# WOOD PRESERVATION ENVIRONMENTAL RESEARCH AT THE UNIVERSITY IN HAMBURG

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## 1 Introduction

Wood is an important renewable source of raw material of high economic importance for the building sector, for the making of furniture, and for the packaging industry. All measures taken for the protection of wood serve to maintain its quality and to derive at the highest value during service. This aim can principally be reached by the use of high durable timbers, by taking care of constructive measures and by the use of chemical wood preservatives.

Traditional wood preservatives are based on biocides, which can cause health risks and environmental problems. The application of chemical wood preservatives has been discussed in the past increasingly controversially. Günther Becker and Buchmann (1966) revealed that waterborne wood preservatives can be leached from different timber species, and Gersonde (1959) showed that an envelope treatment of timber can be washed off by rain. The results confirmed the mobility and the tendency of depletion of wood preservatives during service. In the case of Boron this is common knowledge today (Peylo, Willeitner 1995; Peylo, Willeitner 1997).

Wood preservation environmental research at the University of Hamburg and at the Federal Research Centre of Forestry and Forest Products (BFH) began in 1973 when Willeitner evaluated wood preservative measures under environmental aspects and Wischer and Willeitner (1977), and Illner (1988) published results about preservatives leaking of and leaching from freshly treated wood. These results literally expressed the extend of a possible impact of wood preservatives on the environment which was already known by experts and initiated an open discussion by experts. One possible solution to the problem was the introduction of the accelerated fixation method for treated timber with chromium containing wood preservatives (Daljeet Singh, Peek, Tam 1988; Peek 1984, 1988; Peek, Geick, Willeitner 1985; Peek, Klipp 1990; Peek, Willeitner 1981, 1982, 1984, 1986, 1988a,b,c; Peek,Willeitner, Brandt 1987; Willeitner, Peek 1988a,b; Willeitner, Voß, Peek 1986).

## 2 Reactions from the wood preservation industry and research at the University in Hamburg

In the past, the industry and the research laboratories reacted on these new findings and on the pressure of the regulative bodies. End of the 60th the Quats were developed in New Zealand/Australia and have gained a certain importance for timber out of soil contact. In Germany, these products are more and more used.

Since the 80th research began in order to develop wood preservatives free of Chromium. First approvals in Germany have been given in 1988. These are products on the basis of

Cu-HDO, (Cu–) Quats, (Cu–) Triazole, (Cu–)polymer Betain (Härtner et al. 1995) respectively.

In the 80th, the accelerated fixation method for treated timber has been proposed by Peek, Willeitner (1981) and the basic principles have been improved in different countries since then.

The disposal of treated wood waste became an increasing problem in some of the European member states. The question has been carried forward by the European Community and will continue to be a predominant research topic including logistics in waste management, burning, composting and other disposal techniques and the development of quick detection methods for wood preservatives in waste wood (Leithoff et al. 1995; Leithoff; Peek 1997; Leithoff, Peek 1998; Peylo, Peek 1998; Salthammer et al. 1993, 1994a,b, 1995 a,b,c,d; Peek, Stephan, Leithoff 1993; Stephan, Peek 1992; Stephan, Leithoff, Peek 1995; Stephan, Leithoff, Peek 1996; Stephan, Peek, Nimz 1996; Voss, Willeitner 1993; Voss, Willeitner 1994; Voss et al. 1994; Voss, Willeitner 1995).

With the implementation of the Biocidal Products Directive into local law of the different European member states it might be necessary in the future to develop products and new protection principles with a lower risk potential with the aim to reduce the application of traditional wood preservatives or even to resign of some of them. The use of Melamine resins for the protection of wood without biocides or water repellents might be a approach (Leithoff et al. 1999; Leithoff et al. 1999; Lukowsky, Peek 1998; Lukowsky, Peek Rapp 1998; Lukowsky, Rapp, Peek 1998; Militz, Peek 1994; Peek, Militz, Kettenis 1992; Rapp, Peek 1995a,b; Rapp, Peek 1996; Rapp et al. 1998; Sailer, Rapp, Peek 1998).

In the following major results of some Wood Preservation Environmental Research projects at the University in Hamburg in conjunction with the Federal Research Centre for Forestry and Forest Products will be presented. It should be noticed that this is not at all a complete list and the author appologizes if other valuable resaerch is not mentioned but nevertheless highly appreciated.

### 2.1 The waste wood situation

Waste wood is produced in numerous areas of private and public life, in trade and in commerce, and its re-use is increasingly posing problems in Germany. This development has been reinforced as a result of legislation, such as the Economic Circuit and Waste Law (KrW-/AbfG), the law regarding the supervision and control of the cross-frontier transportation of waste (AbVerbrG), the Ordinance for the regulation of waste materials which need particular supervision (BestbüAbfV), the Ordinance for the regulation of the processing of waste materials which need particular supervision (BestüVAbfV), the Technical Guidance on Settlement Waste[1] (TASI) and other regula-

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[1] In the sense of the administrative regulation settlement waste means "refuse such as household refuse, bulky refuse, industrial waste similar to household refuse, park and garden refuse, market refuse, .. construction waste...".

tions. The construction sector can be expected to produce an increased amount of waste wood in the next few years, particularly within the framework of the new building and reconstruction measures in the new German Federal States.

In the Germany, the orderly, environmentally-correct and thus harmless disposal of timber treated with preservatives is still in its initial phase. The relevant regulations are often of only a general nature and lead to uncertainty on the part of the authorities entrusted with the disposal which in turn leads to different requirements being made of the quality of the disposal in different federal states. For the market operator this means that there are unequal conditions in competition.

Problems with the disposal of wood treated with preservatives arise primarily from the wood preservatives. The potential for contamination, however, does not only depend upon the specific properties of the active ingredients used (e.g. it is unequally greater for mercury, arsenic or pentachlorophenol (PCP) than for copper or boron), but also on the quantities contained in the wood.

The so-called additives which, apart from the biocidal substances, are contained in most wood preservatives, in particular all solvent-based wood preservatives, will not be discussed further at this point. When burnt, the content of heavy metals (e.g. lead, zinc) or halogens (e.g. chlorine) is more important, independent of whether they have been applied as active ingredients or as additives. However, it is necessary to mention that it is not sufficient to consider only the biocidal substances when disposing of waste wood.

The different methods of treating wood with chemical protection and their effect on the quantity (specific to the active ingredient ranges from a few gr/m$^3$ in the case of some pyrethroids up to far more than 100 kg/m$^3$ in the case of creosote) and distribution of the preservatives used makes it difficult to classify timber which has been treated with wood preservatives. In addition, the loss of preservative during use (e.g. washing out by rain) must be taken into consideration.

The potential risk from wood treated with wood preservatives depends to a large degree on the type of wood preservative used, as the active ingredients contained in the preservative differ greatly with regard to their humantoxicological and ecotoxicological behaviour. The prerequisite for the orderly and harmless disposal of this timber is, therefore, the exact knowledge of the wood preservatives used and their composition. It is then possible to ensure that the method of disposal is appropriate to the potential risk.

## 2.2 Quick detection of wood preservatives

The possibility of identifying active ingredients in treated wood is of great importance for the its utilisation. Conclusions as to the type and extent of possible contamination can at present only be made to a limited degree if the age, area of application and origin of the treated waste wood are known. This knowledge narrows down the number of possible active ingredients with which the wood was formerly treated. An exception are the "identification nails" attached to telephone poles which supply information about treatment with wood preservatives.

There is to date no simple procedure available for the quick detection of active ingredients in wood which could be used with the required precision and reliability in every day practice. At present a research project at the University of Hamburg deals with the development of quick detection procedures which should make it possible to carry out an examination in seconds. Presently single or combined methods are being developed which can be used to detect both, organic and inorganic contamination in wood in industrial scale.

The most promising techniques for fast analyses are based on Laser-Plasma-Atomic-Emission-Spectroscopy for inorganic agents and Ion-Mobility-Spectroscopy for organic agents. First results show that laser-induced atomic emission spectroscopy and X-ray fluorescent analysis seem to be particularly suitable for the recognition of inorganic substances. For the identification of organic substances, however, apart from costly gas chromatography-mass spectroscopy (GC-MS) there are to date no possibilities for quick detection (Peylo, Peek 1998; Voss et al. 1994).

## 2.3 Disposal of treated wood

Generally, there are four possibilities given for the disposal of treated poles

- Decontamination
- Re-use
- Energy recovery/thermal treatment
- Landfill

### 2.3.1 Decontamination

The term 'decontamination' comprises two different approaches of detoxifying impregnated wood waste which can be carried out mechanically or chemically (biologically). The mechanical decontamination has been proposed for poles treated with mercury (Voss, Willeitner 1994). About 90-98% of the mercury is located in the outer 10 – 20 mm and a separation of highly treated from nearly untreated material seems to be possible. Similar procedures have been discussed for other waterborne wood preservatives. However, this procedure has never reached the marked for economic reasons.

Different methods of a chemical detoxification of treated wood have been described in the past. Inorganic preservatives such as CCA can be extracted e.g. by using sulphuric acid (Honda et al. 1991; Kamdem, Munson 1996; Smith, Shiau 1996; Munson 1997) or organic acids (Leithoff et al. 1995; Peek et al. 1993; Stephan, Peek 1992; Stephan, Nimz 1996; Stephan et al. 1995, 1996) with a high to extremely high technical effort. The amount of elements remaining in the extracted wood (pulp) is low (Cr: 30 – 50 ppm; Cu: 2 - <200 ppm; As: 30 – 50 ppm).

The idea of a biochemical detoxification of treated wood waste originated from a number of cases were wood exposed to ground contact treated with chromium and copper containing preservatives had been destroyed by brown rot fungi. Figure 1 shows the specific preservative tolerance of different isolates against CCA in laboratory tests according to EN 113. Cr was leached between 50% and 60% and As between 60% to

75% of the initial retention. Regardless of the type of preservative (CCB, CC) Cu was leached to less than 7% as from the undecayed controls.

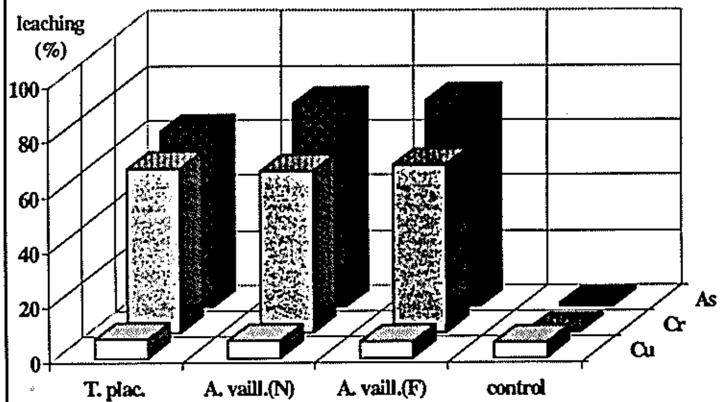


Figure 1: Leaching of a CCA preservative after 16 weeks of fungal decay in % of the initial retention (20 kg/m$^3$).

HPLC-studies showed that the fungi produced large amounts of oxalic acid as well as smaller quantities of acetic and formic acid when colonising the salt-impregnated wood. Cu is precipitated as Cu-oxalate which possesses a low water solubility (25 mg/l) while Cr- and As-oxalate are water soluble and leachable. A second leaching with a solution of 0.2% ammonia in water increased the leachability of copper significantly to 75% of the initial retention.

### 2.3.2 Preliminary results from scale-up experiments

In scale-up experiments feasibility studies for a biochemical processing of treated wood waste were carried out by testing different unsterile inoculation and fermentation methods (Leithoff, Peek 1997, 1998). It was found that a static solid state fermentation in covered containers (about 250 l volume) is promising. Mixing of waste wood chips (80%) made from industrially treated poles with a fresh inoculum (20%, age: 4 weeks) was the most suitable way of inoculation. After a 8-months-fermentation about 80 to 85% of the initial As and Cr could be leached. These tests showed that a solid state fermentation can be carried out successfully under unsterile and unclimated conditions. The experiments showed that the fermentation period can be reduced to 3 to 4 month. Furthermore, it was possible to improve the leaching procedure especially for Cu. However, the process needs further practical experience with scale-up tests simulating the situation of a real plant under practical conditions.

### 2.3.3 The Conversion Process

The flow chart (Figure 2) shows the continuous steps of a possible solid state fermentation and leaching processes. The wood waste is converted into coarse chips and reweted. For inoculation the freshly moistened chips are mixed with about 20% (v/v) inoculum chips from a 4 weeks old pile. During the fermentation the wood moisture content and the temperature of the wood waste chips are monitored. The temperature should not exceed 27-33°C as the fungus stops growing at temperatures above 33°C. Further, the chips have to be reweted when the moisture content drops below 50%. Depending on climate conditions the fermentation process will last between 3-5 month.

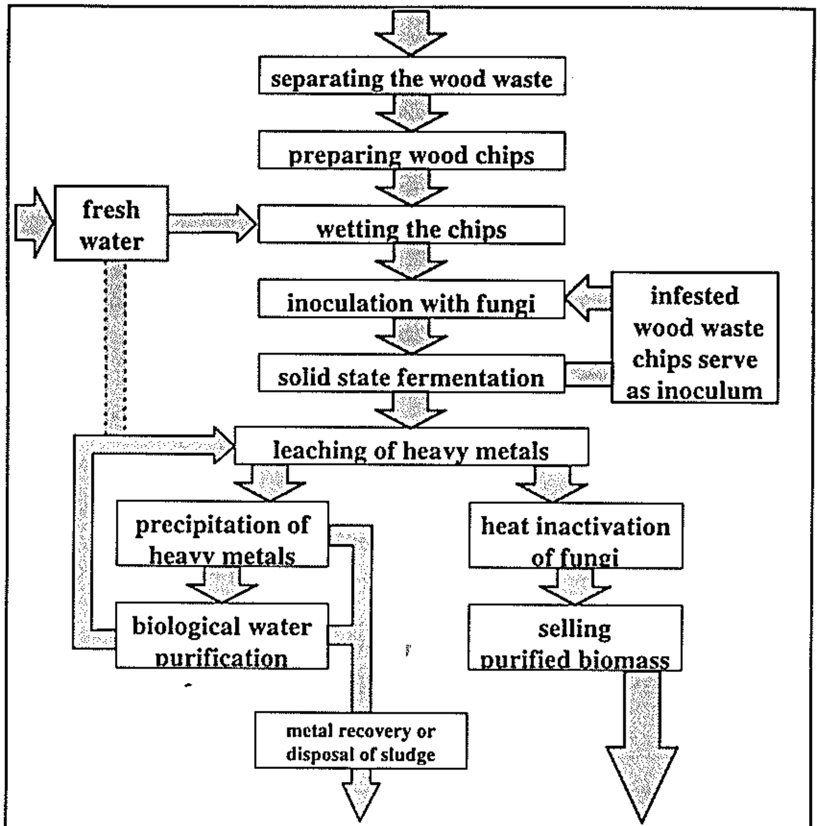


Figure 2: Biological conversion of salt-treated wood waste in a solid state fermentation process using the brown rot fungus *Antrodia vaillantii.* The metal compounds of the preservative are subsequently leached with water/ammonia and recovered as precipitate.

After fermentation the chips are leached with ca. 3-5 vol. water per vol. chips followed by leaching with 1% aqueous ammonia solution and a final leaching with water to recover the surplus ammonia. The leaching procedure is designed as a countercurrent-stream stripping process to save water/ammonia solution and to reduce the amount of run-off water to be cleaned.

The leached chips are dried, sieved and the fungus is inactivated by steaming. For a safe marketing it is necessary to regularly control the heavy metal content of the end product and the inactivation of the fungus.

Methods for the recovery of ammonia, Cr, As and other elements from the leachate is still under development.

## 2.4 Combustion of treated wood

In many cases the combustion of treated wood waste seems to be the most efficient way of disposal. In order to obtain more information about the emission properties, differently treated wood was incinerated in several furnaces after mixing with non-treated wood (Salthammer et al. 1994a,b; Salthammer et al. 1995a,b,c,d).

The results are that the combustion process of residues containing organic or inorganic preservatives is influenced by the elementary composition of the preservative and the thermal and oxidative reaction paths in the flame. Organic preservatives mostly can be thermally cracked by usual combustion conditions. However, elevated conditions are necessary for preservatives based on creosotes.

Among inorganic preservatives, volatile ingredients like fluorine have a considerable environmental impact. Other elements like copper, chromium and boron remain in ashes and cinders reducing the emission towards effective dust removal. It was also found that the concentration of the gaseous emission components CO, $NO_x$ and $C_mH_n$ are not increased in comparison to the values being found of non- treated wood. The concentration of polychlorinated dibenzo-dioxins and –furans in the exhaust gas could be kept low under optimised combustion conditions, however a non-regular incineration process strongly supports increased emissions from PCDD and PCDF.

## 2.5 Fate of preservatives in the environment

In the past, investigations concerning the migration behaviour of wood preservatives were made only to a minor extent and no uniform methods of test exist so far . The migration behaviour of different preservatives without and with Cr was determined using lysimeters of different dimensions (Melcher, Peek 1996; Melcher, Peek 1997; Melcher, Peek 1998). The results of the migration and the distribution of the anions in the soil showed considerable differences between the ions. Approximately the whole quantity of Cr is found in the percolate. That means that Cr migrates very quickly through the soil. In contrast, Cu and F is bound mainly at the top of the column.

From the concentrations of these chemicals in lysimeters and in the percolate it can be concluded that in the case of an accident a contamination of the groundwater by Cr can be expected, however, Cu and F are kept at the surface of the soil (Melcher, Peek 1995).

The very quick movement of Cr was confirmed along with a remedial treatment of a contaminated facility yard (Peek, Illner, Klipp 1992; Peek, Klipp, Brandt 1993; Illner et al. 1996).

## 2.6 Resins in wood protection

In the past decades plastics have received increasing importance in commodities which traditionally belonged to the wood industry. It is therefore obvious to combine the known benefits of the hydrophile lignocellulose framework of wood with other organic material in order to produce a material with a significantly larger range of use. Since about five years a research project is going on at the University of Hamburg and the BFH in which waterbased resins are tested to treat wood for the use in ground contact and under above ground conditions without biocides (Lukowsky, Peek 1998; Lukowsky, Peek, Rapp 1998; Lukowsky, Rapp, Peek 1998; Militz, Peek 1994; Peek 1991; Peek, Militz, Kettenis 1992; Rapp, Peek 1995a,b, 1996; Sailer, Rapp, Peek 1998; Schröder 1983). This research is carried out in close contact with the industry.

The major findings are that the performance of the resins is in most cases independent from the different initial moisture content of the wood. Melamine-resins show the best ASE and absorption results of all resins tested at BFH. ASE of more than 25 % could be achieved with *Pinus radiata*, regardless of whether the samples were treated with low or high resin contents. The most important result is that wood treated with melamine resins shows a high resistance against all kinds of fungi, and has a very high performance especially in soil contact with a high brown rot hazard. Further benefits of Melamine resin treated wood waste are that it can be re-used for particle board production, be burned or even composted and been used as soil fertiliser (Leithoff et al. 1999a,b).

## 3 Outlook

The future development in the wood preservation sector will be characterised by the preference of protection measures without chemical wood preservatives. It also might result in - presumably for a short while - a substitution of timber by other building material. Despite of this, chemical wood preservation will be indispensable for certain commodities, especially for timber in ground contact. It is expected, however, that the position of chemical wood preservation will increase again in the future due to the exaggeration in doing without wood preservatives, as it is noticed at the moment, and defects are programmed.

The latest development in wood preservation is the biocidal products directive 98/8/EC issued by the EU beginning of 1998 which has to be implemented into local law of the European members states until May 2000. Among others, the BPD requires an environmental risk assessment of the treated timber.

This involves questions like:

- What is an environment?
- Does the soil around a treated pole belong to the ‚environment' of the pole or not, where is the boundary?

A research proposal has been prepared and sent to the EU-Commission for financial support in order to develop methods to test possible risks deriving from treated timber in the different hazard classes.

It becomes more and more obvious that research organisations and the industry have to learn to think increasingly in political categories and not in a mixture of scientist, technician or industry manager. Instead of bringing forward rational arguments about the security of plants, treated products or treatment techniques, more and more the restraints and feelings of the public has to be taken into consideration.

The wood preservation industry in Germany takes slowly into consideration alternative protection measures, such as chemical modification of the timber or the combination of waxes and wood preservatives. All new approaches for the protection of wood show a common tendency: to reduce the amount of preservatives. The future will show, whether this is the correct way to go.

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