IRG/WP 01-50170

THE INTERNATIONAL RESEARCH GROUP ON WOOD PRESERVATION

Section 5 Environmental Aspects

Effect of remediation on the release of copper, chromium, and arsenic from particleboard made from CCA treated wood

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Paper prepared for the 32nd Annual Meeting
Nara, Japan
May 20-25th, 2001

IRG Secretariat
SE-100 44 Stockholm
Sweden

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ABSTRACT

This study sought to determine the effect of remediation with oxalic acid (OA) extraction and *Bacillus licheniformis* fermentation on the release of copper, chromium, and arsenic from particleboard made from remediated wood particles and also investigates durability of the particleboard against white and brown-rot fungi. Particleboard samples were manufactured using untreated, CCA-treated, OA-extracted, and bioremediated southern yellow pine particles. Results shows that oxalic acid extraction and bioremediation by *B. licheniformis* significantly increased removal of elements from CCA-treated wood particles. The particleboards containing OA-extracted and bioremediated particles showed generally high leaching losses of remaining elements. Exposure of particleboard samples to decay fungi indicated that *Gloeophyllum trabeum* caused greater weight losses in all samples than *Postia placenta.* In general, leached samples from all particleboard types had greater weight losses than unleached samples. CCA particleboard samples were the most resistant to fungal degradation.

Key words: CCA-treated waste wood; extraction, bioremediation; composite; *Bacillus licheniformis;* white *and brown-rut fungi.*

1: This work was supported by the Research Fund of Istanbul University.

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1. INTRODUCTION

Chromated copper arsenate (CCA) is currently the predominant preservative for various applications and is widely used in the United States. By 1995, 67 million kg of water-borne preservatives were utilized for wood treating purposes and over 90% of this was CCA (Solo-Gabriele et al., 1999). Approximately 17 million cubic meters of CCA-treated wood are generated annually (Cole and Clausen, 1996). It is estimated that the quantity of CCA-treated wood removed from service will increase to 12 million cubic meters by the year 2004 (McQueen and Stevens, 1998). Cooper (1993) predicted that CCA-treated wood being removed from service annually in the United States would increase to 16 million cubic meters by 2020.

Currently, some options for dealing with spent CCA-treated wood include reuse, recycling, incineration, use as fuel, and landfill disposal (Huang and Cooper, 2000). Recycling of treated wood waste into wood based composites is a relatively low-cost alternative to disposal into the environment (Vick et al., 1996). Recycling treated wood into composites is not a new concept. Several studies have recently concentrated on wood-based and wood-cement based composites from CCA-treated wood and compatibility of copper, chromium, and arsenic with adhesives and cement in composites (Boggio and Gertjenjansen, 1982; Hall et al., 1982; Huang and Cooper, 2000; Kamdem and Munson, 1996; Munson and Kamdem, 1998; Schmidt et al., 1994; Vick et al., 1996). However, concerns about introducing preservatives into manufacturing facilities and concerns about leaching from composites made from waste treated wood have limited this management option.

Extraction of the CCA elements from the wood fiber can increase recycling opportunities for the remaining wood pulps. One novel method for recycling CCA-treated wood fiber would be to modify it by removing all or much of the heavy metal components so that both the wood fiber and the metals could be reclaimed. Acid extraction, one option for removal of copper, chromium, and arsenic from treated wood fiber, has been explored by several researchers using different acids (Kim and Kim, 1993; Pasek, 1994; Stephan et al., 1993). Clausen and Smith (1998) and Stephan et al., (1993) showed that the removal of copper, chromium, and arsenic from CCA-treated wood waste increased significantly during oxalic acid (OA) extraction. Bacterial fermentation is another possible method for removal of heavy metals from treated wood since some bacteria are extremely tolerant of toxic metals (Cole and Clausen, 1996; Daniel et al., 1987; Daniel and Nilson, 1985). The mode of action for bacterial removal of heavy metals is often conversion of CCA elements into their water-soluble form. Once converted, copper, chromium, and arsenic can be removed from the wood through a washing process (Felton and DeGroot, 1996). Clausen (1997) Clausen and Smith (1998) Cole and Clausen (1996) and Crawford and Clausen (1999) showed that *Bacillius licheniformis* isolated on CCA-treated wood had a great potential to remove toxic metals when treated wood sawdust was exposed to this organism in liquid culture.

We previously showed that the properties of particleboard made from CCA-treated and remediated wood waste were diminished due to the remedial treatments

(Clausen et al., 2000). The objectives of the present work were to evaluate (i) the leachability of copper, chromium, and arsenic from particles and particleboard made from CCA-treated and remediated wood fibers and (ii) residual decay resistance of the particleboard made from recycled and remediated fibers.

2. MATERIALS AND METHODS

Remediation of CCA-treated wood particles

Oxalic acid (OA) extraction

Two kg of CCA-treated (6.4 kg/m^3) wood particles (hammer milled to 6-16 mesh (3360-1190 µm) were extracted with 18 L of 0.8% OA (pH: 1.7) (Sigma Chemicals, St. Louis, MO) for 18 hour at 25°C in 18 L polypropylene carboys.

Bioremediation

OA was siphoned off and 18 L nutrient broth (Difco, Detroit, MI) prepared according to manufacturer's directions, were added and inoculated with 500 mL of an 18-h culture of *Bacillus licheniformis* CC01. Carboys were then incubated at 28°C and stirred at 100 rpm for 10 days. Spent medium was siphoned off and bioremediated chips were collected on cheesecloth covered screens and oven dried at 60°C (Clausen, 2000; Clausen and Smith, 1998).

Particleboard production

Two 406.4 mm square particleboard panels by 6.4 mm thick, with an approximate specific gravity of 0.80, were manufactured from each type of particles (untreated, CCA-C treated, OA-extracted and bioremediated southern yellow pine). Eight hundred ninety-nine grams of each of four particle types (control, CCA-treated, OA-extracted and bioremediated) were blended with 10% urea formaldehyde (UF) resin (Southeastern Adhesives' 9-2035, Lenoir, NC, U.S.A.). The UF resin was applied in a rotating drum blender using an atomizing Binks spray gun. Particleboard panels were formed by hot-pressing for 5 min to an internal temperature of 121°C. The boards were then trimmed to 406.4 by 406.4 mm and conditioned at 20°C and 65% relative humidity (RH) for 2 weeks (Youngquist, 1999).

Leaching tests

Leachability of copper, chromium, and arsenic components was evaluated on particleboard samples and wood particles according to modified AWPA E11-87 test method (AWPA 1999). The samples with dimensions 19 by 19 by 6.4 mm were cut from the particleboard panels. Samples of wood particles used for particleboard manufacture, equal to the particle weight in the particleboard samples, were exposed to the same leaching test for comparison. Wood particles and particleboard samples were put into glass containers and leached with deionized water using the water-to-wood ratio

specified by AWPA E11-87 test method (AWPA 1999). The water was changed after 6, 24, 48 hours and thereafter at 48 hours intervals, over a total of 14 days. Water samples were collected after each water replacement and the water samples were analyzed for copper, chromium, and arsenic with a Perkin Elmer 5100 PC Atomic Absorption Spectrometer (AAS) (Perkin Elmer, Norwalk, CT, U.S.A.) using flame atomization for higher concentrations and graphite furnace atomization for lower concentrations of the elements to estimate total amounts of CCA elements leached from the board samples and wood particles. The results were expressed as percentage of copper, chromium, and arsenic leached, relative to the amounts contained in the board samples and wood particles originally. Two replicate leaching tests were completed for each board sample and wood particle type.

Analysis of CCA retention

Before and after the leaching tests, the particles and particleboard samples were ground to pass a U.S. Standard 30-mesh (589 µm) screen and analyzed for amount of copper, chromium, and arsenic using an ASOMA X-ray fluorescence spectroscopy (ASOMA Instruments, Austin, TX, U.S.A.) according to AWPA A9-99 test method (AWPA 1999). Percentage reduction of copper, chromium, and arsenic in OA-extracted and bioremediated particles after remediation processes were calculated based on the amount of elements in CCA-treated particles.

Fungal decay resistance tests

Unleached and leached particleboard samples were subjected to fungal decay resistance test. Four replicate samples of each particleboard type were dried to constant weight and steam-sterilized at 100°C, weighed, and exposed to two brown rot fungi, *Gloeophyllum trabeum* (Mad 617) and *Postia placenta* (Mad 698) and the white rot fungus, *Trametes versicolor* (Mad 697) in a modified soil-block test according to AWPA E10-91 test method (AWPA 1999). After 12 weeks of incubation at 27°C and 70% RH, the surface fungus mycelium was removed, the samples were dried at 60°C, and weight losses were determined as percentage of total particleboard mass.

3. RESULTS AND DISCUSSION

Copper, chromium, and arsenic content in particles after remediation

Results of ASOMA X-ray fluorescence spectroscopy analysis for copper, chromium, and arsenic following OA-extraction and bioremediation by bacterial fermentation as well as percentage of elements released after remediation processes based on the amounts of elements in CCA-treated particles are shown in Table 1 and Table 2, respectively. OA-extraction removed about 23.1% CuO, 65.0% CrO_3, and 74.2% As_2O_5 from CCA-treated particles. Further exposure of wood particles to *Bacillus licheniformis* fermentation enhanced the removal of metals compared to removal of elements from CCA-treated wood particles. Acid extraction followed by bacterial fermentation removed about 62.0% CuO, 79.2% CrO_3, and 90.3% As_2O_5 of initial

concentration of these elements in CCA-treated particles. In other words, bioremediation by *B. licheniformis* resulted in about 39% more CuO, 15% more CrO_3, and 16% more As_2O_5 removal than OA-extraction alone. These results show that chemical modification via OA-extraction may act to partially reverse the fixation process of copper, chromium, and arsenic in wood (Clausen and Smith, 1998).

Leachability of copper, chromium, and arsenic

The percentage of copper, chromium, and arsenic leached from the wood particles is shown in Figure 1, 2, and 3, respectively. Total percentage of copper released from OA-extracted particles were considerably higher than from CCA-treated and bioremediated particles (Table 2). The percentage of copper leached from OA-extracted particles was between 2.8% and 9.6% for each leaching time (Figure 1). Compared to copper, the leaching rate of chromium was much greater after 6-h and 1-day of leaching, although the total percentage of chromium released from OA-extracted particles was smaller (Figure 2). Total percentage of chromium leached from CCA-treated particles was 2.6%, whilst 26.5% leached from bioremediated particles (Table 2). It was unexpected that high levels of arsenic would leach from CCA-treated particles compared to copper and chromium leaching rates (Figure 3 and Table 2). While the total percentage of arsenic released from CCA-treated particles exceeded Cu and Cr by 4 to 6-fold, total arsenic leached from OA-extracted particles was 1.8-1.6 times less than copper and chromium (Table 2).

Figure 1, 2, and 3 show the percentage of copper, chromium, and arsenic released from the particleboard samples during the leaching tests. The total percentage of copper released from the boards with CCA-treated particles was insignificant compared to that of the other types of particleboard. Compared to CCA-treated particles, the total percentage of copper removed from the particleboard was smaller. In OA-extracted particleboard samples, the total percentage of copper leached was 42.6% less than OA-extracted particles. In contrast to these results, the total percentage of copper leached from bioremediated particleboard samples increased 32.1% compared to the same type of particles. The percentage of chromium released from the boards made from CCA-treated and OA-extracted particles was not significantly different than that from the particles (Table 2). However, percentage of chromium released from the board samples containing bioremediated particles increased 17.3% compared to bioremediated particles. In contrast to copper and chromium leaching rates, more arsenic was released from particleboard samples with CCA-treated and OA-extracted particles than the comparable particles. The arsenic leached was 40.5% greater in board samples made from CCA-treated particles, while the total percentage of arsenic leached from OA-extracted particleboard samples was approximately 2 times greater than that released from OA-extracted particles.

The total percentage of elements leached from OA-extracted and bioremediated particles was considerably higher than that of CCA-treated particles (Table 2). It may be concluded that exposure of CCA-treated particles to OA-extraction and bacterial fermentation may reverse the CCA fixation process (Smith and Shiau 1997; Stephan et al., 1993). We also conclude that the remediation processes used in this study created

chemical modification on the surfaces of wood particles, essentially reversing CCA fixation. Leaching of residual CCA was even greater after particles (CCA-treated, OA-extracted, and bioremediated) were glued with UF resin, indicating the resin further interfered with fixed CCA components.

Decay resistance of particleboard samples

Gloeophyllum trabeum caused greater weight losses in all particleboard samples than *Postia placenta* or *Trametes versicolor* (Figure 4). The greatest weight losses were seen in control particleboard samples (22% in leached, 38% in unleached samples) and bioremediated particleboard samples (41% in leached, 20% in unleached samples). In unleached CCA particleboard samples, weight loss (5.9%) was much lower than those of the other particleboard samples.

Unleached and leached particleboard samples containing CCA-treated particles had the least weight losses after a 12-week exposure to *P. placenta* (Figure 5). Although weight losses in leached control particleboard samples and particleboard samples with OA-extracted and bioremediated particles were greater than 20%, weight losses were remarkably lower in unleached samples compared to leached samples. It appears that *P. placenta* cannot readily penetrate the UF resin in unleached control samples to gain access to the wood fiber. This is illustrated by increased weight losses for all leached samples.

Compared to *G. trabeum* and *P. placenta, T. versicolor* caused lower weight losses in leached particleboard samples because white-rot fungi preferentially decay hardwoods although little is known about treatability of decay fungi to deteriorate UF particleboard (Figure 6). However, weight losses in unleached samples were slightly greater that those of control samples and particleboard samples with OA-extracted and bioremediated particles exposed to *P. placenta.* Particleboard samples made from CCA-treated particles showed the greatest resistance to *T. versicolor.*

4. CONCLUSIONS

The study shows that oxalic acid extraction and *Bacillus licheniformis* fermentation remediated CCA-treated waste wood via partial removal of copper, chromium, and arsenic. In this study, particleboard made from CCA-treated wood particles seems to be compatible with UF-bonded boards with regard to leachability of CCA elements and biologic efficacy against brown-rot and white-rot decay fungi however arsenic leaching was relatively high. On the other hand, the particleboard samples made from remediated particles show high leaching losses of CCA components. Kamdem and Munson (1996) showed that the particleboard prepared with UF resins leached more CCA components than phenol formaldehyde (PF) resins because UF resins are not water resistant. Our results also showed that leaching of particleboard samples containing OA-extracted and bioremediated particles resulted in higher release rate of remaining CCA components compared to CCA-particleboard samples.

Since UF resins are not water resistant, alternative adhesive systems might be desirable for composite production from remediated fibers depending on their intended use in order to reduce leached CCA components. For example, use of inorganic matrix systems such as cement might be result in improved properties allowing compatibility between CCA-treated and bioremediated fibers and cement. Remediated wood fibers would also provide an excellent source as reinforcing "fillers" in thermoplastic composites.

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Table 1. Percentage reduction of CCA elements in wood particles after remediation process.

Particle type	Reduction (%)		
	CuO	CrO_3	As_2O_5
OA-extracted particles	23.1	65.0	74.2
Bioremediated particles	62.0	79.2	90.3

Table 2. CCA components released from particles and particleboard samples based on original amounts of components

Particle and Particleboard Type	CCA components before leaching			Total leached CCA components[1]		
	CuO %	CrO_3 %	As_2O_5 %	CuO %	CrO_3 %	As_2O_5 %
CCA-treated particles	0,26	0,48	0,31	3,66 *0,00*	2,59 *0,00*	15,62 *0,00*
OA-extracted particles	0,20	0,17	0,08	61,25 *0,15*	56,22 *1,86*	33,88 *4,10*
Bioremediated particles	0,10	0,10	0,03	28,49 *0,21*	26,47 *0,14*	48,49 *0,05*
CCA particleboard	0,22	0,36	0,19	2,60 *0,04*	2,37 *0,00*	21,95 *0,10*
OA-extracted particleboard	0,18	0,12	0,05	35,18 *0,08*	54,71 *0,36*	74,57 *0,31*
Bioremediated particleboard	0,09	0,07	0,03	37,62 *0,16*	31,05 *0,53*	34,58 *0,31*

[1)] Each value represents the means of 2 replicates of 6 samples. Values in italics are standard deviations.

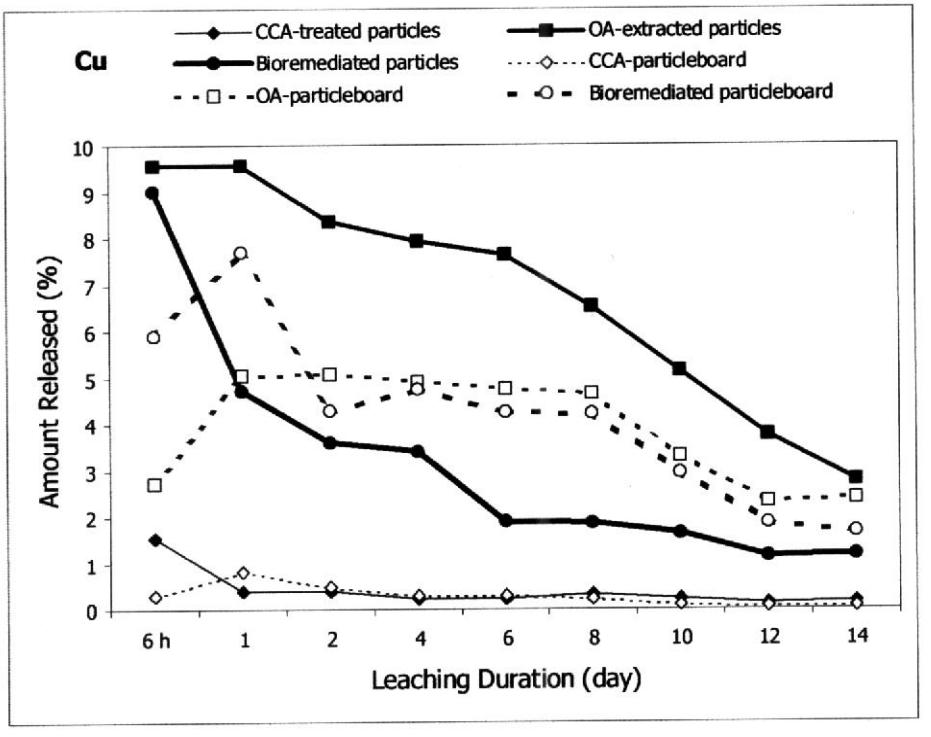

Figure 1. Amount released of copper (Cu) from particles and particleboard samples.

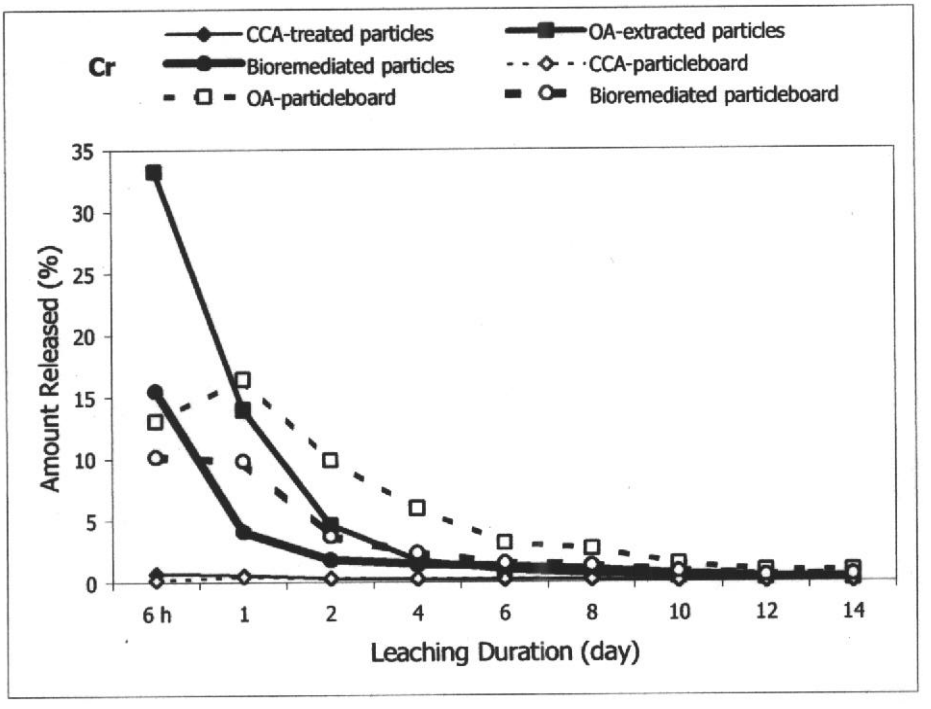

Figure 2. Amount released of chromium (Cr) from particles and particleboard samples.

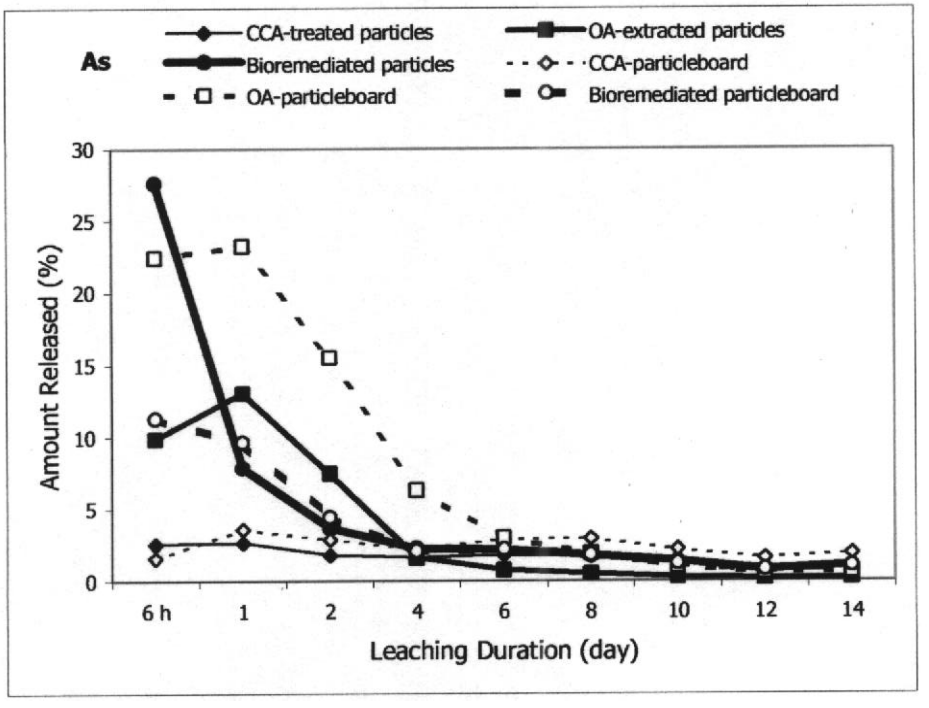

Figure 3. Amount released of arsenic (As) from particles and particleboard samples.

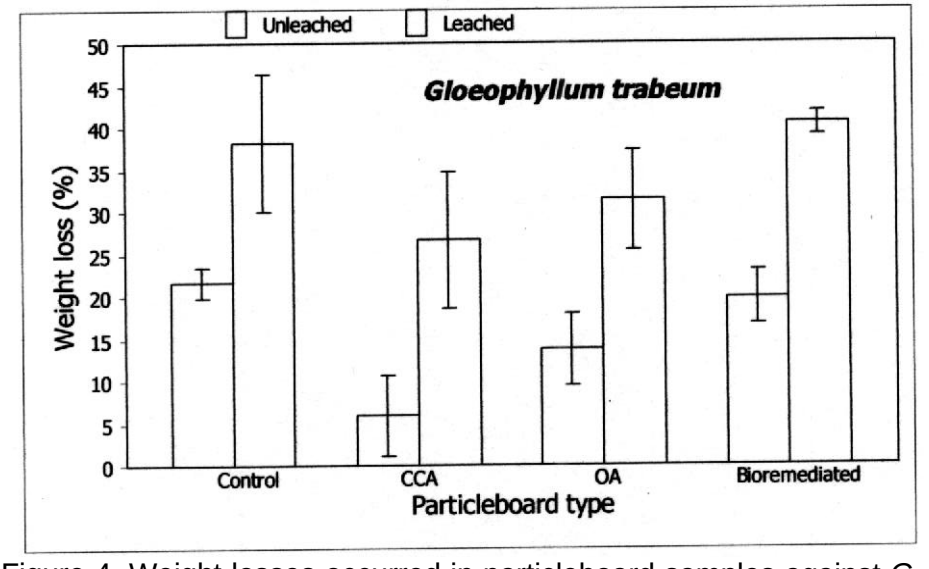

Figure 4. Weight losses occurred in particleboard samples against *G. trabeum.*

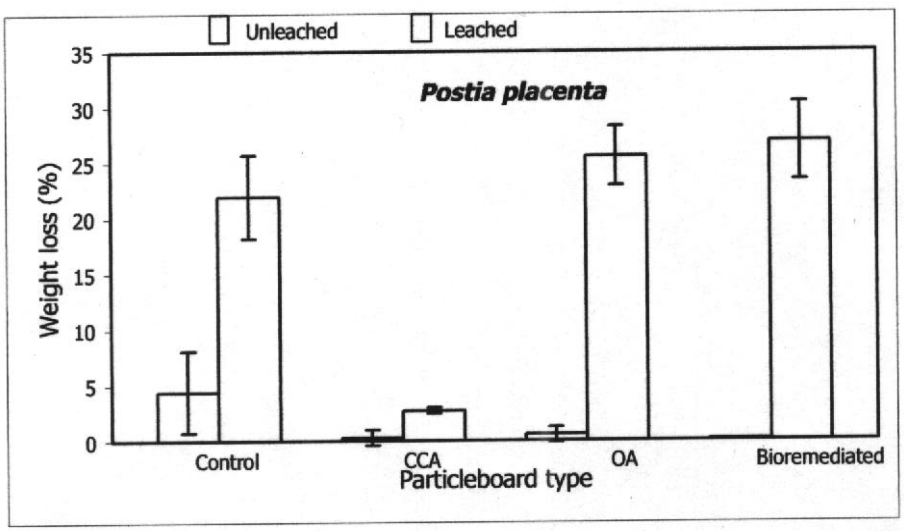

Figure 5. Weight losses occurred in particleboard samples against *P. placenta.*

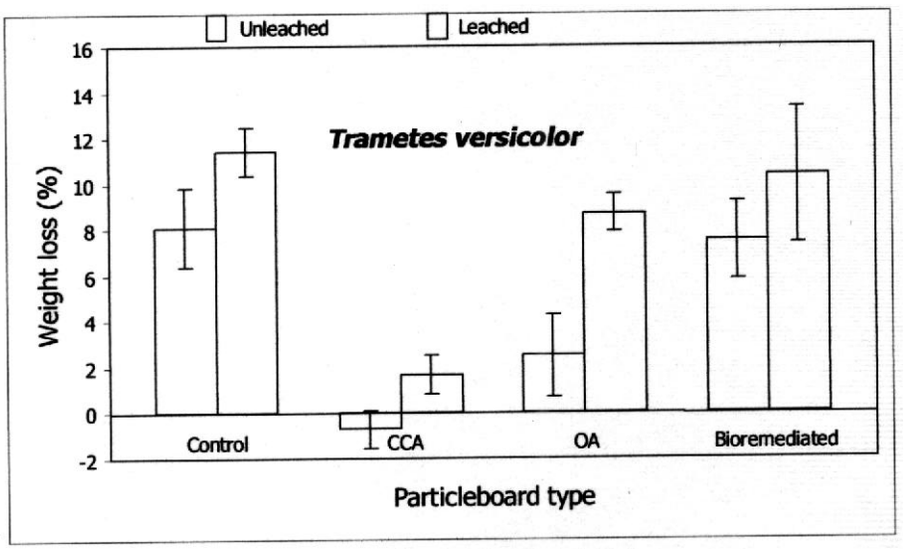

Figure 6. Weight losses occurred in particleboard samples against *T. versicolor.*