

# **The Second International Conference on Wood Protection with Diffusible Preservatives and Pesticides**

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# International Status of Borate Preservative Systems

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## Abstract

The objective of this paper is to assess our understanding of the role of borates in wood preservation and their current commercial status. It reviews boron's natural distribution and chemistry; essentiality and toxicity; efficacy and performance; and boron in preservation. Boron is a ubiquitous element found widely distributed in the environment and is a normal component of a healthy diet. Elemental boron does not exist in nature, but is always found combined with oxygen in compounds called borates. Borates are paradoxical in character as they are both essential micronutrients for plants, and probably animals, but at the same time possess biostatic activity which has allowed their continued use and development in wood preservation over the last 50 years. Both of these effects are thought to relate to the chemistry of boron and its specific interaction in biological systems.

Currently there are approximately 5,000 tonnes of borate products being used in wood preservation, being split roughly into three categories:

- decorative timber treatments and construction in the Asia Pacific area;
- formulation of exterior and remedial wood preservatives in Europe; and
- in construction, wood composites, and pest control in North America.

Recent increases have come from the growth of the wood composite market, the reduction in organic solvent carried systems currently used, and the gradual removal of arsenical-based preservatives from the market. All have been as a result of the critical examination of chemical systems used in wood preservation, and the now permanent trend toward the use of environmentally sound practices.

Wood preservation based on boron is considered of very low risk to both mammals and the environment. Boron-based systems available today offer a highly effective, almost totally flexible option for both stand-alone preservatives and more specialized formulations. This flexibility and continued development has allowed borates to play an expanding role in recent developments within the preservation industry, although perceived leaching problems and aggressive laboratory testing has led to slower growth than would otherwise be expected.

## Natural distribution and chemistry

Boron is a trivalent element widely distributed in the environment, comprising about 0.001 percent of the earth's crust (48); concentrations average from 3 to 10  $\mu\text{g/g}$  in soil (1,49), 4.5  $\mu\text{g/g}$  in ocean waters (64), and about 0.01  $\mu\text{g/g}$  in fresh water (36).

Boron is widely distributed in plant and animal tissues and is known to be essential for plant growth (26,47,60,61,63). In a review by Murray (50), it was reported that the average U.S. daily dietary intake of boron is 1.5 mg/day. The median boron content of U.S. drinking water is 0.031  $\mu\text{g/g}$  with a maximum of 3.95  $\mu\text{g/g}$ . The daily boron intake for humans from

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food and water has been estimated at 0.5 to 3.1 mg for adults (52).

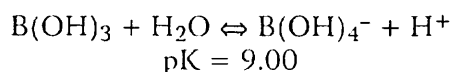
Boron is the only nonmetal in a family otherwise comprised of active metals, Group IIIb of the Periodic Table. As could be expected, boron exhibits bonding and structural characteristics intermediate to both, as do other elements lying to either side of the metal/nonmetal border.

Because of an incomplete electron octet, boron compounds can act as electron pair acceptors. This behavior is demonstrated by the Lewis acid properties of boron. It is this tendency that is fundamental in understanding the action of boron within biological systems.

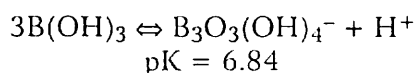
Boron does not occur in nature in its elemental form, but rather as oxygen-containing compounds such as boric acid or borates such as borax. In this paper and elsewhere, references to boron in the environment and in various applications have referred to the elemental boron content, which in some cases allows for comparisons between studies and applications.

It is these natural oxygen-containing compounds of boron that are traditionally used in biological applications. Boric acid, pentahydrate borax, mixtures of the two, or a spray-dried mixture equating roughly to disodium octaborate tetrahydrate (DOT) are the most commonly used.

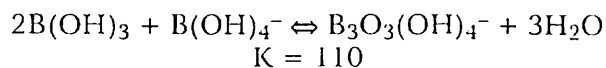
Boric acid is a very weak and exclusively monobasic acid that is believed to act, not as a proton donor, but as a Lewis acid, accepting OH<sup>-</sup>:



At concentrations less than 0.025M, only the mononuclear species B(OH)<sub>3</sub> and B(OH)<sub>4</sub><sup>-</sup> exist, but at higher concentrations the pH becomes consistent with the formation of polymeric species such as:



It is also likely that polymers exist in mixed solutions of boric acid and borates, such as:



Boron in the form of boric acid or more probably ionized as the tetrahydroxy borate anion is one of the chemical elements, whose oxygen compounds will form chelate complexes with certain organic compounds containing *cis* adjacent alcohol groups

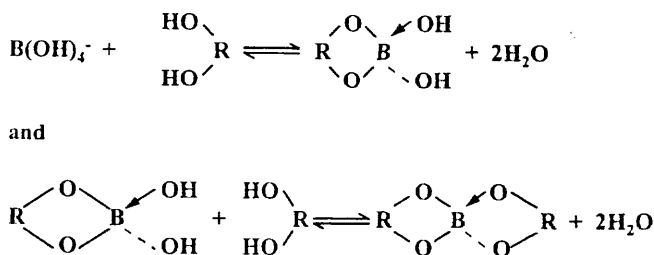
(Fig. 1). These complexes occur in aqueous solution and are well known. As early as 1842, Biot (7) reported that a solution of boric acid became acidic to litmus upon the addition of sugars. Thompson (62) found that boric acid could be determined by titration in the presence of various polyhydroxy compounds (or polyols).

Many chemical and most biochemical reactions of boron are based on the reactivity of the borate anion with adjacent alcohol groups. Steric considerations are critical in the formation of these complexes. Thus 1,2- and 1,3-diols in the *cis*-form only, such as *cis*-1,2-cyclopentanediol are active, and only *o*-quinols react.

Several compounds of biological importance such as vitamins and co-enzymes can react to form complexes with the borate ion as a result of this complexation (4,66). Reactions with these molecules and others within the cell have been found to produce dramatic changes in metabolism which appear to be the key to metabolic regulation related to essentiality and metabolic inhibition related to the preservative properties of borates (Fig. 2).

### Essentiality and toxicity

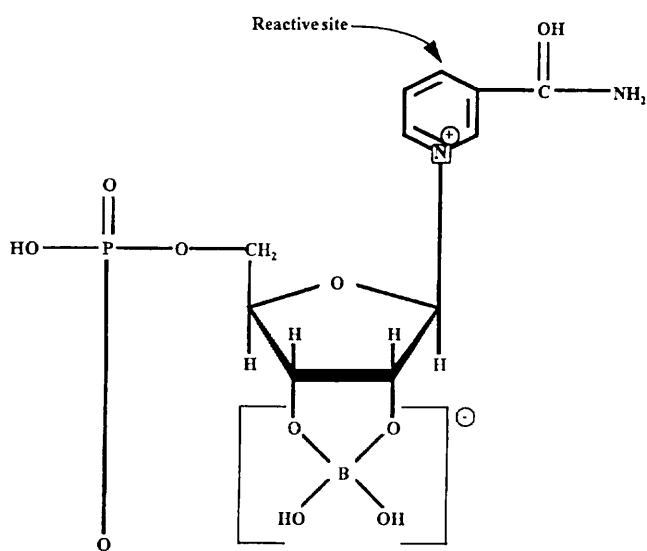
Boron essentiality and toxicity should be considered together as they probably both result from the same biological interaction. It should also be remembered that preservative toxicity is required for, if not synonymous with, its efficacy. The essential element status of boron has been demonstrated for all vascular plants (18,26,47,53,60,61,63) and Schwarz (57) suggests that because boron is ubiquitous in animal tissues and possesses properties expected of an essential element, it should be classified "under special consideration" for trace element function. Other evidence has also suggested that boron has importance in human nutrition, and it has recently been considered to be a "probably essential" trace element (65). Further information on the importance of bo-



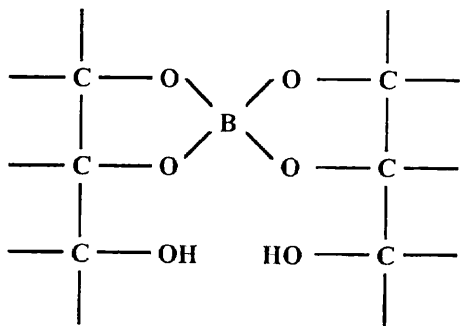
**FIGURE 1.**—Borates undergo chelate complexation with polyhydric alcohols.

ron in animal and human diets has been reported by Hunt (34) and Nielsen (52).

One of the essential actions of borates hypothesized in higher plants is metabolic regulation, by virtue of its ability to form complexes with polyols. When the borate ion complexes with compounds that are reactants or products of enzymatic reactions in plants, it may stimulate or inhibit the course of specific metabolic pathways. The effect of boron on the pentose phosphate pathway (39), production of phenolics and lignification (40,59), is a good example of this. Of course, if intracellular borate concentration is increased to vast excess, such metabolic regulation will turn to inhibition, which can be deleterious to normal metabolic function. This effect in



Interaction of borate with  $\text{NAD}^+$



Interaction of borate with plant cell wall

**FIGURE 2.**—Chelate complexation with molecules of biological significance results in regulation and inhibition of metabolism—important in essentiality and preservation.

microorganisms has been shown to be biostatic rather than biocidal, with these organisms appearing to “starve” (2, 15, 25, 55) if they are not removed from the high boron concentration.

Reports of toxicity in plants are common in areas of naturally very high boron content, although generally relate to symptoms or a yield reduction rather than actual death of the plant or necrosis of tissues. Most plants are actually quite tolerant of boron and are able to accumulate high levels.

Bacteria, fungi, and insects are also affected by relatively high concentrations of boron. Because of this, boron compounds have been used against bacteria in the form of antiseptics and as preservatives in cosmetics and food. Some species of fungi exhibit effects of boron toxicity, resulting in the aborted growth of hyphae, perithecia, and ascospores (8) and in the failure of gametes to cleave (66). The fungicidal mechanism of action of borates has been investigated (41–45). It was hypothesized that its primary mode of action was on general metabolism by interaction of the borate anion with polyols of biological significance, and the oxidized co-enzymes  $\text{NAD}^+$ ,  $\text{NMN}^+$ , and  $\text{NADP}^+$  were suggested as the most likely target of the borate ion (Fig. 2), by virtue of their *cis*-adjacent alcohol configuration and electrostatic stabilization. The hypothesis was tested on *in vitro* enzyme systems and fungal systems and *in vivo* with fungi in decaying wood. It was found that at all three levels of interaction the effects of borate could be completely negated by the addition of nonsubstrate polyols and that similar inhibitory effects could be achieved with other chelating metaloids with similar properties to the borate anion, again at all three levels. The hypothesis was thus upheld and it was concluded that on a cellular level, the same mechanism was likely in all organisms, not just decay fungi.

Boric acid and DOT are used widely as an insecticide against cockroaches and are put down in powder form (12,48). They are also used against the black carpet beetle and for ant control. In the past, EPA registration has been obtained for use against *Alphitobius* sp. and *Dermestes* sp. in poultry houses along with house fly larvae, latrine fly, and in manure. *The Farm Chemical Handbook* (22) listed borax as a larvicide against dog hookworm and swine kidney worm.

With such a broad spectrum of activity at high concentration, environmental toxicity is obviously an area that needs consideration, although impact in this area is very low as a result of rapid dilution to natural levels and its essentiality. The environmental

effects of borate have been reviewed extensively (20). It was concluded that:

Boron is an ubiquitous element, present naturally in sea water, fresh water, rocks, soil, and all plants. Boron neither accumulates in any environmental compartment nor bioaccumulates, but is transported into the oceans which have a high natural environmental background level of borate. Boron is an essential micronutrient for the healthy growth of all plants, and boronated fertilizers are used in agriculture to improve yields and to correct the symptoms of boron deficiency in crops. At the concentrations generally monitored in river water, borate causes no adverse effects to either land plants by irrigation or water plants and aquatic life. Similarly, borate levels generally detected in soil cause no effects to land plants or soil organisms. Organisms in fresh water are the most sensitive to borate. The safe, no-effect concentration of borate to all freshwater aquatic life is at least 1 mg/l.

Ecotoxicity data has been given in Table 1.

Boron toxicity to animals and humans has also been reviewed in detail. It has been concluded to be of low toxicity with an acceptable daily intake of 18 mg/kg of body weight per day (19,35,50). Potential risks in humans and mammals are avoided again by dilution (size effect) and by rapid and almost immediate excretion via the kidneys.

Boric acid and borax have in the past been used as food preservatives, although this use has largely been superseded by the widespread introduction of refrigeration during transport and storage (3). The broad spectrum activity of borates against both microorganisms and insects has led to their extended use in biodeterioration control and wood preservation. Boron compounds are used as fungicides, algae-

cides, bactericides, and insecticides (48) with their use in wood preservation being effective against both fungi and insects.

### Efficacy and performance

Early research with borates as wood preservatives carried out in Europe was concerned primarily with the problems caused by *Hylotrupes* sp. and *Anobium* spp. (37). The first experiments with boron showed it to be as effective as fluorine compounds and these results were later confirmed by Schwarz and Rensch (58). A host of other typical wood-destroying insects have also been shown to be effectively controlled by borates and these are reported in the reviews mentioned previously. Termites are sometimes considered separately from other insects. Good results have been obtained by various workers, both in the field and the laboratory (11). The toxicity of boron compounds to termites in Australia, however, has been shown to vary considerably with species, giving high toxicity to *Coptotermes* sp. and low toxicity to *Nasutitermes* sp. (23). The minimum loading that was found to give protection of matai and radiata pine was in excess of 1 percent (24) and it is generally considered that a retention higher than that needed to control boring beetles is usually needed for the control of termites (23). More recent data on termite control has been produced particularly with regard to the Formosan termite, which is now a very significant problem in many parts of the world. Multiple exposure techniques have demonstrated borates effectiveness against the Formosan termite (27-29). Commercial retentions recommended to provide protection against termites are usually in excess of 1 percent boric acid equivalent (BAE) ( $\approx 4.5 \text{ kg/m}^3$ , depending on timber density). Borates have been approved for use under termite hazard in South Africa at a retention of  $5 \text{ kg/m}^3$  BAE and in Hawaii against the Formosan termite in Douglas-fir at 8

TABLE 1.—Boron ecotoxicity data.

Algal toxicity: Green algae, <i>Scenedesmus subspicatus</i> 96-hr. EC <sub>10</sub> = 24 mg B/l Test substance: sodium tetraborate	Sea water fish toxicity: Dab, <i>Limanda limanda</i> 96-hr. LC <sub>50</sub> = 74 mg B/l Test substance: sodium tetraborate
Invertebrate toxicity: Daphnids, <i>Daphnia magna</i> Straus 48-hr. LC <sub>50</sub> = 530 mg B/l 21-day NOEC-LOEC = 6 to 13 mg B/l Test substance: sodium octaborate tetrahydrate	Freshwater fish toxicity: Rainbow trout, <i>Salmo gairdneri</i> (embryo-larval stage) 24-day LC <sub>50</sub> = 88 mg B/l 32-day LC <sub>50</sub> = 54 mg B/l Goldfish, <i>Carassius auratus</i> (embryo-larval stage) 7-day LC <sub>50</sub> = 65 mg B/l 3-day LC <sub>50</sub> = 71 mg B/l Test substance: sodium tetraborate

kg/m<sup>3</sup> BAE, although 6 kg/m<sup>3</sup> is probably more appropriate in the latter case.

Specific research on the ability of borates to control wood-colonizing fungi has also been extensively reported. There are very many species of fungi that are able to cause problems when they grow on timber. But under conditions where borates are used, these can be put into two main categories:

- the moulds and stainers, which cause only a cosmetic problem; and
- the decay basidiomycetes.

Control against staining organisms has been achieved and indeed, it was for this purpose that simple borates were first used against fungi (56). Borates are most effective against these organisms at high pH, so that sodium tetraborate is more effective than boric acid or more soluble mixtures (54) although the more soluble mixtures are usually used to achieve the normal commercial concentrations required. Typical toxic limits for different sapstain organisms have been given in Table 2, although much lower concentrations give excellent performance in conjunction with many specific organic fungicides.

The effectiveness of borates against wood-decaying basidiomycetes has now been demonstrated in service and in the laboratory by many workers (10), and is accepted. Organisms tolerant to other preservatives, such as copper, arsenate, PCP, creosote, and tributyl tin oxide were amongst the first to be tested against borates. The principal organisms used included *Coniophora puteana* and *Coriolus versicolor* with others such as *Poria* spp., *Gloeophyllum trabeum*, and *Lentinus lepideus*. As yet, no wood-decaying fungus has been reported to be tolerant to borates at normal preservative retention (16).

The most up-to-date efficacy data has been produced very recently for DOT according to the new European standards under EN 599. The relevant standard, test scenario, and corresponding biological reference values have been given in Table 3. Gener-

**TABLE 2.**—Toxic threshold for some sapstain/mold fungi.

Fungal species	Lowest concentration of BAE (kg/m <sup>3</sup> of wood)
<i>Alternaria alternata</i>	2.0
<i>Aureobasidium pullulans</i>	2.0
<i>Phialophora</i> spp. (8 species)	0.1 to 2.0
<i>Phoma</i> spp. (2 species)	2.0
<i>Rhinoctadiella</i> sp.	0.1
<i>Sclerophoma</i> sp.	0.1
<i>Torula</i> sp.	2.0
<i>Trichocladium asperum</i>	2.0

ally these are adopted as commercially acceptable minimum loadings with some consideration to boron mobility and leachability. As an example only, in the United Kingdom (nontermite hazard) the standard for borate treatment based on these results is 0.4 percent (1.8 kg/m<sup>3</sup>) BAE minimal cross-sectional retention with a 3-mm minimum sapwood penetration according to EN 351. This takes into account possible leaching due to exposure during construction, mobility in service, and variability in timber with regard to treatment. However, it should perhaps be noted that this is nearly twice the current retention (1 kg/m<sup>3</sup>), used successfully in Germany for dip treatment over many years, according to DIN 68-800.

### Preservation

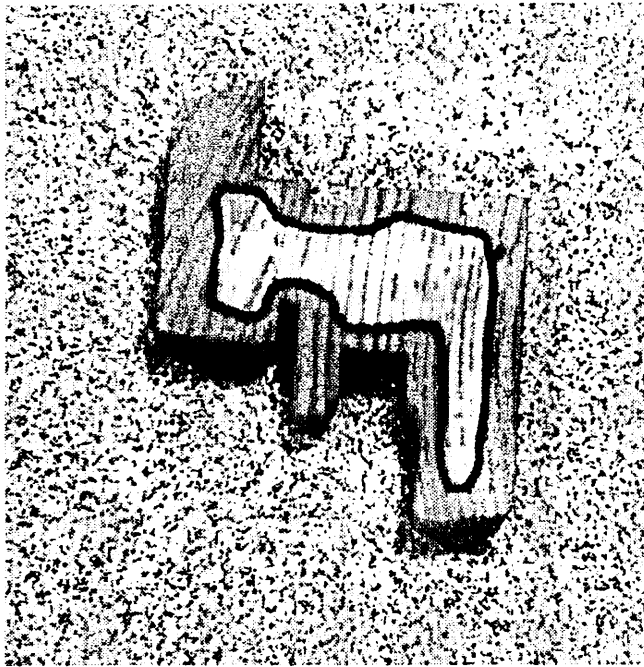
As a wood preservative, boron compounds are one of the safest in current use, and no fatalities or other harmful effects have occurred due to this or other industrial uses. For a comprehensive picture of the use of borates in wood preservation, see the reviews of Carr (9); Cockroft and Levy (11); Barnes et al. (5); Dickinson and Murphy (16); Drysdale (17); and Lloyd and Manning (46). The earliest recorded use of boron in preservation is probably in the chromium boron preservative developed by Wolman in 1913 for the treatment of timber and in fact a wealth of data and information is available in the literature on this topic. Two decades after its introduction by Wolman, boron was proposed as a replacement for dichromate in flame retardant treatment of timber (21), as it possessed fungicidal, insecticidal, and flame retardant properties. Promising toxicity data was then produced by Bateman and Baechler in

**TABLE 3.**—Biological reference values for DOT.

Test	Organism	DOT (kg/m <sup>3</sup> )
EN 113	<i>Coriolus versicolor</i>	0.76
EN 113	<i>Gloeophyllum trabeum</i>	0.59
EN 113	<i>Coniophora puteana</i>	0.32
EN 113	<i>Poria placenta</i>	0.30
EN 49-1	<i>Anobium punctatum</i> superficial treatment	8.53
EN 49-2	<i>Anobium punctatum</i> penetrating treatment	0.30
EN 47	<i>Hylotrupes bajulus</i> penetrating treatment	0.69
EN 20-1	<i>Lyctus brunneus</i> superficial treatment	5.55
EN 20-2	<i>Lyctus brunneus</i> penetrating treatment	2.07 <sup>a</sup>
EN 118	<i>Reticulitermes santonensis</i> penetrating treatment	5.55

<sup>a</sup> 1 kg indicated by other data.

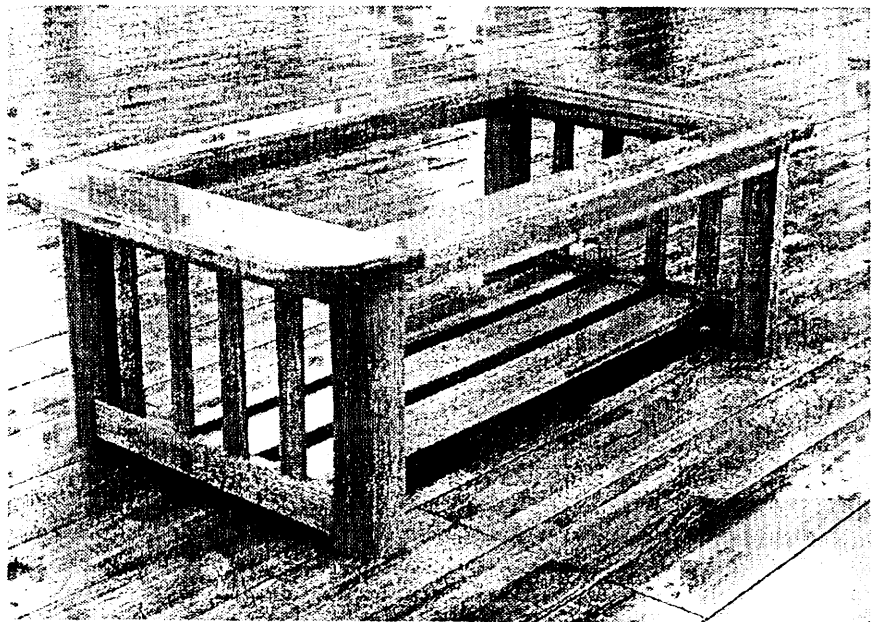
1937 (6). Three of the four commercial fire retardant formulations reported to the American Wood Preservers Association in 1949 contained boric acid (31) and the most successful flame retardant systems used today still rely on borates and their numerous attributes.



**FIGURE 3.**—Fully machined European redwood joinery can be treated by borates as seen here in this double vacuum treated cross-section. Boron is highlighted by the dark curcumin reagent.

Interest in borates as wood preservatives also arose in Australia, where they were recommended for the preservation of hardwoods susceptible to *Lyctus* spp., destined for use in plywood (14). The Forest Products Research Laboratory of Australia found that boric acid treatment made sapwood immune to attack (11,33). Diffusion into veneers and sawn lumber was good, and by 1945 treatments were being carried out on a large scale. Similar work was carried out in New Zealand, but here a treatment was needed for *Pinus radiata* to be used in house construction. The first industrial treatments were carried out in 1949 using a rapidly diffusing boric acid/borax mixture equating to DOT. A spray-dried formulation of similar chemical composition then became available in the early 1950s and was sold under the trade name of Tim-bor. Today borates have gained widespread acceptance in vacuum-pressure treatments, and envelope treatments to dry construction timber are commonplace in Europe, particularly in Germany. More recently even finished joinery treatments have been shown to be possible using double vacuum technology with aqueous borates (Fig. 3) and this is now of particular interest in Europe, where pressure against organic solvent-carried preservatives has reached the position where solvents must be replaced by water where this is shown to be possible.

One recent great success in a completely new market has been in southeast Asia. Boron treatment has been adopted almost exclusively for the treatment of rubberwood and this timber is available commercially in the form of furniture (Fig. 4) and other products on a global basis. Borates find use



**FIGURE 4.**—One of the largest current uses of stand-alone borate wood preservatives is in the treatment of rubberwood. Furniture and flooring (shown here) are common applications of the treated wood.

here, again for their performance against insects but also because of their colorless and odorless features. Powderpost lyctid beetles are significant in these products, although the false powderpost bostrychid beetles such as *Heterobostrychus* spp. are probably more so. The wood is particularly palatable to these organisms when untreated because of its very high natural starch and sugar content, which is typically used as the substrate by wood-boring beetles rather than the actual cellulose component.

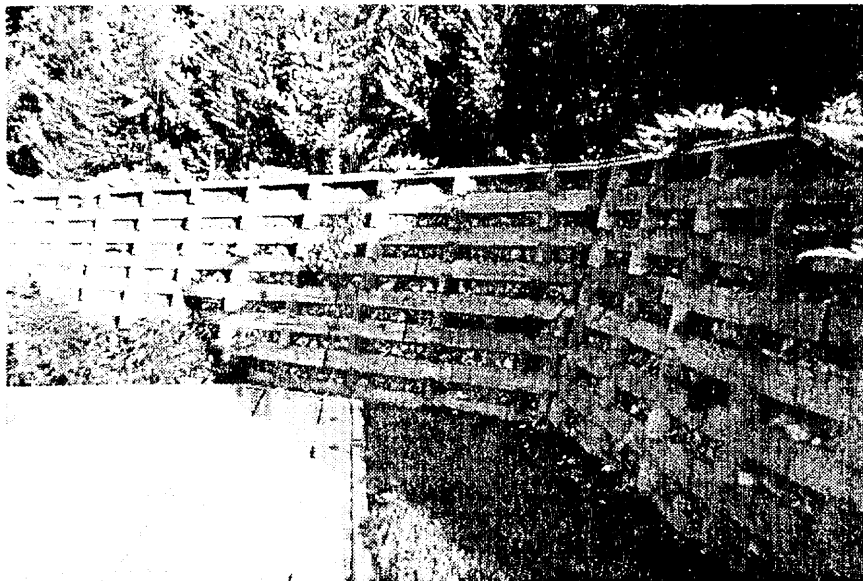
The one major restriction that borates have had in timber preservation has been as a result of their natural solubility and the depletion of borates from treated wood, which can occur in exterior or ground-contact applications. For this reason, borates have been restricted for use as stand-alone preservatives in interior situations such as the treatment of rafters and studs used in the construction of dwellings and in protected environments, such as painted external joinery. This limitation of boron preservatives is, however, directly linked to one of their greatest assets: mobility within the treated timber. This allows them to be highly effective in remedial situations in the home, as solid implants in utility poles, and in industrial pretreatment situations for construction timber in protected situations. Borates in these applications remain mobile in the treated wood and will continue to penetrate, thus providing one of the most effective wood preservative systems available today. Perceived leachability or the inability of borates to pass aggressive laboratory leaching tests remains the greatest commercial barrier to widespread boron acceptance, even in situations where there is no leaching hazard.

This restriction has, however, been to some extent overcome by the use of more complex formulations where boron is one active ingredient in a formulation containing two or more. This is particularly the case with boron-containing copper formulations used in vacuum-pressure application for exterior application. Chromated copper boron (CCB) systems were adopted by Germany during the late 1940s instead of the more widely used chromated copper arsenic (CCA) formulations, because of concerns related to the toxicity of arsenic. CCA products are now being replaced across Europe and in other parts of the world, as registration authorities insist upon their removal from the marketplace. Borates are the obvious substitute for the arsenic component which is required to give performance against insects and copper-tolerant brown-rot fungi. Only low borate concentrations are required for high performance in these systems and so the borate is not exposed to the same concentration-driven leaching pressure (42) (Figs. 5 and 6).

It is particularly the use of borates in exterior preservative formulations which is likely to prove commercially more significant in the future. Current use of borates in preservation is estimated at approximately 5,000 tonnes of  $B_2O_3$  per year, as boric acid, borax, DOT, and zinc borate, although if arsenic in CCA preservatives were restricted from sale globally, this could possibly increase to between 20,000 and 30,000 tonnes of  $B_2O_3$  per year, if the current trend of arsenic substitution with boron continues. It is interesting to note that recent studies have shown similar performance between CCA and CCB formulations (32) and that arsenic-free chromated copper



**FIGURE 5.**—The effect of DOT and a water control against sapstain is shown here, when applied to summer-felled Scots pine (*Pinus sylvestris*) with an 8-week subsequent incubation period.



**FIGURE 6.**—A boron-containing copper based formulation used on landscaping timbers.

formulations give better performance than CCA against soft rot (30). A list of commercially available industrial pretreatment products containing borates has been given in Table 4.

A further way in which borates have moved into higher biological hazard situations as a result of low leachability is in wood composites where they are used in the form of zinc borate.

The species, size, and quality of standing timber available for harvest is changing worldwide. This is promoting the development and extended use of wood composites in applications that require resistance to wood-destroying insects and decay fungi. Traditionally, solid wood products are pressure-treated with solutions of preservative chemicals. However, the nature of a composite makes it possible to incorporate a preservative into the product during its manufacture. This decreases total production costs and yields a superior product in which the composite has a constant loading of preservative throughout its thickness. Both DOT and zinc borate are suitable for incorporation into wood composites, although zinc borate has been used almost exclusively for exterior composite products where there is a perceived risk of leaching. Zinc borate has a very low water solubility at room temperature (<0.28% w/w) and provides efficacy, even after rigorous standard leaching tests (38). It is also relatively simple to incorporate zinc borate powder into a wood composite during blending.

### Conclusions

From the data reviewed in this paper it can be concluded that we have an excellent understanding

**TABLE 4.**—Borate-containing wood preservatives for exterior use.

Type (salts)	Components	Total boric acid (%)
CFB	Copper oxide ammonium bifluoride boric acid	42 (or 58% borax)
CCB	Copper sulphate copper oxide sodium dichromate chromium oxide	43
CCO	Copper oxide chromium trioxide boric acid	25
Copper quat	Didecylpolyoxethyl-ammonium borate copper carbonate boric acid	24
Copper HDO	Copper HDO copper hydroxide carbonate boric acid	22
Copper azole	Copper carbonate tebuconazole boric acid	17.6

of the natural distribution and chemistry of borates. We are also starting to appreciate how they interact with biological systems to inhibit metabolism as preservatives and to play an essential role in plant and animal nutrition.

Commercial experience over 40 years shows that when boron-treated timber is used according to specification, there has not been a single authenti-

cated case of failure in service (13) and that the mobility of borates can be considered a major asset in many application areas. However, even after many decades of successful use and the demonstration that borate mobility is a major advantage in most applications, perceived leachability and the inability of most borates to pass current standard leaching tests remains their greatest weakness and strongest commercial barrier.

On the other hand, toxicity and environmental issues have continued to favor borates. Wood preservation offers a tangible process that is very clearly in line with the current emphasis on conservation of resources and environmental impact. However, the wood preservation industry itself is under continuing pressure due to concerns over the toxicity and environmental effects of preservative chemicals used. Borates are an excellent example of where preservation contributes directly to conservation and this has already led to an increased prominence for boron compounds. Full independent reviews of the environmental impact of borates and their potential mammalian toxicity have both concluded that borates are of low risk in these areas and are having a positive effect to their adoption.

From the developments given as examples here it remains obvious that, as a tool for use in modern preservative systems, borates are probably the most versatile active ingredients available for use today. The potential range of products is almost infinite and end-use applications cover the entire spectrum of timber preservation. Recent successes have all been as a result of major changes within the wood preservation industry, and changes in which environmental concern has played strong role.

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