

Preface

This paper was presented as The Academy Lecture at the Fourth Plenary Meeting of the International Academy of Wood Science, Cape Town, September 27, 1973. Appointment of a Fellow or Fellow Emeritus to present The Academy Lecture is the highest honor the Academy can bestow. The Academy Lecture was established by the Academy in 1972 and is characterized as follows: The Lecture shall deal with a topic of current concern in the field of wood science and associated technology; it shall be concerned with a review of what has transpired in the subject matter field over the years, what is currently being done and future needs; and it shall be comprehensive in scope and emphasize the broad and international aspects. Professor Becker's outstanding treatise is the first in what will prove to be without question a most notable series.

Fred E. Dickinson, President, International Academy of Wood Science

Aspects, Results and Trends in Wood Preservation, an Interdisciplinary Science

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Summary

The paper is to give a survey on problems of wood preservation, selecting examples of fundamental research as well as of results of applied studies significant for the practice. Needs and trends in research are indicated. The interdisciplinary character of wood preservation science is demonstrated.

Introduction

It is a great honour for me to be invited to present the first "Academy Lecture" of the International Academy of Wood Science, and I wish to express my gratitude, for the appreciation thus extended to me.

The Lecture shall serve to report on progress and problems in wood science. My own field of research is wood deterioration by and wood protection against organisms. When contemplating whether I should select a special biological or technological subject or prepare a more general survey, I decided on the latter, speaking to scientists of all branches of wood research. Fire hazard problems will be excluded.

Wood science as a whole is a very complex subject. In wood preservation one has to know and consider morphology, biology, physiology and ecology of wood-destroying and wood-discolourating fungi, of insects and of marine borers. The organisms depend in their development on properties of wood species and wood-based materials. Prevention and control are based on chemistry of wood preservatives. Their efficacy against the noxious agents, their various technical properties and the technology of their application involve other fields of technical sciences. The success of preservative measures again is influenced by micro-morphological, physical and chemical properties of wood species.

A brief survey on such an interdisciplinary science is not an easy subject. Only an overall impression of the field can be given. The illustrating examples of problems and results represent, of course, an individual choice. Preference is given to publications of my own laboratory.

Organisms

In the early days of wood preservation, the organisms considered as main destroying agents were Basidiomycetes, termites and Teredinids. The soft-rot fungi as wood-destroyers were not detected until the middle of this century. Their high tolerance to a number of toxic compounds caused considerable problems in wood preservation. The Cerambycid beetle *Hylotrupes bajulus* and Lyctid beetles became economically important after changes in the use of wood. They involved the development of new types of wood preservatives. The special resistance of one *Limnoria* species to aged creosote has been known for only some 20 years and has complicated the protection of wood in marine environment.

It is not possible to survey briefly the numerous contributions to the knowledge of general biology, geographical distribution and economic importance of wood-destroying organisms made in the last decades. Yet, some examples of biological aspects will be given in order to show part of the factors, mainly the ecological and physiological ones, involved.

Fungi

The "classical" wood-destroying fungi which belong to the Basidiomycetes have been studied for a long time; and it is known which species are the economically most important in Europe, North America and Australia as well as in some areas of tropical countries [51, 84, 86, 167, 179, 218]. In other parts of the world, however, much work is still needed until the distribution and importance of species are known sufficiently. Most Basidiomycetes, once established in wood, grow quickly through it and destroy it faster than the soft-rot fungi, which belong to the lower fungi, the Ascomycetes and Fungi imperfecti [12, 76, 102, 140, 142, 153, 219]. In view of their wide distribution it is remarkable that they remained unrecognized for such a long time. Under special circumstances they can be more dangerous to timber than the Basidiomycetes, e.g. in the biotop where they were first detected: in cooling towers [85, 146, 178].

Succession of fungi including staining and entirely harmless species has been studied under various conditions only for a few years [84, 109, 122, 141, 192]. The association and sequence of species vary very much and depend on wood species, environmental conditions of the place of exposure, on antagonistic actions between species of microorganisms and on treatment of the timber. The problems of succession and establishment of fungi have still to be investigated with respect to various biological and practical aspects, including inhibition of wood-destroyers by special mould fungi [191].

The threshold of wood moisture for the growth of fungi seems to be rather uniform for many wood-inhabiting fungi, and is approximately 22% [202]. In the long run at least part of the fungi may need fibre saturation conditions. Nevertheless, preferences of special biotops or tolerance to ecological situations exist for a number of species. The effect of high moisture differs from species to species. The survival of Basidiomycetes under very dry conditions, long known, has also been studied for many years. It varies with the species and depends on the speed

of desiccation. The mycelium of some fungi may survive for more than 10 years [204].

The dependence on temperature has so far been investigated only for a limited number of species. The optimum for mycelium growth of the majority has been found to be between 25 and 30° C [12]. A well-known exception with a low optimum and a high sensitivity towards temperatures above 25° C is *Merulius lacrimans*, the most important fungus in old houses in Europe. More species of economic significance should be investigated [213].

The enzymatic degradation of wood leads to various types of decay and discolouration [5, 6, 12, 53, 54, 103, 138, 144, 161, 217]. Fungi without cellulase production, such as most staining fungi, are able to grow only through openings in the tissues or the pectinous tori of the bordered pits of conifers. While hyphae of Basidiomycetes can penetrate cell walls in a transverse direction, the attack of soft-rot fungi is restricted to the middle part of the cell wall where they may cause characteristic holes [127, 147]. It is supposed that the material of the primary and of the tertiary walls is resistant against the cellulase of soft-rot fungi. The breakdown of susceptible hardwood is about twice as fast as that of softwood species. This, too, probably depends on the composition of the carbohydrates of the wood. The cellulolytic activity may vary with strains of the same fungus species, especially in the case of Ascomycetes and Fungi imperfecti [125, 145]. Early results of different authors who did not use the same strain therefore seem to be contradictory. The variation of strains in properties and reactions is to be considered as a very general characteristic of microorganisms.

As regards breakdown of cell wall material, the so-called brown-rot Basidiomycetes and the soft-rot fungi are comparable. They decompose the holocellulose effectively, but destroy the lignin to only about 7% and about 20%, respectively, of the original content. The white-rot fungi cause a different type of degradation; their decomposition of lignin may lead to losses of >40% of this substance [189]. The degradation process of Basidiomycetes is characterized by a high production of cellulase shortly after the hyphae have grown into the wood. Then a period follows in which cellulase activity decreases, and the carbohydrates produced from shorter chain-length are used [188].

It is still unknown whether the fact that the oxygen consumption of a Basidiomycete fungus culture always decreases after about 2 to 3 weeks [65, 66, 194] is somehow correlated with the change in enzyme production, whether the oxygen consumption depends on the growth of mycelium, or whether the fungi change their respiratory system after some time of development. Another unsettled question is why the respiration activity shows fluctuations not depending on external factors. Their time rhythms are typical of the individual species.

Growth activity of the mycelium of different strains of a species and capability of wood degradation are not necessarily related [94, 100]. They may differ remarkably with regard to the many factors that influence growth and attack of wood. The dependence on the nitrogen content of the substrate also varies with the species [12, 113]. Both examples demonstrate that much research is required before general statements can be made, tests depending on environmental and nutritional conditions for the fungi be adequately carried out and the results be correctly evaluated and applied.

Bacteria

The question of the significance of bacteria in wood deterioration needs further investigation. Wood in soil and water under conditions of low oxygen content is frequently full of bacteria and probably decomposed by these [46, 55, 106, 108, 148, 152]. Species with cellulolytic ability are well-known. Another field where bacteria can become important are piles of wood chips, mainly in tropical countries [104, 118]. Anaerobic bacteria and species with a high temperature tolerance may develop under ecological conditions which are unsuitable for most fungi.

Insects

The most important wood-destroying insects belong to some groups of Coleoptera (beetles) and to the Isoptera (termites). The insects destroy the wood mechanically for enzymatic digestion in the gut. Properties of wood species, qualities of sapwood and heartwood, humidity and temperature are factors determining the activity and noxiousness of the individual insect species. Comparable with antagonistic influences on fungi, natural enemies may keep down the insects; *Sirex* control in New Zealand is an example of practical importance.

Regarding humidity as an ecological factor, Coleoptera species that need the same moisture content of aged wood as fungi are normally less important than microorganisms growing in the same biotope. The majority of Cerambycids, part of the Anobiids and all Curculionids belong to this group. The beetles attacking freshly cut wood and belonging to the Scolytids and Platypodids, which need a high moisture content of the wood for their ectosymbiotic, mostly staining fungi, are of high economic importance, especially in the tropics. However, dry-wood beetles which are able during the whole life-cycle of the larvae to develop in wood with a moisture content below fibre saturation endanger timber also in environments without fungus occurrence. The lowest limit of relative humidity of the air and the respective wood moisture content needed for growth is found with Lyctids and Bostrychids. *Hylotrupes bajulus* (L.), the Cerambycid, important for conifers in Europe and South Africa, requires a limit of about 45% humidity, whereas *Anobium punctatum* (De Geer), a species distributed in temperate climate zones all over the world, needs a minimum of about 55% relative humidity of the air or about 10% moisture content of wood [15]. A dry-wood Cerambycid genus of the tropics is *Stromatium*.

The optimum temperature for several Cerambycid and Lyctid species is about 27 to 30° C, a range which kills *Anobium punctatum* of Central European origin, the optimum for which is 22 to 23° C [15]. The knowledge of the ecology of more species would not only contribute to the understanding of their distribution [57] and their economic importance, but also make prophylactic measures possible.

As to the significance of humidity for termites, the exceptional position of the so-called dry-wood termites is well known and has long been utilized in their control. The dependence on temperature, however, was investigated only rather recently. It seems that tropical termite species cannot develop at temperatures below 22 or 20° C and die if kept at lower temperatures, while species of temperate climate zones may survive even low temperatures near the freezing point. The

optimum for food consumption has for many termites been found to be around 28 to 30° C [24, 27]. It is much lower only for exceptions such as North American *Zootermopsis* species. Termites of the savannah and some subtropical dry-wood species may be able to tolerate higher temperatures than species living in dense tropical forests for which already 32° C as a constant temperature can be beyond the optimum. Even within the same genus, e.g. *Nasutitermes*, the optimum can differ, probably in connection with the normal exposure of the nests of the respective species to the sun. Much more information is needed about the dependence of the development of incipient termite colonies and the life-time of individuals on temperature conditions and their tolerance of fluctuating temperatures.

The nutritional physiology of wood-destroying insects which depends on enzymatic potentials and on properties of wood species shows a great diversity [28]. It has been known for some time that part of the Coleoptera such as Lyctidae, Bostrychidae and some Cerambycidae do not possess cellulase or hemicellulases and therefore depend on the sugar and starch content of their food material. This fact is of high practical importance, in offering possibilities for protection without insecticides. The breakdown of cellulose by the larvae of the few Coleoptera species that have been studied with regard to this ability is rather limited. Only some 25% of the holocellulose is converted [189]. Termites are, however, capable of using more than 90% of the cellulose [190]. Their efficient system of cellulose digestion based on symbiotic flagellates and bacteria in the gut needs more detailed investigation.

Wood as food material, while extremely rich in carbohydrates, has a very low content of nitrogen [7, 18]. This was neglected for a long time until it became evident that the quantity and quality of amino acids in wood are decisive for xylophagous insects and are a clue to the understanding of their biology and modes of attack [15, 18, 28]. Part of the wood-inhabiting species overcome the nitrogen deficiency through the contribution of their endosymbiotic microorganisms, by feeding mainly in the bast with its high nitrogen content, by breeding ambrosia fungi in the galleries, or by living in wood already decomposed by fungi and thus enriched in nitrogen. The length of the life-cycle depends—so far as nutrition is concerned besides temperature and humidity—to a large extent on specific amino acids. In aging wood, the composition of the amino acids probably changes. The nutritional value of timber decreases for *Hyloterpes* larvae after some decades, while the nitrogen content remains unchanged. Addition of suitable amino acids and vitamin B to timber several centuries old in which *Hyloterpes* larvae died, again renders it a good diet for the larvae [18]. The nutritional value of susceptible hardwoods for *Lyctus* species disappears already after a few years [58]. However, the economically important *Anobium punctatum* is able to grow in very old wood, probably due to its endosymbiotic yeasts which may contribute to the nitrogen and the vitamin supply of their hosts [15].

Termites have an additional source of nitrogen in their symbiotic flagellates and bacteria. Both groups form essential amino acids, as has been demonstrated by experiments in which part or all of the microorganisms were removed by application of specific antibiotics [198]. Endosymbiotic bacteria may fix the nitrogen of the air [48]. Another supplier of nitrogen compounds are fungi. The complicated culture of fungi in special "gardens" by termite species which build

big mounds characterizing the landscape in African and Asian countries has long attracted the interest of zoologists. It has become known only recently, however, that wood-deteriorating fungi contribute much to the nutrition and development of all termites [20]. The insects are strongly attracted by certain compounds in the mycelium and in decayed wood [20, 81, 196]. This attraction has already been applied in practice as a control measure in North America [80]. In laboratory tests, termites eat much more wood, consume oxygen and furthermore build much longer soil galleries in the presence of only the smell of a suitable fungus mycelium without any contact with it [38]. It should also be mentioned that not all fungus species are equally beneficial and attractive to termites. Some are repellent, and these as well as non-repellent ones and special bacteria may be harmful [37, 156, 197]. A number of mould fungi are highly toxic, and termites which eat their mycelia or spores die within a short period [37, 197]. The poisonous effect originates in special toxins. Strains of the same fungus species may differ in content of toxins and be either strongly or moderately toxic or even harmless [33, 37].

Coleoptera larvae also depend on fungi in wood which may be indispensable for part of the insect species or beneficial or toxic. For instance, some blue stain fungi accelerate the development of *Hylotrupes* larvae, while others kill them [26].

The natural durability of wood species against fungi [74, 143, 181, 215] and insects [176, 177] and the efficiency of toxic compounds known to cause the durability are theoretically as well as practically of great interest. Sufficient knowledge of the chemical principles of toxic and repellent effects on insects can become a basis for the development of new synthetic preservatives needed after part of the contact insecticides has been banned. A number of compounds from wood extracts prepared in Hamburg-Lohbrügge proved to be of such a high efficacy that this can be compared with the toxicity of contact insecticides.

Wood species and extractives were tested against termites mostly with only one or very few species of these insects. Comparative studies on the efficiency of toxic or repellent wood substances against a larger number of termite species belonging to various families, however, demonstrated that remarkable differences can occur [39]. Especially within the family of Kalotermitidae an exceptional tolerance was observed, while termite species of the family Rhinotermitidae showed more uniform reaction. The investigation of the influence of the toxic substances on the symbionts of the termites showed that the species of these organisms may be influenced very differently by the individual compounds and that they thus contribute to the difference in the sensitivity of the termites. General conclusions cannot be drawn before further detailed investigations with many termite species are carried out.

It may be worth mentioning that the astonishing repellency to termites of sapwood of freshly cut *Pinus sylvestris*, which is not present in spruce or fir, was found to be caused by furfural [40]. The repellency disappears by watering or short ageing of the wood.

It is still unknown which compounds cause the toxicity of hardwoods to *Hylotrupes* larvae, which is based on substances that can be extracted with low alkaline solutions such as 0.5% sodium hydroxide solution [14].

Marine borers

In the field of marine borers taxonomical work has been an important precondition, and revisions made on a world-wide basis are fundamental for further work. In the case of Teredinids, a number of species names proved to be synonyma [208]; with the genus *Limnoria*, on the contrary, many new species have been detected [158]. *Limnoria tripunctata* Menzies was found to be widely distributed and of great economic significance. The question, whether *Chelura terebrans*, which is always associated with *Limnoria*, is an important wood-destroyer, found an interesting answer. This species feeds preferably on the faeces of *Limnoria* and its own. *Limnoria* reduces the cellulose content of the wood only to about 20%. Conversely, *Chelura* by its activity improves the oxygen conditions of *Limnoria* to some extent [132]. *Sphaeroma* uses the wood mainly [131] and *Martesia* only for shelter. Their nutrition depends on microorganisms at the walls of the holes of *Sphaeroma* and on plancton in the case of *Martesia*.

Teredinid larvae are attracted for settling on the wood by microorganisms. The primary film formed by bacteria and especially the attack of the wood near the surface by marine fungi are a precondition for the attack of the borers [124]. The *Teredo* species which was used in Berlin-Dahlem for laboratory tests had the advantage of a short swarming period of only a few days and of lunar-periodical spawning so that tests could be well planned. It could be concluded from the results that prevention of the microorganisms should be a protective measure against the molluscs. Observations in nature seconded this hypothesis. This is an obvious example of the importance and necessity of biological investigations also with respect to practical tasks of wood protection. *Limnoria* is favoured in growth and reproduction by the presence of marine fungi in the wood, probably largely because of the increase in nitrogen [128, 159]. These relations, which are not easy to investigate, should be studied in more detail.

The influence of ecological factors such as salinity, temperature, oxygen concentration, light conditions and water movement on the marine borers has been extensively investigated mainly in various European countries [79, 132], the U.S.A. [158, 208] and India [169].

Wood preservatives

Wood without natural resistance against organisms needs to be protected if the environmental conditions under which the timber is in service make an attack possible. Wood preservatives are to be efficient against deteriorating agents, but must not be dangerous to humans. Good penetration into wood and permanence of efficiency under the service conditions are essential. The preservatives have to meet a number of technical prerequisites. These are mainly: no reduction of mechanical strength of the wood or of glueability and paintability, no increase in inflammability of timber or in metal corrosion and no change of properties of other materials in contact with the treated wood. As little odour as possible and no change of colour are required for special purposes. The possible hazard for the environment has long been observed in wood preservation in various countries.

Compounds and types

Two main groups of wood preservatives, namely water-soluble inorganic salts and organic substances are known, and some elements or substances have been used for a long time [49, 83, 117, 210]. Compounds of copper, zinc, arsenic, fluorine and boron have a long history as wood preservatives. The addition of chromium compounds for rendering the other salts more or less stable against leaching is still the only solution for successful performance of water-soluble preservatives in wood exposed to rain or wet soil.

Creosote which has been used for wood preservation for more than a century varies in its composition from country to country. Developments tend to improve its penetration into wood or its permanence or to make it more effective against special organisms.

Numerous organic compounds have been screened as to their efficiency against wood-destroying organisms. Pentachlorophenol has become a substance of outstanding importance, mainly with regard to fungi [470., 111, 117, 175, 210]. Some organo-tin [8, 63, 91, 114] and organo-mercury compounds have been introduced into wood preservation for special purposes—to mention only a few of many successful substances.

Some synthetic contact insecticides proved to be very effective also against wood insects. Oily preservatives containing about 1% of these are of enormous efficiency. The toxic limits obtained are extremely low, and the permanence is satisfactory. Nothing in wood preservation against fungi is, until now, comparable to the high efficacy of contact insecticides against insects.

The development of organic solvent-type wood preservatives has not been limited to pentachlorophenol, metal-organic compounds and synthetic contact insecticides. Special fractions of creosote and various other organic substances were also combined with different carriers and additions for special purposes. Deep penetration, when applied without pressure differences, lack of colour, little odour, easy glueing and painting of the treated wood were desired properties in conjunction with high efficacy for these products. They are mainly used for wood in buildings, for joineries for interior and exterior use, and for furniture and wood-based materials.

Water repellency combined with preservation and reduction of cracking are required in many cases. Satisfactory results have recently been obtained [45, 174].

Testing problems

The number of wood preservatives which were to be tested has been constantly very high in some countries. This explains why laboratory testing and screening methods providing answers on efficacy, permanence, applicability etc. after rather short a time have been thoroughly studied and discussed, also on an international basis. Reproducibility and acceleration as two preconditions for laboratory testing are to be based on detailed knowledge of the organisms, their biology, nutrition and dependence on environmental factors. The latter may influence also the permanence of preservatives. Contrary to testing physical and mechanical properties of wood, in the case of wood preservation many results cannot be trans-

ferred immediately to practical conditions, and correct translation is part of the art—if I may use this word—of testing the efficacy of wood preservatives.

The main laboratory testing method against fungi is the determination of thresholds of the fungicidal efficacy. The principle is that small blocks of selected timber are treated with concentration-series of the preservative and the limit of fungal attack is then determined under defined conditions. The substrate on which the fungi are grown and the sizes of the blocks differ with the individual standards [89, 112]. A basic problem is the suitable choice of representative species of fungi which should be the most important ones in the respective area. The tolerance of species against the same toxic substance may differ remarkably, as has long been known [61, 62, 73]. Differences can be large even among strains of the same species [94, 183]. The multitude of factors involved which influence the final result and the accurate definition of attack at the threshold cannot be dealt with here in detail. Only one observation should be added. The fungi are able to grow through layers of several millimeters with a much higher concentration of a preservative than the threshold represents. This has several consequences.

An essential difficulty in using the threshold obtained by means of nationally or internationally accepted testing procedures is the difference between the quantitative distribution of the preservative in the small testing blocks where axial penetration prevails, and in timber of larger dimensions treated under practical conditions. In order to fill this gap, a test method using test specimens of larger dimensions has been developed, i.e. the so-called fungus cellar test [95, 97]. Its principle is to expose timber of practical size, e.g. boards for floors, to a well-known single fungus species under constant conditions optimal for the fungus. Details of the method will not be repeated here. The results obtained can be transferred immediately into practice. The exchange of air prevents a concentration of gaseous evaporated substances as it takes place in small testing containers. Therefore *Merulius lacrimans*, a very sensitive fungus with lower thresholds in Kolle-flasks than those of other species, proved to be as tolerant as these under the fungus cellar conditions. When particleboards treated with fungicides in order to render them resistant under wet conditions in buildings were to be tested, block methods in small containers failed due to the inhibiting influence of the glue material. In the fungus cellar, however, the testing of the efficacy of their thresholds has been found to be possible [99].

Laboratory tests with soft-rot fungi are being discussed, and international expert groups carry out comparative round-robin-tests [47]. It seems that the burial of blocks in unsterile soil with a number of different soil types or a polyvalent one gives the most appropriate answer [203]. The water-holding capacity of the soils is to be considered [36]. For laboratory tests with single species the use of vermiculite as a substrate is recommendable [126].

In field tests, mainly performed as stake tests, the wood is exposed to natural associations of various fungi and termites—if these insects are represented at the places, long experience is available with stake tests in many countries [44, 82, 88, 136, 171, 172, 199]. The distribution of preservatives in stakes does not represent, however, conditions with larger dimensions of timber. The respective IUFRO-Committee has prepared a proposal for unification of the stake test.

The efficacy against insects can be tested by determining the toxic values representing 100% and just non-100% death rates of the larvae under defined conditions, the prevention of development of freshly hatched egg-larvae under conditions simulating attack in practice, or the eradication of larvae in infested wood. It has been determined to what extent the results of the semi-practical laboratory methods can be transferred to practice. A comparison of the resistance of Coleoptera species is possible only for a small number of Cerambycids, Anobiids and Lyctids.

In laboratory tests with termites most institutes have used one species only. Comparative investigations in Canberra [92, 93] and Berlin-Dahlem [21, 23] have, however, shown that serious differences among species may exist, especially between Rhinotermitidae species on the one side and Kalotermitidae or Termitidae species on the other. It seems that part of these differences is based on the reaction of the symbionts of the termites which are indispensable for these insects [135]. The most efficient toxicants with regard to termites are some contact insecticides and arsenic compounds [21, 23, 92, 93]. Termites behave quite differently from Coleoptera larvae. Their ability of moving freely and their reaction to repellent substances influence the performance of a preservative under practical conditions as well as the results of laboratory tests [90].

Water-soluble preservatives

The chromium compounds which are still the only means for obtaining fixation of water-soluble salts in wood were originally introduced into wood preservation in order to reduce the iron corrosion caused by fluorine compounds in treating tanks. The finding that leaching of these compounds is reduced was originally a "by-product". Arsenic compounds, the fixation of which by chromium is better than that of fluorides, were later added in order to improve the efficacy against *Hylotrupes bajulus* in poles. This, however, was erroneous since fluorine compounds are more efficient against these larvae than arsenic compounds. The mixture of chromium, fluorine and arsenic was, however, better than the CF mixture.

Copper sulphate has long been used in various European countries. Failures of poles due to attack by *Polyporus* species in Germany and Switzerland terminated the use of copper in Germany in the 30s. In other countries, mainly in the tropics, where hardwood species with their considerably higher susceptibility to soft-rot fungi were used in stake tests, copper-containing salt mixtures showed a much better performance than mixtures without copper [44, 123, 172]. Salts containing chromium, copper and arsenic compounds proved efficient against both types of fungi, termites and beetle larvae, and were resistant to leaching [19, 42, 60, 82, 139]. Recently, the influence of relations between the three components on efficacy and permanence against leaching was studied [64, 193]. Other combinations contain chromium, copper and boron or chromium, copper and fluorine compounds [19].

The introduction of bifluorides into wood preservation in Germany was prompted by search for a water-soluble compound efficient for in-situ eradication of *Hylotrupes* larvae. Their special efficacy was detected when medium-size *Anobium* larvae, which are resistant against stomach poisons but killed by hydrogen fluoride gas, were used for screening tests; with the less tolerant *Hylotrupes*

trupes egg-larvae it would have been overlooked. The depth of penetration of bifluorides which is based on HF-ion diffusion is very advantageous also for preventive treatment without pressure application [29, 185, 186]. The phenomenon of splitting off of the HF-molecule in wood and the loss of fluorine by evaporation have been studied in detail [30, 31]. Fluorine compound, concentration of salt and its distribution, wood species and its penetrability, pH-value, humidity, and temperature are factors that influence the HF-evaporation. The remaining fluorine protects sufficiently for a long period if the treatment has been adequate.

Boron is another element the water-soluble compounds of which have importance in wood preservation [11, 17, 50, 52]. The efficacy against fungi is similar to that of fluorine compounds. Protection against beetle larvae is provided if the penetration is deep enough. The leachability is as high as with fluorine compounds, but no evaporation has been observed. Low toxicity to human beings and fire retardance are advantages.

Organic preservatives

The efficiency of fractions and compounds of creosote against fungi, beetle larvae and termites has been tested thoroughly. Polyvalence in efficacy and permanence are based on the combination of many compounds. The ageing process is now known [25, 165, 195]. The results are the basis for deliberations about possible modifications.

The resistance of *Limnoria tripunctata* against aged creosote without the lower-boiling compounds prompted many trials in the U.S.A. to improve the permanence of the efficiency against this borer [72, 115, 116]. Addition of copper compounds presents one way [133], use of toxic organic compounds another [115]. In Australia, the attack of the interior of creosoted poles by *Coptotermes* species led to the addition of arsenic compounds to the creosote [119]. The effect of an increase of high-boiling fractions on the permanence of creosote in tropical countries is being tested with railway sleepers in Liberia.

The ageing of creosote has been studied as to quantitative loss and qualitative change of composition in connection with the question why beech railway sleepers show a better performance and have a much longer service time than pine sleepers [25, 165]. The creosote evaporates from the interior of the tracheids of the latter through the pits. At first the low and medium-range-boiling compounds disappear, and after several years the pine sapwood contains only high-boiling substances known to have a relatively small effect against fungi and insects. In the beechwood, however, only the few outer millimeters show some loss of creosote, whereas in the vessels in the interior of the wood it was found nearly unchanged still after 20 to 30 years' service time. The vessels of beech have no pits or other openings in their wall; fluids or gases can penetrate into and move out of the vessels only in the axial direction. At the ends of the long vessel, evaporation leads to a sealing of the vessels, and thus the creosote in their interior remains nearly unchanged. Most probably the same situation exists in other hardwoods with a similar morphology.

The organic solvent-type wood preservatives gained increasing importance mainly for timber in buildings and furniture during recent years [1]. Their composition shows a large variation. Experience and problems cannot be reported here.

A comprehensive survey completed recently in France within a scheme of the IRGWP provides valuable information. In the case of new organic solvent types, the permanence must always be tested thoroughly. For a number of organic products a permanence of efficiency of more than 15 years has been proved [22]. Combinations with colouring pigments which may influence the penetration of the efficient compounds led to the development of a modified testing procedure. Thresholds against fungi and preventive action against *Hylotrupes* egg-larvae were tested also after removal of wood by planing layers of one or several millimeters. While many preservatives showed a deep penetration of insecticidal efficiency, only part of the preservatives protected against fungi after more than a 1-mm-deep removal [41].

The performance of a wood preservative does not only depend on its efficacy against the deteriorating agents and its permanence against ageing factors. Properties of wood species may also play a decisive role. Chromium/copper/arsenic wood preservatives, effective in many softwoods and hardwoods under severe conditions, fail in some hardwood species despite high loadings injected by pressure treatment. In wood species with this exceptional attack of soft-rot fungi, electroscan and electronprobe investigations carried out independently in two laboratories [71, 105] showed that there was no penetration of the salt into the cell walls. The crystals were found on the surface of the interior of the vessels only. The reason for the lack of penetration into the cell walls is still unknown. Possibilities to treat the respective wood species satisfyingly will be explored.

Another example of the influence of wood properties is that kilndried wood leads to other toxic values of contact insecticides than air-seasoned wood [35]. The type and temperature of drying influence the absorption and distribution of preservatives and the loss of organic preservatives by evaporation and thus the long-term performance.

Wood preservatives can be decomposed by organisms, mainly by bacteria. Observations were made first with creosote and pentachlorophenol [56, 72, 75, 137, 180, 209]. It is known that special microorganisms may be able to use substances which are very toxic to others. The decomposition is not only important with respect to the permanence of preservatives in treated timber but also for the degradation of toxic substances in soil.

Treating methods

The selection of adequate treating methods and their effective application depend on the morphology and permeability of wood species, the moisture content of the wood, the preservatives to be used, the final destination of the treated timber with probability of hazard by organisms, and ageing factors of the service conditions influencing the permanence of the preservative.

Cylinder treatment

The injection of fluids in closed cylinders by means of pressure and vacuum is the main preservation technique for most timber in many countries. There exist various modifications that have been used for quite a time. The success depends on the permeability of the wood tissue. Much experimental work in recent decades

has been devoted to refractory wood species. In order to improve their treatability, seasoning, presteamer, incising and other pretreatments were studied [9, 200]. The results have been compiled in surveys and are therefore easily accessible [43, 117]. Pretreatments were investigated also in connection with the prevention of bleeding of creosote from pine poles [150].

The oscillation process, developed in Sweden and using short-term sequences of pressure and vacuum of changing intensity, has some advantages for the treatment of refractory European spruce in semi-dry condition [110, 214]. In Australia, high pressure of up to 70 atm is applied for the treatment of refractory *Eucalyptus* species [200]. With many other species, however, high pressure leads to a mechanical collapse of the wood tissue.

The strong blockage of the pits in spruce and Douglas fir during seasoning which causes the refractory behaviour of these species [149, 151] can be changed by the activity of mould and staining fungi [121, 154, 155] or by watering the logs; in the latter case bacteria may then decompose the pectin of the tori and thus reopen the connection between the tracheids [77, 78]. The procedure of adding pectinase-enzymes to the storage water has also been studied under practical conditions in order to render the wood permeable for pressure and vacuum treatment [10, 207].

For application of water-borne preservatives, full-cell treatment was propagated which is to provide as high an absorption of fluids by the wood as possible so that an adequate amount can reach also the interior layers [130].

A rather new development is the use of organic fungicides and insecticides in an appropriate solvent in a vacuum cylinder for the treatment of building timber, especially window frames and other wood for exterior use and to recover the carrier-solvent [3, 173]. The timber can be well treated by this method and be glued and painted easily.

Sap-replacement and related methods

Sap-replacement, the method devised by Boucherie already some 120 years ago, is a very suitable method, especially for wood species that are refractory to pressure treatment. This is true not only for spruce and part of Douglas fir but also for many tropical and subtropical hardwood species. Besides that, the lack of pressure-plants in tropical areas can be another reason for giving preference to a sap-replacement method [96, 157].

Disadvantages of the original method were the very long treating time from 2 to 3 weeks and the poor treatment of the outer layers of the bottom end due to the form of the caps. By the development of caps which allow the use of pressure from 1 to 2 atmospheres, the treating time has been reduced to 24 hours or even less. Furthermore, the most susceptible outer layers of the sapwood are much better treated by means of the new type of caps than before [96, 101].

The sap-replacement method can be modified and combined with the open-tank treatment. Debarked trunks are submerged in a treating solution which can penetrate in the radial direction through adhesion and diffusion, and the same or another solution is injected in the axial direction by means of pressure from the bottom end or suction from the top end or both. The effect of these variations on the qualitative and quantitative distribution of salts has been studied recently [101].

Non-pressure methods and diffusion

Brushing, spraying, dipping and open-tank treatment have long been applied, mainly for building timber to be used under conditions with little or minor risks. These non-pressure methods found wide application, e.g. in Germany [187] where only a small part of constructional timber is treated in cylinders. One important reason for this situation is the development of water-soluble and oily preservatives with a penetration into wood two to three times deeper than that of the older products. These deeply penetrating products are also used for the eradication of insect larvae in infested wood by means of in-situ treatment; this task was a stimulus for the development of such preservatives.

For the surface treatment of large quantities of wood, either dipping and open-tank soaking or spraying are the methods mostly applied [160, 182, 206, 211]. For the treatment of window frames and other prefabricated material with oily preservatives spraying tunnels have been developed through which the timber is moved with constant speed, thus providing an even application of fluid.

An important field for dipping or spraying is the treatment of veneers, especially in tropical countries [88]. Plywood can be protected by means of the application of special preservatives to the glue from which the fungicides and insecticides penetrate into the wood. Particleboards may also be treated by addition of preservatives to the resin [69].

Non-pressure application can carry a risk when insufficiently seasoned timber is treated. Cracks may develop later, thus opening the untreated interior of the wood which is then subject to attack by fungi and insects [13]. Renewed treatment recommended for such a situation is not always practised.

Fluids applied without pressure differences move into wood through adhesion and capillarity in the tracheids and vessels. In the case of water-soluble salts, diffusion is also of great importance for preservation. This possibility was first used for the treatment of freshly felled and debarked trees on the surface of which the salt was brushed as a paste. The process later became important for the remedial treatment of the ground zone of standing poles by means of salt bandages [129]. Diffusion does not depend on the existence of fresh sap, but only on the water content of the timber. The depth of penetration of salts applied by various non-pressure methods was improved by storage of the treated timber in stock and covering it with plastic sheets or other appropriate material in order to keep it in a moist condition. Diffusion is mainly used with fluorine and boron compounds, for the velocity of diffusion is highest with elements of low molecular weight. It can, however, be used also with other elements. In the case of fluorine compounds, HF diffuses into wood with a moisture content even below fibre saturation [29]. Additional diffusion may provide rather deep a penetration also when the salt solution is applied by spraying, brushing, or dipping [201]. Diffusion depends mainly on the water content of the wood. In softwoods its velocity is highest parallel to the fibre direction and lowest in the radial direction.

The speed of movement of preservatives in the wood injected by pressure or vacuum, or by adhesion and capillarity during dipping and soaking, or by diffusion is represented by minutes, days and months.

Quantitative distribution of preservatives

Results of treatment have been tested and recorded for a long time only by determination of the quantity of absorbed fluid and of the depth of penetration in a concentration which could just be seen by eye or detected by special colour reactions [205]. For an exact investigation of the quantitative distribution of preservatives in various layers of the treated wood, methods of chemical analysis had to be developed which are applied on a semimicrobasis. In the case of inorganic salts, besides the classical analytical possibilities, the atomic absorption spectrometry provides a time-saving method for a number of important elements [64, 134]. Organic substances, even of very high complexity in composition, can now be quickly identified by means of gas-liquid and thin-layer chromatography [163, 164, 166, 167]. After the development of appropriate analytical methods, information about the quantitative distribution of preservatives became available, which contributed much to the understanding of the specific advantages or shortcomings of methods and preservatives. Quite recently the use of electron-probe X-ray microanalysis and of energy dispersion X-ray ancillary apparatus to the scanning electron microscope made possible investigations of the distribution of preservatives in the microstructure of the cell walls [59, 71, 105, 170, 173]. It can be expected that some special unsolved problems in wood preservation will be solved by means of these highly advanced techniques.

The quantitative distribution of preservatives throughout the cross-section of a piece of treated timber shows a more or less steep decrease in concentration. This is true not only for the application of non-pressure methods [16, 107, 184], but also for pressure and vacuum treatment [67, 98, 212, 216]. Wood species with good radial permeability through rays or other open tissues, such as pine sapwood, show a less rapid decrease and a deeper penetration than refractory species such as spruce.

Some migration of preservative and thus a limited equalization of concentration throughout the wood takes place with organic substances by adhesive and capillary creeping of oils and evaporation of gases into the interior of the wood and with water-soluble salts by diffusion, unless they become insoluble through fixation. After months or years the curves of concentration of the preservative may differ remarkably [16]. If evaporation or leaching takes place, the originally high concentration in the layers next to the surface is reduced. The whole curve of concentration representing the distribution becomes more equalized.

The process of leaching was studied under various conditions. With wood species refractory to treatment, leaching takes place somewhat slower than from species with good penetrability [32, 68], but the velocity depends mainly on the speed of diffusion from the interior to the surface of the wood. The reason is that rain can wash away or migration into wet soil can remove only a small amount of salt at the surface of the wood. With poles, observations in connection with renewed treatment by salt bandages have shown that the migration of salts in the axial direction and the loss through the lower end of the pole are more important for the leaching effect than the diffusion in the radial direction [34]. As regards the leachability of elements in combination with chromium compounds, it is highest with boron, followed by fluorine compounds; arsenic compounds and copper are fixed much better [32, 68].

Recent quantitative investigations have contributed to a better understanding of characteristics of treatment processes and of factors influencing the performance of preservatives. A number of questions, however, requires further research.

Final remarks

The need for further research on biological fundamentals and on testing methods has been pointed out in the respective chapters. In spite of the numerous preservatives on the market, the search for new compounds will continue. Products with higher efficacy against lower fungi causing discolouration and soft-rot as well as fungicides and insecticides with low toxicity to human beings are needed for several purposes. The degradability of preservatives by micro-organisms needs further investigation. The idea to develop preservatives with universal efficacy has been given up since preconditions in the fields of application are too diverse. Appropriate recommendations available in many countries provide consumers with the necessary information.

The world-wide discussion and observation of sound ecological conditions for the human population and nature in general has its consequences also for the use of wood preservatives. National committees and an international group are studying the precautions to be taken during treatment with respect to the consumer of treated wood. Antagonistic and other biological effects may be applicable in some special cases. But for part of the wood, chemical preservation is as unavoidable as plant protection. It is only one condition for the safe and successful use of wood and the increase in its consumption. Protection of freshly cut timber is of high importance in tropical countries. The production and transport of veneers in these areas require chemical treatment, which is not always necessary in other climatic zones.

Sources of high-quality wood species are decreasing everywhere in tropical rain forests. The use of other, less known species becomes necessary. Their durability and, if they are susceptible to attack by organisms, their treatability must be tested. In the industrialized countries the use of timber for external work such as windows etc. involves various problems, especially in the case of refractory wood species. The combination of preservation and painting causes several additional difficulties. Reduction of sorption in connection with preservation is to be promoted.

Wood-based materials, plywood as well as particleboards, gain increasing importance as building materials including prefabricated parts. The protection against fungi, termites and Lyctids, which is indispensable under circumstances where attack can be expected, requires further studies. Prefabrication is a general trend in the building industry. The continuing use of solid wood and wood-based materials depends on wood preservation. In this connection quality tests mainly for non-pressure treated wood are required.

Refractory wood species including little known tropical hardwoods and failure of normally successful treatment due to unknown properties of some wood species call for further investigations.

These tasks are to be solved on an international basis. It is evident how useful and important cooperation sponsored and promoted by our International Academy can be in the interdisciplinary field of wood preservation.

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