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CCA removal from treated wood using a dual remediation process

Novel approaches to remediate chromated-copper-arsenate (CCA)-treated waste wood are needed to divert this material from landfills. In this study, CCA-treated wood wafers were treated with a two-step remediation process to remove copper, chromium and arsenic. Process conditions, involving an acid extraction and bacterial culture with a metal-tolerant bacterium, were optimized. It was determined that 18 h of exposure to 1.0% oxalic acid was an optimal exposure time for the size of wood used in this study. Oxalic acid concentration was then varied to determine optimal concentration for metal removal. Increasing concentrations of oxalic acid removed increasing quantities of copper, chromium and arsenic. Acid extraction at 0.80% and bacterium exposure resulted in a total reduction of 78% copper, 97% chromium and 93% arsenic. An 0.8% of oxalic acid followed by bacterial culture with *Bacillus licheniformis* CC01 afforded the best conditions for maximum removal of copper, chromium, and arsenic from treated wood with minimal exposure of the wood fiber to acid conditions.

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Introduction

Chromated copper arsenate (CCA) is the primary wood preservative used to treat more than 80% of all treated lumber in the United States (Micklewright 1994). The increase in demand for CCA-treated wood products since the preservative gained popularity in the early 1970s has resulted in an annual production of an estimated 1.4×10^7 m^3 (6×10^9 board feet) of this product (Micklewright 1994). This material, as it comes out of service, is currently placed in approved landfills, although future removal of CCA-treated wood products from service is going to greatly increase the demand on landfill space. There are also concerns about environmental contamination from chemicals leaching from treated wood, both in service and after the wood is removed from service and placed in landfills. Thus, remediation methods that divert this material for reuse will alleviate contamination concerns as well as decrease the demand on landfill space.

The quantity of CCA-treated wood that will be removed from service around the turn of the century represents a substantial wood fiber source. Based on an average service life of 20 to 50 yr, Cooper (1993) estimates 1.6×10^7 m^3 of CCA-treated wood will be removed from service in the United States annually by 2020. This estimate is modest. In a survey on the service life of decks, McQueen *et al.* (1998) found that the actual service life of a deck (average of 9 yr) is much shorter than the expected functional service. They

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also determined that 43% of the time, deck replacements resulted from aesthetics, either of the wood or the deck design.

Acid extraction as a means of chemically leaching copper, chromium and arsenic from treated wood has been explored (Kim & Kim 1993; Stephan et al. 1993, 1996; Pasek 1994; Clausen & Smith 1998*a,b*). Stephan *et al.* (1993, 1996) studied microbial conversion of CCA-treated wood with *Antrodia vaillantii*, a brown-rot fungus known for its copper tolerance. *A. vaillantii* produces high levels of oxalic acid, thereby increasing the acidity in the substrate. These same authors postulate that the high production of oxalic acid increases the acidity of the substrate, thereby increasing the solubility of chromium and arsenic.

Minimal exposure of wood fiber to acids is desirable because prolonged exposure to a strong acid, such as sulfuric, damages the fiber integrity by hydrolyzing carbohydrates. Warner & Solomon (1990) reported that 100% copper was found in the leachate after 40 days of continual leaching of CCA-treated wood with NaOH/citric acid buffer at pH 3.5. Cooper (1991) also demonstrated that this high level of copper loss was caused by the citric acid buffer rather than simply by low pH. Clausen & Smith (1998*a,b*) noted a cumulative effect of oxalic acid extraction and bacterial culture with *Bacillus licheniformis* CC01 on metal removal from CCA-treated wood chips. They postulated that exposure of treated wood to oxalic acid partially unfixed CCA components and allowed the bacterium to remove metals in the wood beyond the level achieved by the bacterium alone.

The objective of this study was to optimize the oxalic acid extraction parameters in relationship to the bacterial culturing in order to maximize the removal of copper, chromium and arsenic from CCA-treated wood, while minimizing exposure of the wood fiber to the acid.

Material and methods

Acid extraction time optimization

An aqueous 1% solution of oxalic acid was used to determine the optimal exposure for extraction of copper, chromium and arsenic from wood wafers (1.9 cm by 1.9 cm by 0.32 cm) treated with CCA type C using a full-cell process. A 1% active ingredient was used to obtain a nominal 6.4 kg m^{-3} retention; however, the actual retention was determined by atomic absorption spectrometry (AAS) according to the American Wood Preserver's Association standard A11-93 (AWPA 1995). Sixteen CCA-treated wafers (quartersawn Southern Pine sapwood) were soaked in 100 ml of 1% oxalic acid solution for 4, 8, 18 or 24 h at 25°C and rotated at a speed of 10.47 rad s^{-1} (100 r.p.m.). Control wafers were soaked in water for 24 h at 25°C and rotated at 10.47 rad s^{-1} (100 r.p.m.). Four wafers were removed at each time interval. Two of these wafers were oven-dried at 60°C for 24 h, ground to 20 mesh (0.841-mm sieve openings), and analyzed for copper, chromium, and arsenic values by AAS.

Bacterial fermentation of acid-extracted chips

The two wafers that were acid-extracted for each time interval were additionally treated by exposure to *Bacillus licheniformis* CC01 (Cole & Clausen 1997) in 100 ml of 0.8% nutrient broth (Difco Laboratories, Detroit, MI, USA) for 7 d at 28°C and 15.7 rad s^{-1} (150 r.p.m.). Control wafers were incubated in uninoculated nutrient broth under the same conditions. The wafers were then dried at 60°C for 24 h, ground, and analyzed by AAS as described above.

Acid extraction concentration optimization

Four CCA-treated wafers were placed in each of 10 concentration percentages of oxalic acid (0.0, 0.04, 0.06, 0.08, 0.10, 0.20, 0.40, 0.60, 0.80 and 1.00) for 18 h at 25°C and 10.47 rad s^{-1} (100 r.p.m.). Controls were soaked in water under identical conditions. Two wafers were dried at 60°C for 24 h, ground and analyzed as described above. Two wafers were exposed to *Bacillus licheniformis* CC01 as described before. The wafers were then dried at 60°C, ground and analyzed by AAS as described above.

Results and discussion

Optimal extraction time

Optimal acid extraction time was determined by exposing CCA-treated wafers to an aqueous 1% solution of oxalic acid for 0, 4, 8, 18 or 24 h. Test wafers were then analyzed or additionally exposed to the metal-tolerant bacterium Bacillus *licheniformis* CC01 for a single incubation time. Fig. 1 shows the cumulative effect of acid extraction time on the removal of copper, chromium, and arsenic (mg metal per g of wood). The greatest metal removal was demonstrated at 8 h for copper and 24 h for chromium and arsenic in wafers exposed solely to oxalic acid. However, when the wafers were subsequently exposed to the bacterium, the largest metal depletion was seen after 8 h exposure to oxalic acid for copper (80% removal) and 18 h for both chromium and arsenic, 80% and 91%, respectively.

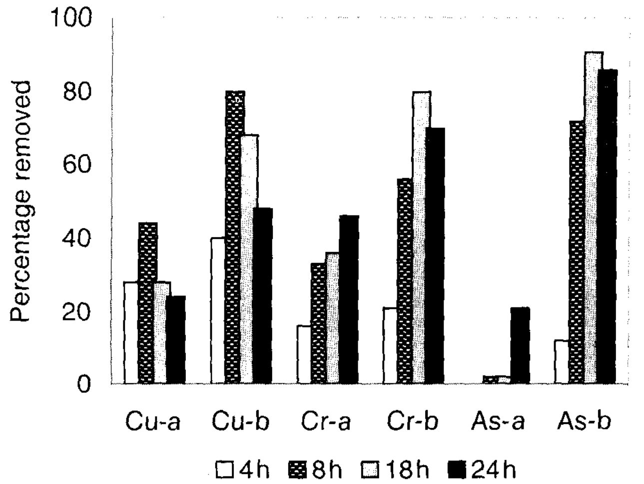

Fig. 1. Percentage of copper, chromium and arsenic removed following (a) 1% oxalic acid extraction for varying amounts of time and (b) 1% oxalic acid extraction for varying amounts of time followed by 7-d bacterial fermentation of CCA-treated wood chips. CCA-treated wood contained 2.50 mg g^{-1} copper, 4.06 mg g^{-1} chromium, and 2.15 mg g^{-1} arsenic at T_0.

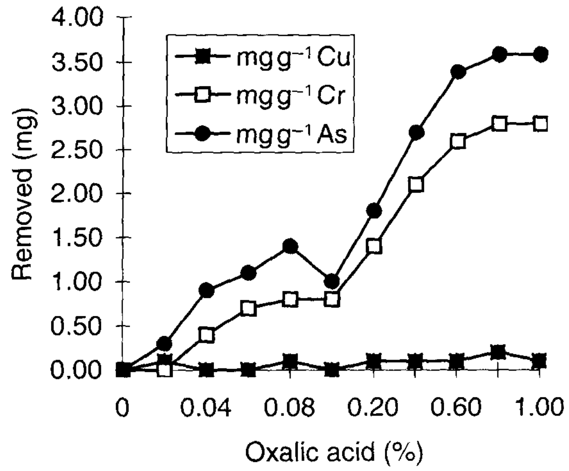

Fig. 2. Amount of copper (Cu), chromium (Cr) and arsenic (As) removed from CCA-treated wafers after combined exposure to oxalic acid extraction for 18 h at various concentrations and bacterial culture.

Optimal oxalic acid concentration

An 18-h exposure was selected for determining the optimal oxalic acid concentration for maximum removal of copper, chromium, and arsenic following bacterial culture with *Bacillus licheniformis* CC01. Table 1 shows the oxalic acid concentration levels tested, pH of each test solution, and mg of each metal per g of treated wood following oxalic acid extraction. Values represent the average of duplicate samples. There was a distinct decrease in the color of the wood as the oxalic acid concentration increased, especially > 0.10%, although no concentration removed enough chemical to bleach the wafers to the color of untreated Southern Pine. Variations were observed in control values (Fig. 1, Table 1). Untreated control values for each experiment were analyzed independent of prior experimental controls. These differences may have been the result of a variable uptake of chemical during treatment.

Combined acid extraction and bacterial culture

Fig. 2 demonstrates the amount of metal removal from CCA wafers that were exposed to the combined acid extraction for 18 h with various concentrations of oxalic acid followed by bacterial culture with *Bacillus licheniformis* CC01 for 7 d at 28°C. Values represent the average of two samples. The theory that oxalic acid increases the solubility of chromium and arsenic (Stephan *et al.* 1993, 1996) may account for the additive effect when acid extraction was followed by bacterial culture. Any increase in solubility of chromium and arsenic may enable *Bacillus licheniformis* CC01 to more efficiently release chromium and arsenic from treated wood, which this bacterium would otherwise have had difficulty removing.

Copper remained relatively unchanged, although note that 11% to 35% of the copper was removed from the original sample solely by oxalic acid extraction, and the bacterial controls showed a removal of 64% of the copper. Exposure to the bacterium released between 55% and 75% of

Table 1. Amount of copper, chromium, and arsenic remaining in CCA-treated wafers following exposure to various concentrations of oxalic acid for 18 h.

% oxalic acid	pH	Copper (mg g^{-1})	Chromium (mg g^{-1})	Arsenic (mg g^{-1})
0*	5.5	1.37	3.58	1.50
0.02	2.8	0.89	3.20	1.16
0.04	2.5	0.94	3.27	1.36
0.06	2.3	0.94	3.20	1.12
0.08	2.2	1.03	2.63	0.96
0.10	2.1	1.05	3.26	1.17
0.20	1.9	0.95	2.35	0.82
0.40	1.7	1.17	2.38	1.00
0.60	1.6	1.13	2.13	0.70
0.80	1.5	1.22	1.89	0.69
1.00	1.4	1.12	1.61	0.53

Control value represents the amount of metal in CCA wafers soaked in water for 18 h.

the copper remaining in the wood sample after treatment at each concentration of oxalic acid. Previous studies using 1% oxalic acid to extract chipped CCA wood showed an additive effect when acid extraction was followed by bacterial culture to remove approximately 90% copper (Clausen & Smith 1998*a,b*). Stephan *et al.* (1996) believe that treatment of CCA-treated wood with oxalic acid leads to the formation of copper oxalate in the wood. Copper oxalate is an insoluble precipitate; therefore, its formation may prevent additional leaching of copper by the acid under certain conditions.

Conclusions

Data obtained in this study suggest that an 18-h exposure time to oxalic acid provides optimal release of copper, chromium and arsenic from CCA-treated wood wafers (1.9 cm by 1.9 cm by 0.32 cm). No additional removal was seen by increasing the extraction time. The cumulative effects of oxalic acid extraction and bacterial culture on the removal of chromium and arsenic were demonstrated when CCA-treated wood was extracted in at least 0.60% oxalic acid. By this dual remediation process, 92% chromium and 80% arsenic were removed with 0.60% oxalic acid extraction. Copper removal was not increased by oxalic acid extraction. Rather, exposure to *Bacillus licheniformis* CC01 was solely effective at removing 64% of the copper. An additional 14% copper was removed in samples extracted with 0.80% oxalic acid. Results suggest that a dual remediation process involving acid extraction and exposure to metal-tolerant bacteria removes significant quantities of metals from CCA-treated wood. Under these conditions, 0.8% oxalic acid extraction for 18 h and 7 d bacterial culture with *Bacillus licheniformis* CC01 were effective at removing 78% copper, 97% chromium and 93% arsenic from CCA-treated wood. The implications of this research reach beyond diverting this waste material from landfills and preventing possible groundwater contamination. This method is a novel, environmentally benign approach for recycling a significant fiber source into value-added composite products.

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