

Leaching Behavior and Flux of Toxic Metals from Chromated Copper Arsenate-Treated Wood and Chromated Copper Arsenate-Treated Wood Ash

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Wood preserved with chromated copper arsenate (CCA) chemical is currently the most commonly used treated wood in Korea. It represents approximately more than 90 % of the treated wood market by volume in Korea (KFRI, 2002). It is widely used to build structures such as decks, fences, poles, pilings, playgrounds and picnic tables because of its excellent performance in prolonging the structural integrity. The copper within the wood serves as the fungicide, while the arsenic protects against insects. Chromium acts as a fixing agent to bind the metals to the wood. The metals in CCA solution are introduced into the pore space of the wood under high pressure. It is reported that approximately 227 metric tons of CCA chemical was imported and utilized to produce approximately 120,000m³ of treated wood in 2002 (KEI, 2004). In recent years, a significant amount of CCA-treated wood from foreign countries such as Canada and the United States is being imported as well.

While CCA chemicals are considered “fixed” from a wood preservation efficacy standpoint, a number of researchers have reported that chromium, copper and arsenic do leach from CCA-treated wood over time when exposed to water or when disposed of in the environment (Cooper, 1991; van Eetvelde et al., 1995; Cooper et al., 1997; Hingston et al., 2001; Jambeck, 2004; Stook et al., 2005; Townsend et al., 2005). The potential impacts of CCA preservative on human health and the environment via leaching have been investigated by a number of researchers (Weis et al., 1992; Rahman and Hughes, 1994; Anderson et al., 1996; Stilwell and Gorny, 1997; Weis et al., 1998; Jambeck, 2004; Townsend et al., 2005). Leaching risk may be evaluated either by theoretical predictions of the total fraction of the metal concentration expected to leach, or by conducting leaching tests. A number of standard short-term (typically <24 hr) batch leaching protocols that have been proposed and used for CCA-treated wood include the toxicity characteristic leaching procedure (TCLP, EPA SW-846 method 1311, US EPA, 1996) and the synthetic precipitation leaching procedure (SPLP, EPA SW-846 method 1312, US EPA, 1996), Standard M-10 (AWPA, 1983), and British Standard EN.84 (BSI, 1997). Results of these short-term leaching tests have often been used for regulatory decisions on management of discarded CCA-treated wood. For example, in the US, TCLP results of CCA-treated wood are compared to risk-based target levels (e.g., toxicity characteristic limits) to assess potential impacts on waste disposal systems such as landfills. The TCLP is designed to simulate the leaching

expected to occur in a municipal solid waste landfill under the acid-forming waste decomposition conditions.

Although many short-term leaching tests have been routinely used for regulatory purposes and in risk assessments at a fixed solid-to-liquid ratio, most tests do not provide the kinetic behavior and release rate of chromium, copper, and arsenic from CCA-treated wood in the environment. It should be recognized that potential environmental impacts are highly dependent upon the rate at which the toxic metals are released. The amount of time during which a leaching solution is in contact with CCA-treated wood samples may influence the quantity of contaminant leached unless equilibrium conditions are established. Previous studies have shown that CCA-treated wood leaches preservative compounds at greater rate when first exposed to leaching solution as compared to prolonged exposure period of time (Lebow et al., 1999; Hingston et al., 2001). Thus, extrapolating short-term data may lead to an overestimation of losses of metals expected during the longer service life or disposal period of CCA-treated wood. Limited studies regarding kinetic leaching of toxic metals from CCA-treated wood and CCA-wood ash have been conducted.

The objectives of this work were to examine the leaching dynamics of copper, chromium, and arsenic from CCA-treated wood and CCA-wood ash by means of laboratory leaching tests and to determine the release rate of toxic metals from the wood and the ash. Leaching studies of up to 60 days duration were conducted on different types of CCA-wood materials in leaching solution. Results obtained from this study allow for a better understanding of the leachability of the metals from different CCA matrixes and may be used with site specific data to provide a more thorough assessment of leaching risk.

MATERIALS AND METHODS

New CCA-treated dimensional lumber (i.e., wood that had not been exposed to weathering in the environment) was obtained from a local CCA-treated wood manufacturer in Chungnam Province, Korea. The lumber (Western hemlock-*Tsuga heterophylla*) was treated with 2.0% CCA Type C (CrO_3 47.5%, CuO 18.5%, and As_2O_3 34.0%) in accordance with Korea Wood Preservation Standard (KFS, 2003). The CCA retention level of this lumber was 3.5 kg/m^3 (reported by the manufacturer). In Korea, CCA-treated wood used for above ground purposes requires at least 3.5 kg of CCA/m^3 of wood. Untreated dimensional lumbers (yellow pine) from several wood-based housing construction sites in Daejeon, Korea, were also collected to mix with CCA-treated wood samples as well as to serve as a control. The following CCA-treated wood samples were prepared for laboratory leaching tests: CCA-treated wood block, chip and sawdust, and CCA-wood ash. The CCA-treated wood block was prepared by cutting the dimensional CCA-treated lumber to dimensions of $9 \times 6 \times 4 \text{ cm}$ (approx. 100 g). In order to produce CCA-treated wood chip, approximately 1.0 m^3 of the lumber was processed using an industrial wood chip shredder. Following size reduction, the chipped samples were sieved (less than 1-cm diameter) to obtain a homogenous sample. Sawdust

samples were prepared by cutting the lumbers in multiple locations using an electric power drill. An effort was made to cut the entire wood piece to obtain a sample representative of the entire piece. Untreated wood samples were processed as the same sample preparation of CCA block, chip, and sawdust above. All the samples were stored in sealed polyethylene bags in the absence of light prior to initiation of leaching tests.

In order to prepare CCA-wood ash, the chipped wood samples (approx. 240 kg) produced from the shredder were combusted using an industrial furnace (Batch Type, Model KB-HBS-1500). The dimensions of the furnace were approximately 4.2 x 2.4 x 1.8 m. Catch pans were placed at the bottom of the furnace to capture the ash. Prior to the combustion process, the industrial furnace was cleaned to remove any foreign materials, if present. The samples were combusted for approximately two hours at a temperature that varied between 550°C at the beginning of the burn to 650°C toward the end of the burn. Following the combustion, the ash sample (approx. 3.2kg) was collected from the catch pan and used for total and leaching analyses.

Total metal concentrations of chromium, copper, and arsenic were measured for the sawdust and the ash using microwave acid digestion (EPA Method SW-846 3015). The acid digestates were analyzed by using inductively coupled plasma-atomic emission spectroscopy (ICP-AES, Model Optima 3000DV, EPA Method SW-846 6010B). Samples with high concentrations of metal were diluted to fit within the linear region of the calibration curve. Analysis for total hexavalent chromium (Cr(VI)) in the solid samples was performed and required an alkaline digestion to preserve the chromium species (EPA Method SW-846 3060A). The alkaline digestates were analyzed for Cr(VI) using the 1,5-diphenylcarbazide colorimetric method (EPA Method SW-846 7196). The colorimetric analysis involved adding 1,5-diphenylcarbazide to a 25 ml digested sample. The intensity of the resultant samples containing Cr(VI) was measured using a spectrophotometer (HACH Model DR/4000U) at 540 nm. The detection limit of Cr(VI) was 0.003 mg/l.

A laboratory leaching experiment was conducted on the CCA-treated wood and CCA-wood ash samples over a period of 60 days to determine the kinetic leaching behavior and flux changes of toxic metals from the samples. The flux was calculated by the differences of concentration in leaching period and mass of sample (or surface area for blocks). The experiment used for this study is similar to TCLP and SPLP protocols with an exception of particle size and leaching solution. A 100-gram block sample was placed in a 2.2-liter polyethylene container. In case of CCA-treated wood chip and sawdust, a 50-gram CCA sample was mixed with a 50-gram untreated wood sample. Two liters of de-ionized water as a leaching solution were then added to the container (i.e., liquid-to-solid ratio of 20:1). A leaching test for the ash sample was conducted using 10 g of ash sample with 200 ml of the solution. De-ionized water was used to simulate the leaching of wood preservative from land-applied CCA-treated wood and CCA-wood ash under rainfall. One untreated block sample was similarly prepared and leached as a control. The container was placed in a rotary extraction vessel (30±2 rpm) and rotated end-over-end at room

temperature ($20\pm 2^{\circ}\text{C}$). Duplicate samples were run for all samples except for the block samples. Three replicate blocks were leached with liquid-to-solid ratio of 20:1. Leachate samples were collected at the following intervals: 6 hr., 12 hr., 18 hr., 36 hr., 3 days, 7 days, 10 days, 20 days, 30 days, 40 days, and 60 days. Leachate was completely removed after the designated intervals and on each sampling occasion the leaching solution was replaced with a completely fresh volume of leaching solution (i.e., de-ionized). After collecting leachate, the final pH of the leachate was recorded using a pH meter (Accumet Model HM-7E), and the leachate sample was filtered through a $0.7\text{-}\mu\text{m}$ borosilicate glass fiber filter and acidified to less than pH 2 and stored in an acid-rinsed polyethylene bottle at 4°C prior to metal analysis. A portion of the filtered leachate was digested following liquid digestion for metals (EPA Method SW-846 3010A). The acid digestate was analyzed for chromium, copper, and arsenic using the ICP-AES or inductively coupled plasma-mass spectrophotometry (ICP-MS, Model Elan 6000, Method SW-846 6010B), depending on metal concentrations. Hexavalent chromium concentrations in the leachate were also measured using the colorimetric method. The Cr(VI) samples were analyzed within 24 hours of collection.

All glassware was washed with 1-N HNO_3 and thoroughly rinsed with de-ionized water prior to use. Laboratory blanks, duplicates, sample spikes, and calibration check samples were included during chemical analysis for quality assurance and quality control (QA/QC). Leaching from a control sample of an untreated wood resulted in concentrations below detection limits ($10\text{ }\mu\text{g/L}$). Metals were not detected above the detection limit for any of the blanks. The sample spike recovery percentage of arsenic, chromium, and copper ranged from 84% to 97%. All reagents used were analytical grade.

RESULTS AND DISCUSSION

Total metal concentrations (mg/kg) in the CCA-treated wood and CCA-wood ash are shown in Table 1. The metal concentrations from the sawdust samples ranged from 840 to $1,200\text{ mg/kg}$. These levels correspond to the entire cross section of the wood samples; they differ from theoretical levels based on the manufacturer-reported retention level (3.5 kg/m^3) that is based upon measurement of the outer 1.5 cm of the wood surface. It should be noted that retention levels could be quite variable even in the same piece of lumber (Hingston et al., 2001). The total concentrations were used to estimate the percentage of metals leached during the long-term leaching test. The percent leaching was calculated by leaching results divided by total metal concentrations. For the wood ash samples, the concentrations of the metals ranged from 19,000 to $22,000\text{ mg/kg}$ (Table 1). The levels in the ash are much higher than the non-combusted CCA samples, indicating that much of the metals are concentrated in the ash residue after the combustion. The sum of all the metals accounts for 6.2% of the ash by weight. Of the total chromium in the ash sample, approximately 30% was in the form of Cr(VI), while the non-combusted CCA-treated wood contained only 2.5% of Cr(VI) in total chromium. This indicates that much of chromium present in the CCA-treated wood was

Table 1. Total content (mg/kg) of CCA-treated wood and CCA-wood ash.

| Parameters | CCA-Treated Wood | CCA-Wood Ash |
|---------------------|-------------------------|----------------|
| Arsenic | 1,200 ± 50 ^a | 21,300 ± 4,480 |
| Copper | 840 ± 35 | 19,000 ± 420 |
| Chromium | 1,100 ± 20 | 22,000 ± 3,330 |
| Hexavalent Chromium | 27 ± 3.8 | 7,100 ± 220 |

a. Mean and standard deviation based on triplicate samples.

oxidized to Cr(VI) during the combustion process. Figure 1 presents the results of cumulative metals leached from CCA-treated wood and CCA-wood ash as a function of time. In case of the wood samples (Figure 1. (a), (b), and (c)), arsenic leached much greater amount than chromium and copper. A recent study also showed a similar result, indicating that arsenic leached the most over the 14-day experimental period, and the amounts of chromium and copper leached from CCA-treated wood were relatively small over the same period (Townsend et al., 2004). Metals from the CCA-wood chip sample showed a very similar leaching trend to the sawdust sample, but with slightly lower concentrations. The block samples leached less metals than the chip and sawdust samples because the smaller surface area in the block samples available may have limited leaching of the metals. Arsenic from the block samples appears to continually leach over the time, suggesting that a considerable amount of arsenic could be mobilized in a relatively longer period of time and made available to the surrounding environment. Following 60 days of leaching, cumulative amounts of arsenic leached from the CCA-wood block, chip and sawdust samples were approximately 160, 500, and 610 mg/kg, respectively. These correspond to 6.5%, 41%, and 50 % leaching of total arsenic. While no detectable leaching of Cr(VI) occurred from the CCA-wood block, 60-day cumulative amounts of Cr(VI) leached from the CCA-wood chip and sawdust were 5.8 and 10.1 mg/kg, respectively. The final pH values for CCA-wood block samples slightly decreased over time (6.52 to 5.72), while the final pH for both CCA-wood chip and sawdust ranged from 6.08 to 4.98.

For the CCA-wood ash (Figure 1 (d)), chromium leaching was noted to be the highest of all metals. The percentages of chromium leached from CCA-wood ash averaged approx. 31% of total chromium after 60-day leaching, while only 7.5% of arsenic present in the ash leached into the solution. Most of chromium leached from the ash was in the form of Cr(VI). This can be attributed to substantial amounts of Cr(VI) present in the ash that is mobile and readily leached into the solution. Copper leached into the solution below or near the detection limit (0.003 mg/L). This suggests that most copper was bound to the ash and would not be expected to readily leach into solution. There are limited data pertaining to the leaching of ash from the combustion of CCA-treated wood (Solo-Gabriele et al., 2002; Khan et al., 2004). A previous leaching study conducted on CCA wood ash using deionized water has observed similar trends, with arsenic and chromium leaching the most and copper leaching the least (less than the detection limit of 0.1 mg/l) (Solo-Gabriele et al., 2002).

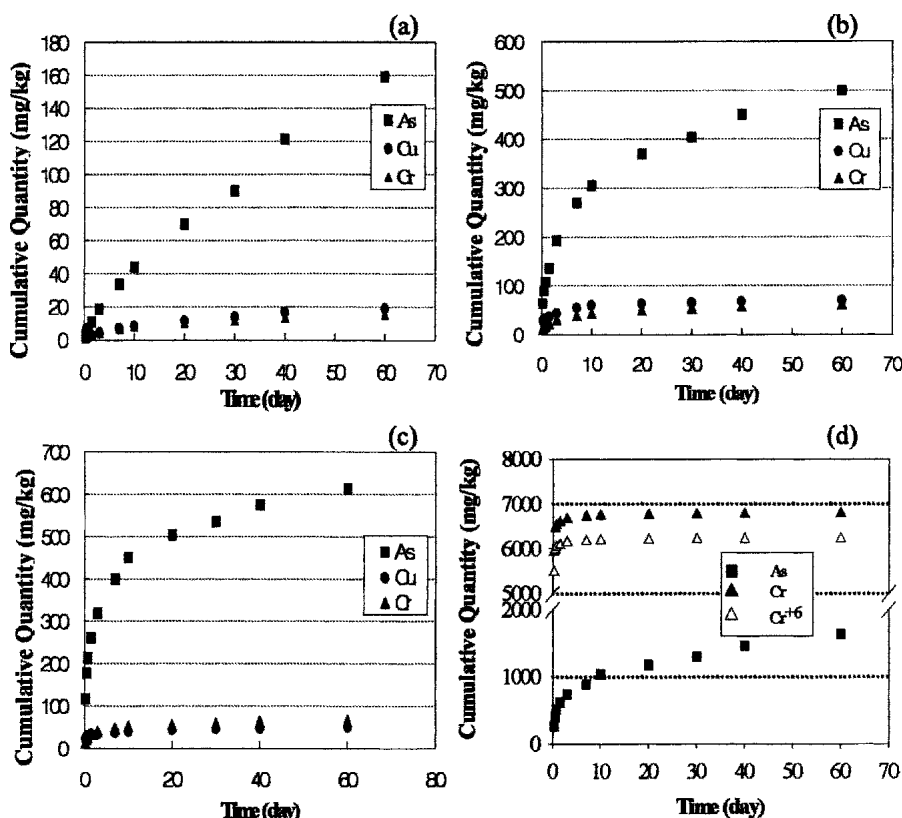


Figure 1. Cumulative metals leached from CCA-treated wood and CCA-wood ash as a function of time: (a) CCA-wood block, (b) CCA-wood chip, (c) CCA-wood sawdust, (d) CCA-wood ash.

Figure 2 illustrates the flux changes of chromium, copper, and arsenic from the CCA-treated wood and CCA-wood ash as a function of time. For CCA-treated wood samples (Figure 2(a), (b), and (c)), leaching fluxes of arsenic are noted to be highest of all metals. The copper release rate in each interval was comparable to the chromium rate. All of these data show that the fluxes for each element are high initially and then decrease with time. The rapid reductions in flux may be explained by the relatively high proportion of the total preservative content that is subject to very early loss resulting from readily leachable CCA chemicals on the surface or in the pores of treated wood. In case of the wood block sample in Figure 2(a), leaching fluxes of As, Cr and Cu after 60 days declined to 15%, 2.0% and 2.1% of 6hr, respectively (see Table 2). Much of leaching by dissolution may have occurred only on the exposed surface area of the block sample. Thus, the leaching fluxes of all metals from the CCA block samples were much lower than those of the CCA-wood chip and sawdust, especially at the beginning of the leaching experiment. It is known that wood is an inherently variable material which makes comparison of different studies and its environmental impact more challenging.

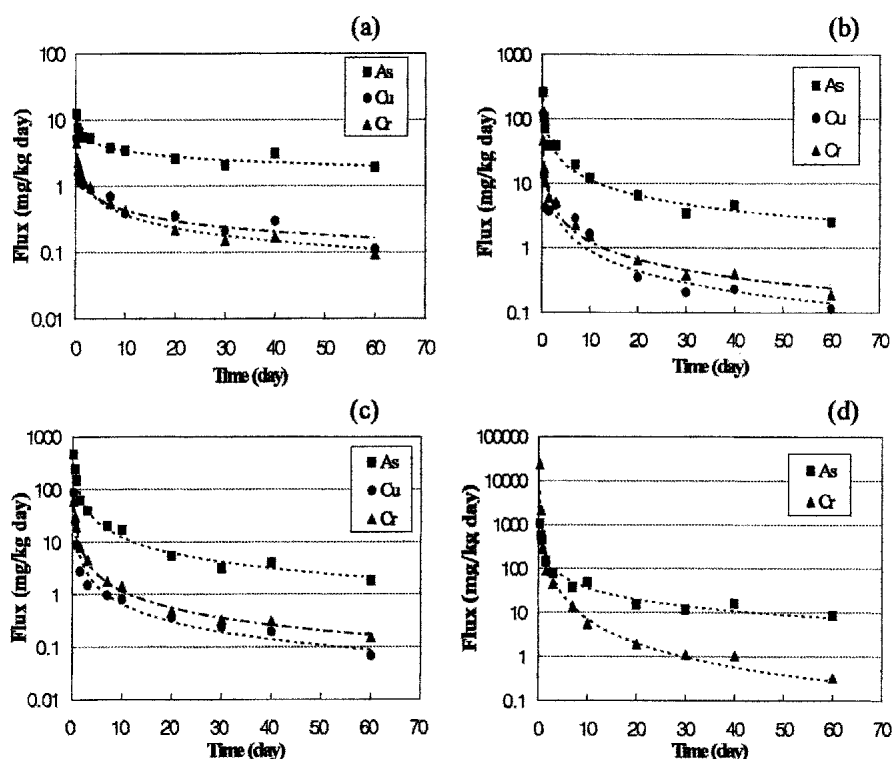


Figure 2. Flux changes of As, Cr, and Cu leached from CCA-treated wood and CCA-wood ash as a function of time: (a) CCA-wood block, (b) CCA-wood chip, (c) CCA-wood sawdust, (d) CCA-wood ash.

Table 2. Leaching Flux Changes of Copper, Chromium, and Arsenic from CCA-Treated Wood and CCA-Wood Ash between 6 hr and 60 day.

| Parameters | Leaching | Block | | Chip | Sawdust | Ash |
|------------|----------|---------|-----------------------|---------|---------|---------|
| | Time | mg/kg·d | μg/cm ² ·d | mg/kg·d | mg/kg·d | mg/kg·d |
| As | 6 hr | 12.3 | 5.54 | 254 | 456 | 1,050 |
| | 60 day | 1.9 | 0.85 | 2.49 | 1.84 | 8.68 |
| Cu | 6 hr | 5.16 | 2.32 | 118 | 86.4 | <0.05 |
| | 60 day | 0.11 | 0.05 | 0.11 | 0.17 | <0.05 |
| Cr | 6 hr | 4.53 | 2.04 | 46.1 | 59.1 | 23,720 |
| | 60 day | 0.09 | 0.04 | 0.19 | 0.15 | 0.33 |
| Cr(VI) | 6 hr | <0.05 | <0.05 | 5.48 | 7.6 | 21,920 |
| | 60 day | <0.05 | <0.05 | <0.05 | <0.05 | 0.25 |

Several factors such as the treatment process, retention levels of treated wood, type of treated wood, and exposure environment may contribute to the variability in the CCA-treated wood leaching studies (Lebow et al., 2004). A kinetic study conducted on CCA-treated wood block using deionized water has observed similar trends over the period of leaching experiment of 21 days (Dubey et al., 2005). Although direct comparison of the flux obtained from this study may be difficult, due to the varying effects of the factors above, the fluxes of CCA-wood block (Table 2) fell within the ranges of other studies reported (Breslin and Adler-Ivanbrook, 1998; Hingston et al., 2001).

For the wood ash sample, the leaching rate of chromium is relatively higher than the arsenic leaching rate at the beginning of the experiment (within 12 hr), and then the rapid reduction was observed over time. The rate at which chromium leaching decreased was more than the arsenic rate. Calculated arsenic flux at day 60 (approx. 10 mg/kg·day) was more than one order of magnitude greater than the calculated chromium flux. A slower rate of arsenic release from CCA wood ash may be particularly important in assessing the long-term loss of arsenic from the ash when disposed. Recent research has also found that management in traditional combustion systems poses a concern with respect to ash quality and leaching due to the presence of CCA-treated wood (Solo-Gabriele et al., 2002). The final pH for CCA-wood ash samples ranged from 10.6 to 11.1. The relatively rapid reduction in chromium (mainly consisted of Cr(VI)) leaching compared to that of arsenic may be attributed to the relatively higher mobility of Cr(VI).

Results of this study showed that the leaching fluxes of arsenic from CCA-treated wood were consistently much higher than those of chromium and copper throughout the 60-day leaching period. Increased leaching time significantly reduced leaching fluxes of all CCA elements. The rate of preservative leaching from CCA-treated wood has been observed to decrease with time (Dubey et al., 2005). It was reported that higher concentrations of preservative leaching occur in the beginning just installation of treated wood structure. Thus, several studies have showed the declining flux rate using empirical methods (van Eetvelde et al., 1995; Brooks, 2001; Hingston et al., 2001). The particle size of CCA-treated wood used also affected the leaching rates of the elements. Relatively large surface area per unit weight of CCA-treated wood allowed more wood available for leaching. In case of CCA-wood ash, the rate of chromium release was much greater than that of arsenic for the first 12-hr leaching. However, the steady-state release rate of arsenic from the ash was greater than that of chromium. Much of the chromium was present in the hexavalent form in the leachate, which is more toxic and mobile than trivalent chromium. According to the leaching levels of chromium, copper, and arsenic from CCA-treated wood by means of kinetic leaching experiments, arsenic may become a potential problem for soil and groundwater contamination via leaching due to its mobility and toxicity, while chromium in CCA-wood ash, especially Cr(VI), may become a major of concern for CCA-wood ash disposal after incineration. However, further study should be carried out to provide a more thorough assessment of risk along with site specific data.

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