for penetration despite incising, whereas 12 of 14 charges that used ACZA met both penetration and retention specifications. Blew et al. (1967) found that pressure-treating wood grown in Alaska offered less protection than treating the same species grown in Oregon. These results were based on retention differences in round and sawn wood in Sitka spruce and other species.

Others factors to consider in Alaska—

Even if enough chemical can be impregnated in Sitka spruce grown in Alaska by using conventional wood-treating processes, there are still other factors to consider. The capital investment in treating cylinders of any size is unfeasible for most Alaskans. The climate in Alaska usually forces mills to close during winter, thus reducing the production time available to help repay any capital investment. The low annual production for any one mill, often less than 1 mmbf per year, results in a high capital cost per unit treated.[2] Transporting chemicals, especially oilborne chemicals, over the marine highway system in southeast Alaska can be very costly, cutting into an already limited profit margin. Because many Alaska wood manufacturers use portable processing equipment, the treating system should also be capable of moving seasonally or as the harvest location changes. Treating fresh-cut wood is required because there is a very limited amount of drying capacity available. Taking into account all of these factors, conventional treating does not appear advantageous for Alaska. Therefore, alternative methods of treating wood ought to be investigated.

The treating system should also be capable of moving seasonally or as the harvest location changes.

Alternative Wood Treating

Alternative wood-treating methods include nonpressure processes such as brushing, spraying, dipping, and many variations of soaking (Hunt and Garratt 1967). Many alternative treating methods require much less equipment than conventional methods and are typically limited to small-scale applications by homeowners and farmers (Zabel and Morrell 1992).

Brushing and spraying—

Typically, oilborne preservatives are used when treating wood by brushing or spraying, but waterborne preservatives can also be used. Penetration via these processes is shallow, and therefore protection is limited. Abrasion or checking can easily break the envelope of protection. Wood has to be dry and warm enough to avoid congealing of the oilborne preservative on the wood surface (Hunt and Garratt 1967).

[2] Kilborn, K. 2002. Personal communication. Former team leader, Alaska Wood Utilization Research and Development Center, 204 Siginaka Way, Sitka, AK 99835.

Dipping and soaking—

Hunt and Garratt (1967) differentiated numerous treatment processes that involved dipping or soaking wood. For example, dipping consists of momentarily immersing wood in a bath of preservative, whereas steeping consists of submerging wood for several days or even weeks in an open container. With steeping, dried wood is treated with waterborne chemicals. Cold soaking is similar to steeping except that wood is soaked in unheated oilborne chemicals for 2 days to 1 week.

The thermal process involves immersing dried wood in successive baths of hot and cool preservative. The purpose of the hot and cool baths is to form a partial vacuum, whereby atmospheric pressure would force the preservative into the wood. Either oilborne or waterborne solutions can be used with this method if the temperature does not cause excessive chemical loss through evaporation. Depending on the standard, the hot bath is around 102 °C and the cool bath around 38 °C. Several variations of this method were patented. In one variation, the wood was heated in a kiln instead of a hot bath and then submerged in the cool bath. The theory of creating a partial vacuum by using hot and cool baths was improved by actually creating a vacuum in an airtight container by exhausting the air with a pump (Hunt and Garratt 1967).

Diffusion methods are similar to steeping in that there is bulk flow of solution into the wood. Yet, the diffusion method has a second mechanism for moving preservative into the wood by using a diffusion period. Wood is wet-stacked for a period in order to facilitate diffusion. The theory of diffusion states that chemicals will move from zones of higher concentration (treating solution) to those with lower concentration (water in wood). Therefore, green wood and waterborne chemicals are used for diffusion treatments. This diffusion method typically involves soaking wood in solutions, but theoretically can extend to the use of pastes and wraps to deliver chemicals into the wood (Hunt and Garratt 1967).

Single-diffusion applications that use boron have been commercially accepted in New Zealand, Australia, and New Guinea for decades, and account for 28 percent of all wood treated in the region (Vinden et al. 1997). Chemicals placed in the wood only by diffusion, however, are susceptible to leaching, because chemicals that diffuse into the wood matrix can easily leach out during service. Products treated by the diffusion method are used in low-hazard building timbers, or out-of-ground contact (Vinden 1990).

The double-diffusion process was developed to overcome leaching issues associated with the single-diffusion processes. In this method, green wood is sequentially soaked in two aqueous chemical solutions that react within the wood matrix to form a precipitate that is highly resistant to leaching and toxic to fungi and termites (Baechler 1953).

Other factors to consider in Alaska—

There are many factors to consider when comparing alternative treatment methods for use in Alaska. Dipping is only recommended as a method to deliver long-term wood protection for wood that has been dried and is impractical to treat by more effective methods. Limited drying capacity in Alaska makes dipping impractical. The schedule for the steeping method recommends steeping 1 day for each 25 mm of material thickness plus 1 more day for good measure, but penetration rarely exceeds 6 mm. The poor penetration and the requirement for dried wood eliminate steeping as a choice for Alaska. The thermal process can attain suitable penetration, but the hot bath temperature may be unattainable or not maintainable in Alaska. In addition, the hot and cool solutions need to be pumped into and out of the treatment tank, or the wood must be moved between two separate tanks. This requires either pumps or equipment to move the wood back and forth between tanks, and equipment to heat the solution. The vacuum method requires a sealed container and only works well with easily treated wood, again precluding refractory species. Diffusion methods use green wood, open tanks, and are the suggested alternative for treating refractory species (Hunt and Garratt 1967). Taking into account all of these factors, the alternative wood treating method of diffusion, particularly double-diffusion, appears most suitable for Alaska.

Double-Diffusion Wood Treating

As previously stated, treating wood by diffusion typically refers to soaking wood, but theoretically can also extend to the use of pastes and wraps to deliver chemicals into the wood. Two mechanisms help move preservative into the wood: bulk flow and diffusion (Greaves 1990, Hunt and Garratt 1967). Bulk flow is considered the initial mechanism of treatment by the diffusion method, and consists of liquid flowing into the wood owing to a pressure difference. The second mechanism is diffusion whereby the chemical absorbed in the bulk flow phase becomes more evenly distributed as it moves from areas of high to lower concentration. This allows the chemicals to penetrate deeper and more uniformly into the wood (Vinden 1990). Baechler (1953) noticed a possible third mechanism: capillary pull. If the water column inside the wood matrix was still continuous, evaporation from the top of the post would draw solution upward. Capillary pull is a form of bulk flow that mimics a tree's natural water transport system; it is limited to extremely green posts, treated upright in a barrel with post tops exposed.

Theory of diffusion—

If the wood is at its highest possible moisture content and there is no interaction with the wood substance, the rate of diffusion of such chemicals should follow Fick's law. This law states that the rate of transfer per unit area of a section equals the negative of the diffusion coefficient times the derivative of the concentration with respect to the space coordinate measured normal to the section (i.e., the direction of diffusion). The rate of diffusion is greatest in the longitudinal direction and lowest in the transverse directions (Vinden 1983).

Mathematical models can help predict real world results, establishing relationships between variables, and optimizing treatment schedules. Models must take into account the moisture content and density of the wood, the interactions between the wood matrix and the preservative, temperature, preservative retention, time and type of wood (heartwood, sapwood, earlywood, and latewood), as well as the concentration of the preservative. Vinden (1984) compared the calculated mathematical models for steady-state and non-steady-state diffusion coefficients for copper ions through saturated samples of Scotspine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* (L.) Karst.), and European white birch (*Betula pendula* Roth). His data indicated that the pathway for diffusion was limited to the area of the free water in the lumens, and that that diffusion ceased below the fiber saturation point. He also found that the steady-state diffusion coefficient for spruce air-dried and resaturated wood was significantly lower than the coefficient for spruce in the green condition. This may be because the pathway for diffusion is slowed by pit aspiration (Flynn 1995) resulting from increased capillary tension caused by the removal of free water in lumens during drying (Siau 1984), highlighting another raw material variable not previously mentioned. Therefore, the coefficients of diffusion differ for green and previously dried wood. Vinden (1984) also found that during the initial or non-steady state diffusion, the coefficient of diffusion will deviate from Fick's law, owing to the number of fixation sites (hydroxyl groups) within the wood matrix. He found that all the fixation sites must be filled before diffusion proceeds. Other researchers have also shown that copper ions fix to the wood matrix (Bland 1963, Cooper 1991, Jin and Archer 1991).

Although explainable in mathematical terms, the numerous variables have a significant impact on the retention and penetration of preservatives. Therefore, pilot studies and chemical retention analyses are still needed.

Equipment—

Treating by double-diffusion requires that the wood be soaked in two chemicals sequentially and then wet stacked for a period of time. Depending on the amount

Table 1—Relative advantages and disadvantages of chemicals used in double-diffusion treatments

Chemical	Advantages	Disadvantages
Copper	More toxic to fungi More economical	More corrosive to tank
Nickel	Less corrosive to tank	Less toxic to fungi Less economical
Zinc	Less corrosive to tank	Less toxic to fungi
Reacting with:		
Arsenic	Restricted use	Restricted use
Boron		Did not form an insoluble precipitate with any metal
Chromium	Restricted use	Restricted use
Fluoride	Consumers familiar (toothpaste)	Did not form an insoluble precipitate with nickel or zinc
Phosphorous	Helps fix copper inside wood	Does not contribute to toxicity

Source: Baechler 1953.

and size of the wood to be treated, the double-diffusion method can require fairly simple equipment. Each chemical could be pumped into and out of one treatment tank, or the wood could be moved between two separate tanks. This requires either pumps or the ability to move the wood back and forth between tanks. The material for the tanks can be stainless steel, wood with plastic lining, or other material depending on the corrosivity of the chemicals employed. Tank size would depend on the product being produced. Fence posts could be treated upright in a barrel, whereas decking would have to be fully submerged. Depending on the amount of wood treated per month or the volume of chemicals used per year, containment equipment around the tanks may be necessary (EPA 1996). Depending on species and moisture content, the buoyancy of wood may make hold-down hardware necessary. As with all wood-treating methods, equipment is needed to transport the wood to and from the treating vessel. Personnel protective equipment, as mandated by the Material Data Safety Sheets for each chemical and the Occupational Safety and Health Administration, is also needed.

Ideally, the two chemicals used in the double-diffusion method will form a precipitate that is highly resistant to leaching and toxic to fungi and termites.

Chemical combinations—

Ideally, the two chemicals used in the double-diffusion method will form a precipitate that is highly resistant to leaching and toxic to fungi and termites. To be toxic and insoluble after forming a precipitate, salts of very strong acids are used with weakly basic metals (Baechler 1953). Baechler (1953) initially reacted nickel, zinc, or copper with chromate, fluoride, arsenate, borate, or phosphate. Advantages and disadvantages for each chemical are given in table 1.

Restricted use is listed as both an advantage and a disadvantage for two chemicals. These chemicals are highly toxic, making personnel training and extra containment equipment essential. The disadvantage would be the costs associated with the added safety measures. The advantage would be the awareness of toxicity personnel would gain from training. Recent efforts to revive double-diffusion as an effective but low-cost treatment option for rural areas have focused on sodium fluoride and copper sulfate (Hoffman 2002a, 2002b, 2002c; Kilborn et al. 2003, [and see footnote 3]; Reader 2000; Wheat et al. 1996).

Chemical labels—

Treaters have to legally abide by the wording on the chemical labels. Chemical labels are proprietary to a given company and have either an EPA registration number or NSF-60 certification. Labels contain information on the uses for which the chemical manufacturer is willing to take liability, based on past research. It is illegal to use pesticides for nonlabeled uses or to use them at levels above or below label recommendations.

The copper sulfate labels from Old Bridge Chemicals, Inc.[4] (2000) and Chem One, Inc. (2000) have the same wording for use in a wood treatment. Both labels are for peeled, green posts treated "butt end down first in the copper sulfate solution for three days, then butt end down in sodium chromate solution for two days, and finally turn the post upside down in the sodium chromate solution for one additional day."

The label for Blue Viking's Copper Sulfate Instant (Griffin 1997) states that the first solution is a solution of sodium salt or sodium chromate. Therefore, sodium fluoride could be used with this product label. It states that green material is soaked in the sodium solution for up to 3 days, and then soaked in the Blue Viking Copper Sulfate Instant solution for up to 3 additional days.

The only registered label found for sodium fluoride states: "For Pesticide Formulation Use: Only in formulation into a fungicide for wood preservation" (Osmose 2002). As the label stands, the term "formulation" precludes the use of sodium fluoride in double-diffusion. This is because the wood is treated sequentially in the two chemical solutions, and the formulation of copper fluoride could not occur until after the chemicals are inside the wood matrix.

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[3] Kilborn, K.; Crawford, D.M.; Lebow, S.T.; Lebow, P.K. 2003. Double-diffusion treatment of Alaska inter-coastal wood species [Poster]. In: Forest Products Society annual meeting; 2003 June; Bellevue, WA.

[4] The use of trade or firm names in this publication is for reader information and does not imply endorsement by the U.S. Department of Agriculture of any product or service.

According to Curtis (2003, see footnote 5), Tyonek's Wood Double-Diffusion Treatment Plant in Kenai Borough, Alaska, had a sodium fluoride label that included atmospheric pressure immersion. Because the plant is no longer operating, the whereabouts and status of that label are unknown. Besides the legal issue with the use of sodium fluoride, there is not enough information on these labels for someone to develop treating schedules based on wood species, retention, and penetration.

Preservative threshold—

The minimum amount of preservative needed to prevent wood decay by selected fungi can be determined by using AWPA Standard E10-01 Soil-Block Method (AWPA 2001b). The standard treats sapwood test blocks of a nondurable conifer (e.g., southern pine [*Pinus* spp.]) or a medium-density angiosperm (e.g., sweetgum [*Liquidambar styraciflua* L.]) with different concentrations of the chemical. A minimum of three species each of brown rot and white rot fungi are required when determining thresholds of new preservatives. Depending on the size of the test blocks and fungi used, the incubation period extends from 8 to 24 weeks. The threshold is then calculated by plotting weight lost after incubation against chemical retention to determine the point where fungal-induced weight loss ceases. Duncan (1958) reported that the threshold for a given preservative changes with wood species even within a genus, e.g., *Pinus*. Therefore, the wood species used in the soil block test should match the wood species in question for the preservative application.

Baechler and Roth (1956) conducted decay tests by using *Neolentinus lepideus* Fr., *Gloeophyllum trabeum (*Pers.) Murr., and *Postia placenta* (Fr.) M. Larsen and Lombard fungi on 19 mm southern pine cubes treated with copper sulfate, zinc chloride, sodium arsenate, sodium borate, sodium fluoride, or sodium dichromate water-borne solutions. The only reference to treating schedules was: "the cubes were treated to refusal with solutions of known concentration." The threshold for copper sulfate and sodium fluoride are given in table 2. Units were converted from pounds per cubic foot (lb/ft^3) to percentage on a weight per weight basis (percent wt/wt) by using a specific gravity for southern pine of 0.51 (USDA FS 1999).

These tests were not standardized nor was it stated how the thresholds were determined. Furthermore, because cubes were treated with only one chemical for

[5] Curtis, K. 2003. Personal communication. Director, Ketchikan Wood Technology Center, 7559 N Tongass, P.O. Box 519, Ward Cove, AK 99928.

Table 2—Threshold concentrations for copper sulfate and sodium fluoride with southern pine sapwood

Fungus	Retention threshold range: Copper sulfate	Retention threshold range: Sodium fluoride
	Percent wt/wt	
Neolentinus lepideus	— – 0.59	0.26 – 0.41
Gloeophyllum trabeum	0.94 – 1.31	.49 – .59
Postia placenta	.96 – 1.67	.49 – .57

— = lower threshold immeasurable.

Source: Baechler and Roth 1956.

each decay test, no inferences can be made about the combined fungicidal effect of copper and fluoride. Copper is almost always used with another biocide, i.e., chromated copper arsenate, ammoniacal copper arsenate, copper naphthenate, and copper-8-quinolinolate. Cowling (1957) presented threshold values for several preservatives inoculated with 18 wood-destroying fungi, including the 3 fungi listed in table 2. The threshold reported for copper (as metal) in copper naphthenate was 0.50 percent wt/wt. This value may be a more accurate threshold assumption for copper in copper fluoride than those listed in table 2.

Panek (1963) immersed southern yellow pine (*Pinus palustris* P. Mill.) poles for 15 minutes to 4 hours in 20 or 30 percent aqueous ammonium bifluoride. Pole conditions after months of air-seasoning were compared to fluoride retentions. A retention of 0.8 kg/m^3 was ascertained as an above-ground fluoride threshold for the outer 25 mm of southern yellow pine poles. The condition of the poles was rated for one of six categories; no visible stain, light, medium, or heavy sapwood stain, incipient decay, or decay. For wood with a specific gravity of 0.51, that threshold could be expressed as 0.16 percent wt/wt (USDA FS 1999). Therefore, 0.16 percent wt/wt could be interpreted as an above-ground fluoride threshold based on visual inspection of poles not in ground contact.

Previous studies—

Other investigations into the treating of wood by using the double-diffusion method were conducted by the U.S. Department of Agriculture, Forest Service. The first double-diffusion study (Baechler 1953) resulted from increased interest in treating fence posts for farm use. In 1941, 100 green southern pine posts were treated in copper sulfate followed by sodium arsenate. After treatment, the posts were dried and installed in a fence post plot in the Harrison Experimental Forest in Mississippi. Eleven years later, only one failure occurred and only a few had decay. Five posts have failed after 22 years (Blew and Kulp 1964), and a total of eight posts had

Table 3—Copper sulfate and sodium fluoride absorptions by jack pine posts treated by the double-diffusion process

	Treating schedule					
	Copper sulfate		Sodium fluoride		Chemical absorption	
ID	Time	Concentrate	Time	Concentrate	Copper sulfate	Sodium fluoride
	Days	*Percent*	*Days*	*Percent*	*- - - - - - - - Percent wt/wt - - - - - - - -*	
A	1	7.95	4	3.2	1.42	1.04
B	2	7.95	4	3.2	2.35	.93
C	2	7.95	7	3.2	2.16	1.27

Source: Baechler 1953.

failed after 29 years (Gjovik and Davidson 1975). The incomplete copies of these reports did not indicate the service life of untreated southern pine posts in this plot. Because 92 percent of the treated posts were sound after 32 years of service, it would be safe to say that the double-diffusion method delivered satisfactory amounts of chemical into the wood matrix.

Laboratory tests were also part of Baechler's (1953) initial study. Jack pine (*Pinus banksiana* Lamb.) posts were treated with copper sulfate, followed by either disodium phosphate or sodium fluoride. Copper sulfate and sodium fluoride absorptions by jack pine posts treated by the double-diffusion process are given in table 3.

Several fence posts were treated by double-diffusion at the Matanuska Experimental Station farm in Palmer, Alaska, in 1954.

Several fence posts were treated by double-diffusion at the Matanuska Experimental Station farm in Palmer, Alaska, in 1954. Species included Alaska-grown white spruce, paper birch (*Betula papyrifera* Marsh), balsam poplar (*Populas balsamifera* L.), and quaking aspen (*Populas tremuloides* Michx.). Posts were treated for 3 days in 8-percent copper sulfate solution and then treated for 3 days in 11-percent sodium chromate solution. After 32 years in service, 100 percent of the aspen, balsam poplar, and white spruce posts were sound, whereas only 58 percent of the paper birch posts were sound. The controls for aspen, balsam poplar, paper birch, and white spruce failed after 9, 4, 7, and 9 years, respectively (Mayer et al. 1995).

Baechler et al. (1959) treated several species of hardwood posts native to the Southeast United States. The wood was completely submerged for treatment to replicate larger scale commercial-type treating, permitting a more efficient utilization of hardwoods than the method of treating upright in a barrel. Treating was conducted at ambient temperature, with only one solution concentration for each chemical used. Treatment times were ½, 1, 2, or 3 days for both tanks. The first tank was zinc sulfate and arsenic acid. The second tank was sodium chromate. Five posts from each treatment and species group were analyzed for chemical retention and penetration. The remaining 25 posts were installed in a test plot at the Whitehall Experimental Forest in Georgia. Chemical analyses showed that sapwood was much more treatable than heartwood, and that "double-diffusion appears to offer considerable promise." After 29 years in service, only 3 of 25 pine posts had failed. The overall service lives for white oak (*Quercus garryana* Dougl. ex Hook.), red oak (*Quercus falcate* Michx.), and yellow-poplar (*Liriodendron tulipifera* L.) for all of the treatment times combined were 16.3, 16.4, and 16.2 years, respectively (Vick and Baechler 1986).

Twelve species of wood grown in Hawaii were treated by double-diffusion in 1960, as a demonstration of the process for local landowners, salesmen, and industry personnel. At the time, commercially treated posts were not readily available and this method appeared feasible. Copper sulfate was used as the first solution, followed by sodium chromate. Freshly peeled posts were treated butt down for 3 days in each solution, by using one barrel for each solution. Discs were cut from the top, middle, and butt after a 2-week diffusion period, and analyzed for chemical retention. Analyses showed that the chemical retention for most of the species were within a satisfactory range, based on the desire to retain equal amounts of each chemical. The demonstration showed promise for a commercial double-diffusion treating operation using Hawaiian species (Smith and Baechler 1961).

Baechler (1963) explicitly told farmers "How to treat fence posts by double diffusion." This report recommended sodium fluoride and copper sulfate as the first and second treatment solutions, respectively.

The double-diffusion process was investigated in the late 1960s for its ability to treat Engelmann spruce (*Picea engelmannii* Parry ex Engelm.), lodgepole pine (*Pinus contorta* Dougl. ex Loud.), and Rocky Mountain Douglas-fir (*Pseudotsuga menziesii* (Mirb.) Franco var. *glauca* (Beissn.) Franco) posts. These species resist conventional treatment. One hundred and twenty-six posts of each species were treated with one of the four treatment combinations given in table 4. Posts were fully submerged in solution for treatment. Sixty posts were analyzed for sapwood thickness, and chemical retention and penetration. The remaining 225 treated posts and 75 untreated posts were installed in a fence post plot at the Central Plains Experimental Range in Colorado (Markstrom et al. 1970).

Markstrom et al. (1970) found that Engelmann spruce and lodgepole pine could be successfully treated based on the average penetration exceeding 19 mm. Full sapwood penetration occurred in Rocky Mountain Douglas-fir, but the average minimum penetration was less than 19 mm for all treatments. They also found that

Table 4—Treatment schedule used in fence post study

		First solution 10 percent copper sulfate		Second solution 13 percent sodium chromate / arsenic acid	
ID	**Pretreatment**	**Time**	**Temperature**	**Time**	**Temperature**
		Days	*Fahrenheit*	*Days*	*Fahrenheit*
A	—	1	Ambient	1	Ambient
B	—	3	Ambient	3	Ambient
C	—	1/3	200°	1	Ambient
D	Incising	1	Ambient	1	Ambient

— = no pretreatment occurred.
Source: Markstrom et al. 1970.

both Engelmann spruce and lodgepole pine could be treated to meet the 6.41 kg/m^3 of CCA retention specified by AWPA Standard C5-00 fence posts (AWPA 2001a).

Thirty years later, all of the treated posts withstood a 22.7-kg load applied laterally at the top of the post. Untreated posts had service lives of 16, 17, and 9 years for Engelmann spruce, lodgepole pine, and Douglas-fir, respectively. All untreated posts failed at or near the ground line (Markstrom and Gjovik 1999).

By 1985, double-diffusion studies were extended to treating railroad ties in an effort to demonstrate the use of nonpressure processes to treat native Alaska species. Western hemlock ties and timbers, and Sitka spruce and yellow-cedar timbers were in the combinations shown in table 5.[6]

The first solution was heated for half of the charges. The goal of heating to 82 to 88 °C was unattainable; the actual temperature never exceeded 52 °C. Treating with a heated solution is referred to as the modified double-diffusion process (see footnote 6). Forty-eight hemlock ties went into the railroad track near Palmer, Alaska, and are still in the track (see footnote 2). The remaining wood was to be analyzed for chemical retention and penetration, but no reports of the results were found.

To increase retention with the double-diffusion process, the use of ultrasonic energy was investigated. Alaska-grown white spruce was treated with approximately 4 percent sodium fluoride while ultrasonic energy was applied. Wheat et al. (1996) found that using ultrasonic energy during treatment increased chemical retention. However, the second treatment in the double-diffusion process was not

[6] Gjovik, L.R. 1985. Double-diffusion treatment study for southeast Alaska. Study 3-85-37. 9 p. Unpublished progress report 1. On file with: USDA Forest Service, Forest Products Laboratory, One Gifford Pinchot Drive, Madison, WI 53726.

Table 5—Treatment schedule used in the Alaska demonstration project

Chemical	Concentrate	Temperature
	Percent	
First solution:		
Sodium fluoride	4	Ambient
Sodium fluoride	4	Hot
Copper sulfate	8	Ambient
Copper sulfate	8	Hot
Second solution:		
Copper sulfate	8	Ambient
Copper sulfate	8	Ambient
Sodium chromate/arsenic acid	11	Ambient
Sodium chromate/arsenic acid	11	Ambient

Source: Gjovik 1985 (see footnote 6).

used. Therefore, it is not known if the additional chemical uptake would remain in the wood matrix during submersion into the second treatment solution.

In 1995, the Wood in Transportation Program awarded Tyonek Native Corporation a grant to develop a double-diffusion treating facility. The facility would, in turn, use locally grown species of wood and provide long-term employment for local residents. Operations began in 1997, by treating wood for a bridge to be built near Fairbanks, Alaska (Russell and Kilborn 1997). However, operations ceased shortly thereafter owing to lack of infrastructure.

Treating demonstrations using ponderosa pine (*Pinus ponderosa* P.& C. Lawson) posts and poles have also taken place in Colorado, Arizona, and Utah. Posts were treated by using sodium fluoride and then copper sulfate. Because these demonstrations were to inform the public of a low-cost wood preservation treatment for their refractory species, only one charge of wood was treated at each site. Chemical penetration and retention were not assessed (Reader 2000).

The most recent double-diffusion field project took place near Copper Center, Alaska. To access proposed agricultural land, the Trans Alaska Pipeline had to be crossed by using a bridge. The State of Alaska and Alyeska Pipeline Service opted to use Sitka spruce for the bridge abutments, because wood abutments would not transfer heat to the soil and disturb the permafrost supporting the pipeline. Timbers were treated with heated 4-percent sodium fluoride and then 10-percent copper sulfate (Hoffman 2002a). Samples were treated along with timbers for the purpose of analyzing chemical retention and penetration, but no results have been made available at this time.

Table 6—Chemical retentions for Alaska-grown railroad ties

		Copper		Fluoride (estimated)	
Species	**MC**	**0-13 mm**	**13-25 mm**	**0-13 mm**	**13-25 mm**
	Percent	*- - - Percent wt/wt - - -*		*- - - Percent wt/wt - - -*	
Sitka spruce	28	0.22	0.06	0.13	0.04
Sitka spruce	34	.26	.08	.15	.05
Western hemlock	26	.28	.06	.17	.04
Western hemlock	31	.48	.09	.28	.05
White spruce	39	.37	.08	.22	.05
White spruce	32	.44	.16	.26	.09

MC = moisture content.

Source: Kilborn et al. 2003 (see footnote 3).

In the most recent laboratory study on double-diffusion, railroad ties from western hemlock, Sitka spruce, and white spruce grown in Alaska were treated with 4-percent sodium fluoride followed by 8-percent copper sulfate. Ties were fully immersed for 20, 10, 5, or 2.5 days in each solution. After a 2-week diffusion period, copper content was analyzed. Fluoride content was estimated from copper content based on a previous study indicating that fluoride was found in excess of copper. Because copper sulfate labels limit treatment time to 3 days, only the chemical retentions for the 2.5-day treatment are given in table 6. The authors did not discuss the implications of the chemical analyses (see footnote 3). Furthermore, it is unknown if decay tests will be performed on these treated ties in order to determine the actual copper and fluoride thresholds needed for service in Alaska.

Several gaps still exist in the literature.

Discussion

This review found double-diffusion as the most advantageous wood-treating method that could potentially treat locally grown species in Alaska. Although there appear to be many studies treating wood by double-diffusion, several gaps still exist in the literature.

- Although copper sulfate manufacturers include wood treatment on their label, it is not clear why a maximum treatment time of 3 days in each solution was selected. It is unclear if a 3-day treatment can deliver enough chemical under all conditions of initial moisture content, density, temperature of the wood or solution, and type of wood (species, heartwood, sapwood, earlywood, and latewood). Furthermore, there is no established threshold for copper and fluoride for double-diffusion.
- The only report found on the leach resistance of chemical precipitates formed in double-diffusion treatments did not include copper fluoride. The

report only included copper arsenate, copper chromate, nickel arsenate, nickel chromate, and magnesium ammonium arsenate (Baechler 1941). It has never been established that copper sulfate forms an insoluble precipitate with sodium fluoride.

- Optimum treatment schedules to deliver adequate copper and fluoride retentions into Sitka spruce grown in Alaska, and the optimal length of time for the treated wood to be wet-stacked to facilitate the diffusion remain unknown.
- Previous studies were limited to posts, poles, and railroad ties, all of which are likely to have more easily treated sapwood. The treatment times for dimensional lumber that do no contain any sapwood are unknown.

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English Equivalents

When you know:	Multiply by:	To find:
Meters (m)	3.28	Feet
Millimeters (mm)	.03937	Inch
Kilograms (kg)	2.205	Pounds
Kilograms per cubic meter (kg/m^3)	.06243	Pounds per cubic feet
Degrees Celsius (C)	1.8 C + 32	Degrees Fahrenheit
Kilopascals (kPa)	.14504	Pounds per square inch
Cubic meters	2,360	Million board feet (mmbf)

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