Fractionation of heavy metals in liquefied chromated copper arsenate (CCA)-treated wood sludge using a modified BCR-sequential extraction procedure

Hui Pan [a,*], Chung-Yun Hse [b], Robert Gambrell [c], Todd F. Shupe [d]

[a] Calhoun Research Station, Louisiana State University Agricultural Center, 321 Highway 80E, Calhoun, LA 71225, United States
[b] Southern Research Station, USDA Forest Service, 2500 Shreveport Highway, Pineville, LA 71360, United States
[c] Dept. Oceanography and Coastal Sciences, Louisiana State University, Baton Rouge, LA 70803, United States
[d] School of Renewable Natural Resources, Louisiana State University Agricultural Center, Baton Rouge, LA 70803, United States

ABSTRACT

Chromated copper arsenate (CCA)-treated wood was liquefied with polyethylene glycol/glycerin and sulfuric acid. After liquefaction, most CCA metals (98% As, 92% Cr, and 83% Cu) were removed from liquefied CCA-treated wood by precipitation with calcium hydroxide. The original CCA-treated wood and liquefied CCA-treated wood sludge were fractionated by a modified Community Bureau of Reference (BCR) sequential extraction procedure. The purpose of the BCR-sequential extraction used in this study was to examine the availability of CCA metals in treated wood for reuse. Both As and Cr had a slightly higher concentration in the sludge sample than in original CCA-treated wood. The sequential extraction showed that As and Cr were principally existed in an oxidizable fraction (As, 67%; Cr, 88%) in original CCA-treated wood. Only 1% of both As and Cr were extracted by hot nitric acid with the last extraction step. The distribution of As and Cr changed markedly in liquefied CCA-treated wood sludge.

The amount of As in the exchangeable/acid extractable fraction increased from 16% to 85% while the amount of Cr increased from 3% to 54%. Only about 3% of As was present in the oxidizable fraction. However, there was still about 34% of Cr in the same fraction. Based on these results from sequential extraction procedures, it can be concluded that the accessibilities of CCA metals increase markedly by the liquefaction–precipitation process.

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1. Introduction

For many years, chromated copper arsenate (CCA) was the most commonly used waterborne wood preservative in the world. Despite the decision of the US and Canadian wood preservation industry to voluntarily withdraw the use of CCA-treated wood from residential and consumer uses as of December 31, 2003 (USEPA, 2003), the use of CCA-treated wood for industrial purposes (e.g., utility poles and marine pilings) continues (Song et al., 2006; Block et al., 2007). In addition, with an expected average service life between 20–40 years, the amount of spent CCA-treated wood in the US and Canada will greatly expand from the current 3–4 to around 12 Mm^3 y^{-1} within the next 15 years (Kazi and Cooper, 2006). Many efforts have been made to develop appropriate alternative technologies for disposal of spent CCA-treated wood other than traditional landfills and incineration, which have raised environmental and human health concerns. A method to remove Cr, Cu, and As from spent CCA-treated wood based on wood liquefaction has been developed by Lin and Hse (2005). They reported that CCA-treated wood can be liquefied under similar liquefaction conditions as non-treated wood and up to 99% of the heavy metals can be removed from CCA-treated wood by adding ferrous salts during the liquefaction stage. In addition, CCA-wood complexes are either released from the decomposed lignin and cellulose or still remain as the complexes or chelates with decomposed lignin and cellulose during the liquefaction reaction, which should provide better accessibility for the extractants in the heavy metal removal step.

Although many CCA metal removal methods, such as chemical extraction (Kartal and Clausen, 2001a; Kakitani et al., 2006) and bioremediation (Clausen, 2004b), have been proposed, very few studies have focused on both removal and reuse of CCA metals. It has been generally considered that Cr(VI) is reduced to Cr(III) and which then precipitates with As(V) and Cu(II) in wood components during CCA fixation process. Therefore, it is essential that the recovered CCA metals should be in an appropriate oxidation state which can be reused as a treating solution. Kazi and Cooper (2006) proposed a combination of extraction and oxidation treatment for CCA-treated wood waste using an oxidizing agent to oxidize Cr(III) to Cr(VI) after the extraction process. However, the chemistry of CCA fixation is largely a matter of conjecture because of the difficulty of *in situ* analysis (Bull et al., 2000). Removed CCA

* Corresponding author. Tel.: +1 318 6442662; fax: +1 318 6447244.
E-mail address: hpan@agcenter.lsu.edu (H. Pan).

metals, either in extracted solution or other residues, are mixtures of three metals and are associated with other organic or inorganic substances. Knowledge of the total amount of removed CCA metals without considering their speciation is not sufficient for separation or recovery of the metals for reuse.

Chemical speciation can be divided into three classes: (1) classical speciation refers to specific chemical compounds or oxidation states of elements; (2) functional speciation refers to the observed role or behavior of the element; and (3) operational speciation refers to the situation where the reagent used to extract the sample defines the species (Ure, 1991). Sequential extraction is a typical operational speciation method. The sequential extraction procedure was first developed for trace metal analysis in sediment (Tessier et al., 1979). It has been adapted and applied to determine metal form or speciation for a variety of materials, such as soil and sewage sludge (Silveira et al., 2006; Pöykiö et al., 2006). To standardize the methodology used in a wide range of procedures, the Community Bureau of Reference (BCR) began an evaluation program of the sequential extraction schemes for determining metals in soils and sediments (Ure, 1991). Despite some limitations, such as lack of phase selectivity and absence of validation, sequential extraction is recognized as the best available method for gaining information on the origin, manner of occurrence, bioavailability, mobilization, and transport of heavy metals (Ma and Rao, 1997).

In the study presented in this paper, spent CCA-treated wood was liquefied with polyethylene glycol (PEG) using sulfuric acid as a catalyst. CCA metals were then precipitated in the liquefied wood sludge with calcium hydroxide. Considering the similarities and differences between liquefied CCA-wood sludge and sediments or soils, a modified BCR-sequential extraction procedure was applied to fractionate Cr, Cu, and As in liquefied CCA-treated wood sludge. The purpose of this study was to investigate the possible forms and availabilities of the CCA metals after being removed from CCA-treated wood waste and thus to provide information for CCA metal recovery and reuse.

2. Materials and methods

2.1. Materials

CCA-treated wood waste was obtained from a local preservative treated wood product company and was reduced to sawdust. Then the sawdust was oven-dried and sieved to pass a 20-mesh (0.853 mm) before liquefaction. PEG with a molecular weight of 400 (PEG 400) and glycerin were used as the liquefaction reagent solvent at a weight ratio of 2:1. Sulfuric acid (95%) was used as a catalyst in the liquefaction reaction. All chemicals were reagent grade. Chemicals used in sequential extraction procedures were trace metal grade.

2.2. Sample preparation for sequential extraction

Three types of samples were prepared for sequential extraction as follows: (1) Liquefied CCA-treated wood sludge: the wood sludge samples were obtained by liquefaction of CCA-treated wood and precipitation of CCA metals. In brief, CCA-treated wood sawdust was mixed with PEG 400/glycerin at a weight ratio of 1/3. Sulfuric acid was added at 3% of the wood weight. The mixture was reacted under reflux at 150 °C for 120 min. After the liquefaction reaction, the mixture was cooled down and diluted 6 times its volume with acetone/distilled water (4/1, v/v) mixture. Then calcium hydroxide (4% of liquefied wood weight) was added to the diluted liquefied wood and stirred for 2 h. The above mixture was then subjected to filtration to separate the liquid and the sludge. The

wet liquefied CCA-treated wood sludge was oven-dried at 85 °C for 24 h and then ground in a Wiley mill to powder for sequential extraction. The preparation procedures were summarized in Fig. 1. (2) Liquefied CCA-treated wood residue: CCA-treated wood was liquefied and diluted as described above and no calcium hydroxide was added. After stirring for 1 h, the mixture was filtrated. Solid wood residue was oven-dried and weighed to determine the wood residue content as divided by the weight of original CCA-treated wood used in the liquefaction. The wood residue content was 24.5% after liquefaction. Then the residue samples were ground for sequential extraction. (3) Original CCA wood particle without liquefaction.

2.3. Sequential extraction

Three types of samples described in the previous section were subjected to the sequential extraction. A modified three-step BCR-sequential extraction procedure (Rauret et al., 2000) was followed. For each extraction specimen, about 0.5 g oven dried sample was weighed to three decimal places into a 50 mL polyethylene centrifuge tube. The extraction procedures are briefly described as follows:

Step 1 (exchangeable/acid extractable): A total of 20 mL acetic acid solution (0.11 M, pH = 3.06) was added to the centrifuge tube and shaken for 16 h at room temperature. The extract was separated from the solid residue by centrifugation at 3000g for 20 min. The supernatant was collected for later analysis. The residue was rinsed with 10 mL distilled water by shaking for 15 min, centrifuging, and decanting the supernatant.

Step 2 (easily reducible): A total of 20 mL freshly prepared hydroxylamine hydrochloride solution (0.5 M, pH = 1.60) was added to the residue in the centrifuge tube from step 1 and shaken for 16 h at room temperature. The separation of the extract, collection of the supernatant, and rinse of residue were the same as in step 1.

Step 3 (oxidizable): First, 5 mL of hydrogen peroxide (30%) was added to the residue from step 2 in the centrifuge tube. The tubes were loosely covered to prevent substantial loss of hydrogen peroxide. The digestion at room temperature was allowed to
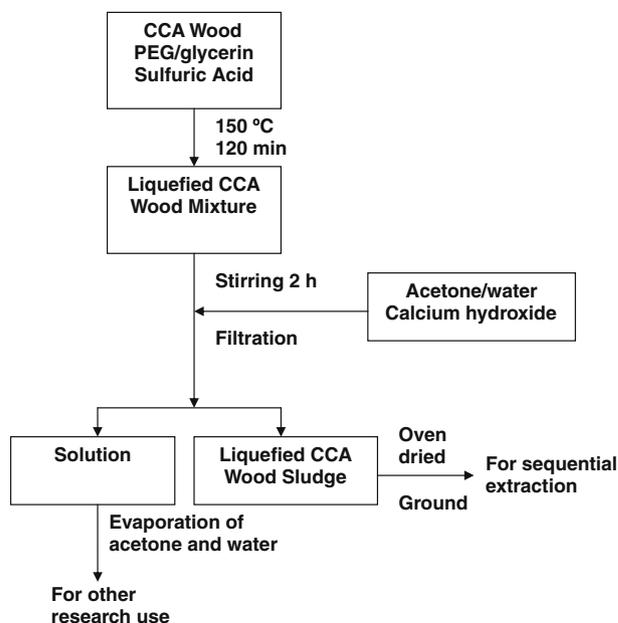
Fig. 1. Flowchart of liquefaction of CCA-treated wood and precipitation for liquefied CCA-treated wood sludge.

proceed for 1 h with occasional manual shaking and followed by digestion at 85 °C for another 1 h. The centrifuge tubes were then uncovered and heating continued to reduce the volume to 1–1.5 mL. Another 5 mL hydrogen peroxide was added to the tube and digested with a cover at 85 °C for 1 h, and again, was the cover was removed and the volume reduced to 1–1.5 mL.

All liquefied CCA-treated wood sludge samples and liquefied CCA-treated wood residue samples turned to a clear solution after the hydrogen peroxide digestion at step 3. The final extract from step 3 was diluted to 50 mL and stored for further analysis. There was a residue that remained after hydrogen peroxide digestion for all original CCA-treated wood samples. This residue was shaken with 25 mL ammonium acetate (1.0 M) for 16 h at room temperature. The separation of extract and rinse of residue were the same as described in step 1.

Step 4 (hot nitric acid extractable): For original CCA-treated wood sample, residue from step 3 was transferred to a glass digestion tube and digested with nitric acid at 120 °C.

2.4. Digestion method for element concentration determination

Because the samples in this study were woody material or from woody material, hot nitric acid, instead of *aqua regia* was used to digest the original CCA-treated wood, liquefied CCA-treated wood sludge, and liquefied CCA-treated wood residue samples. The total metal concentrations in each sample were determined by digestion of about 0.5 g sample with 10 mL nitric acid according to AWPA A7-08 (2008). Liquid samples from sequential extraction steps 1 and 2 were also digested according to the same method but with less nitric acid to eliminate the matrix interferences from extraction reagents. After digestion, the concentrations of As, Cr, and Cu in the samples were determined by the inductive coupled plasma atomic emission spectroscopy (ICP-AES). Once the heavy metal concentrations in each sample were determined, the percentage removal of the CCA metals from CCA-treated wood and the percentage of the CCA metal left in the liquefied CCA-treated wood residue was calculated by following equation:

$$Percentage \; removal(\%) = \frac{C_p W_p}{C_o W_o} \qquad (1)$$

where C_p is the metal concentration in oven-dried liquefied CCA-treated wood sludge or liquefied CCA-treated wood residue and C_o is the metal concentrations in original CCA-treated wood, W_p is the weight of oven-dried liquefied CCA-treated wood sludge or liquefied CCA-treated wood residue and W_o is the weight of the original CCA-treated wood used in the liquefaction.

3. Results and discussion

3.1. Percentage removal of CCA metals from CCA-treated wood

All three metals can be removed from original CCA-treated wood at high percentages by the liquefaction–precipitation method. The percentage removal of As, Cr, and Cu calculated from Eq. (1) was 98%, 92%, and 83%, respectively. Only about 2% of As, 6% of Cr, and 7% of Cu were found in the liquefied CCA-treated wood residue, which was much lower than their original concentrations in CCA-treated wood.

It is worthy noting that the percentages of all three metals left in liquefied CCA-treated wood residue were even much lower than the average percentage of wood residue content itself (24.5%). This result indicated that during the liquefaction reaction, the majority of the metals were either released from the complexes with wood components or remain as complexes with decomposed wood components that dissolved in acetone/water solution. Lin and Hse

(2005) had the same conclusion in their study on liquefaction of CCA-treated wood. However, this study obtained a different percentage removal of the heavy metals with a similar liquefaction and precipitation method used in their study. All three metals in this study showed a much higher percentage removal without adding a ferrous salt during the liquefaction stage as Lin and Hse did. A stainless steel (T316) Parr reactor was used in Lin and Hse's study and the liquefaction was performed in a sealed system, while a glass reactor was used in this study and the liquefaction was performed under refluxing. We have previously reported that liquefaction reactions could undergo different reaction mechanisms in these two types of reactors (Pan et al., 2007). This could be the reason for the differences in percentage removal of the heavy metals in these two reactor systems. Despite the different CCA percentage removal, calcium hydroxide was confirmed in this study to be an effective precipitant for CCA metals. Other studies also showed that the most common method applied for chromate control is to reduce Cr(VI) to Cr(III) and then shift the pH to alkaline values by lime addition. Cr(III) will form insoluble Cr(OH)$_3$ and can be removed by precipitation (Zouboulis et al., 1995; Guo et al., 2006). Lime precipitation is also frequently used to remove arsenic in water by forming calcium arsenate with low solubility (Johnston and Heijnen, 2001; Ghimire et al., 2003).

3.2. Total metal concentration and CCA metal fractionation

Table 1 lists the total heavy metal concentrations determined by the AWPA method and their concentrations in each fraction of the BCR-sequential extraction procedures. The recoveries of the three metals were mostly above 95% in original CCA-treated wood and liquefied CCA-treated wood sludge samples. With much lower metal concentrations, the liquefied wood residue sample also showed lower metal recovery from 77% of As to 88% of Cu. Comparing the concentration of each element in original CCA-treated wood and liquefied CCA-treated wood sludge, both As and Cr had higher concentrations in the sludge sample than in original wood sample. These results implied that wood liquefaction as a recycling option for CCA-treated wood waste has the advantage of concentrating heavy metals into liquefied wood sludge and thus helped to reduce the processing volume of the recycling materials. Fig. 2 shows the CCA metal distributions in original CCA-treated wood and liquefied CCA-treated wood sludge.

Arsenic was principally present in the oxidizable fraction in original CCA-treated wood sample, accounting for around 67% of the sum of all sequential steps. Similar amounts (16%) of As existed in exchangeable/acid extractable and reducible fractions. Only 1% was extracted by hot nitric acid at the last step of the sequential extraction. The distribution of As in original CCA-treated wood is reasonable. According to current knowledge on the fixation mechanism of CCA metals in treated wood, arsenic was precipitated in wood by Cr as CrAsO$_4$ (Bull et al., 2000). Most As should be fixed in wood after a complete fixation process and associated with organic matter; in other words, extracted as oxidizable fraction. A small amount of As was found in both exchangeable/acid extractable and reducible fractions, which very likely came from some fixed arsenic gradually released from wood due to the changes of chemical environment during the service time of treated wood products. It has been reported that the changes in pH of the leachant, temperature or type of leaching agent greatly affect the leaching of Cr, Cu, and As at disposed CCA-treated wood areas (Moghaddam and Mulligan, 2008; Lebow et al., 2008). These two fractions could also come from the As that failed to fix onto wood during the fixation process. Arsenic present in the hot nitric acid extracted fraction (step 4) is very likely associated with wood cellulose in crystalline region. Wood is known as a non-uniform material. It consists of amorphous lignin, hemicelluloses, cellulose

Table 1
CCA concentration in each fraction of the liquefied CCA-treated wood sludge by BCR scheme.

	Step 1[a]	Step 2[b]	Step 3[c]	Step 4[d]	Sum	Total[e]	% Recovery[f]
Original CCA wood particle							
As	559 ± 91	578 ± 54	2340 ± 209	26 ± 10	3503	3726 ± 125	94
Cr	123 ± 24	338 ± 27	3733 ± 390	51 ± 16	4245	4101 ± 123	104
Cu	853 ± 145	1219 ± 99	406 ± 53	6 ± 1	2484	2573 ± 63	97
Liquefied CCA-treated wood sludge[g]							
As	3482 ± 180	407 ± 37	215 ± 12	–	4104	4262 ± 201	96
Cr	2365 ± 133	504 ± 34	1509 ± 42	–	4378	4349 ± 166	101
Cu	1167 ± 63	1157 ± 40	104 ± 4	–	2428	2425 ± 113	100
Liquefied CCA-treated wood residue[h]							
As	156 ± 29	21 ± 2	78 ± 5	–	255	330 ± 27	77
Cr	100 ± 9	14 ± 1	702 ± 58	–	816	955 ± 79	85
Cu	476 ± 258	143 ± 75	20 ± 5	–	639	724 ± 361	88

Note: The concentration is in ppm and the results are expressed as mean ± standard deviation (six replicates for each sample).
[a] Exchangeable/acid extractable.
[b] Reducible.
[c] Oxidizable.
[d] Hot nitric acid extractable.
[e] Obtained from hot nitric acid digestion.
[f] Ratio of the Sum over Total.
[g] Obtained from precipitation with $Ca(OH)_2$ after liquefaction.
[h] Obtained directly after liquefaction.

and crystalline cellulose and thus to be expected to has different accessibilities to different regions. Metals associated with crystalline cellulose represent the least amount of total fixed metals and also the most difficult portion to access or extract. Therefore,
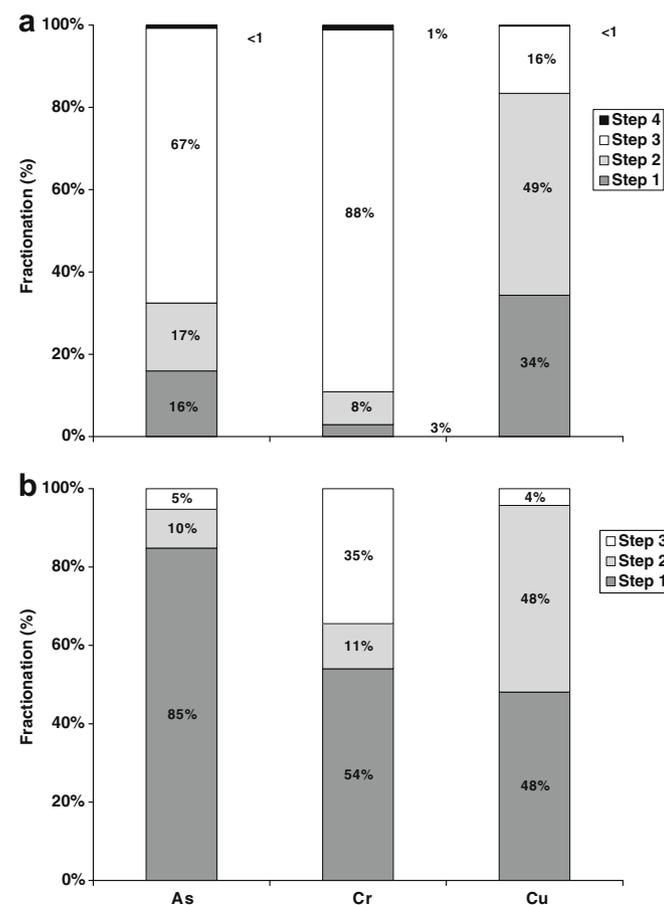

Fig. 2. Distribution of three elements in (a) original CCA-treated wood particles and (b) liquefied CCA-treated wood sludge among each fraction from the sequential extraction.

harsher conditions are required to liberate this fraction of metals than those associated with wood components in amorphous regions.

As shown in Fig. 2b, the distribution of As changed dramatically after liquefaction. First of all, about 85% of As in liquefied CCA-treated wood sludge was present in exchangeable/acid extractable fraction, which indicated that most As compounds were liberated from their association with wood components during the liquefaction reaction. Wood liquefaction is a thermo-chemical decomposition process of wood material. The reaction is normally carried under acidic conditions due to both the addition of acid catalyst in some cases and the formation of levulinic acid or formic acid from decomposed wood cellulose during the liquefaction reaction (Yamada et al., 2007). Arsenic is very susceptible to acid extraction as reported by other researchers (Kartal and Clausen, 2001a; Clausen, 2004a). In addition, fixed CCA compounds may undergo reversed reaction of the fixation process under strong acidic condition and further convert insoluble CCA compounds into more soluble compound (Kartal and Clausen, 2001a). Therefore, most As was extracted during acetic acid extraction at step 1. The remaining 10% and 5% of As were in reducible and oxidizable fractions, respectively, which indicated that there was small amount of As still bound to some metal oxides and decomposed wood components.

Up to 88% of Cr presented in the oxidizable fraction in the original CCA-treated wood sample. In the CCA treating solution, Cr acts as a fixing agent to precipitate As and Cu onto wood when Cr(VI) reduced to Cr(III). This function of Cr in the CCA fixation process implies that Cr easily forms a complex with wood and thus most Cr is associated with organic matter. Chromium in the reducible fraction (8%) can be presented as simple chromium hydroxide (Audry et al., 2006). For a type C CCA solution, the ratio of Cr/Cu/As is about 1.6/0.78/1. Bull (2001) suggested that part of the Cr was present with As as $CrAsO_4$ and the remaining Cr must be present as $Cr(OH)_3$.

After the liquefaction–precipitation process, the distribution of Cr changed markedly as showed in Fig. 2b. Similar to As, the major change of Cr distribution is the large increase (51%) in the exchangeable/acid extractable fraction and the decrease (54%) in the oxidizable fraction compared to original CCA-treated wood. There was also a slight increase in the reducible fraction. However, compared to As and Cu, Cr has a larger portion (34%) associated

with organic matter in liquefied CCA-treated wood sludge. Fig. 2a shows that Cr has the highest portion in oxidizable fraction among the three heavy metals in original CCA-treated wood. As discussed in previous sections, exposure of CCA-treated wood to acid conditions may reverse the CCA fixation process. However, metal sorption by natural organic matter is reduced less at lower pH than metal sorption on oxides (Basta et al., 2005). In other words, as shown in the study, metals associated with organic matter (e.g., oxidizable fraction) are more resistant to acid extraction than those associated with metal oxides (e.g., reducible fraction). Other researchers also reported that Cr leached the least despite being present in the greatest proportion (Moghaddam and Mulligan, 2008).

Unlike As and Cr, Cu was largely present in reducible fraction (49%) and exchangeable/acid extractable fraction (34%) in original CCA-treated wood (Fig. 2a). This could contribute to the different "fixing" forms of the three heavy metals in CCA-treated wood. Previous reports about the chemistry of CCA fixation in wood speculated that As, Cr, and Cu are fixed as $CrAsO_4$–lignin complexes, $CuCrO_4$–lignin complexes, and Cu(II)–lignin/cellulose complexes in CCA-treated wood (Pizzi et al., 1984; Pizzi, 1990). A more extensive investigation done by X-ray absorption fine structure spectroscopy showed that Cu is not closely associated with any other heavy metals (Bull et al., 2000). It has been reported that during chemical extraction processes to remove CCA metals from CCA-treated wood waste, only 23% of Cu could be removed with oxalic acid solution while 74% and 65% of As and Cr could be removed under the same conditions (Kartal and Clausen, 2001b). On the other hand, about 93% of Cu can be removed by ethylenediaminetetraacetic acid while only 36% of Cr and 38% of As can be removed using the same chelating agent (Kartal, 2003). Cu consistently performed differently than Cr and As under the same extraction conditions, which can further implied that Cu was not associated with either Cr or As. Only some chelating agents that have a strong affinity with Cu(II) can liberate it from its complexing with lignin/cellulose. Copper hydroxide $(Cu(OH)_2)$ is the only plausible inorganic fixation product in CCA-treated wood (Bull et al., 2000). The reported forms of heavy metals in treated wood are somewhat variable and more work needs to be done to fully elucidate this point. Copper could be extracted as the hydroxide compound in both exchangeable/acid extractable and reducible fractions, but it could be fixed via different association mechanism with wood. Although it was reported that Cu was mainly present in the oxidizable fraction in many soil samples (Audry et al., 2006; Cuong and Obbard, 2006) and is preferentially retained on organic matter by complexation rather than ion exchange (Balasoiu et al., 2001), only about 17% of copper existed in oxidizable fraction in the original CCA-treated wood sample in this research, which is substantially lower than those of arsenic and chromium.

The distribution of Cu in liquefied CCA-treated wood sludge compared to the wood did not change markedly as arsenic and chromium did. The amount of Cu in the reducible fraction was close to that in the original CCA-treated wood. There was a moderate increase (14%) in the exchangeable/acid extractable fraction and a decrease (13%) in the oxidizable fraction, which indicated that most copper complexed with the wood component had been liberated after liquefaction reaction.

4. Conclusions

CCA-treated wood waste was liquefied and the heavy metals were precipitated by calcium hydroxide after liquefaction. Most CCA metals (98% As, 92% Cr, and 83% Cu) were removed from CCA-treated wood by liquefaction and precipitation. Only about 2% of As, 6% of Cr, and 7% of Cu were found in the liquefied CCA-treated wood residue, which indicated that during the liquefaction reaction, the majority of the metals were either released from the complexes with wood components or remain as complexes with decomposed wood components that dissolved in acetone/water solution. The results from the sequential extraction showed that the CCA metals were mostly bound to organic matters (i.e., wood component) in original CCA-treated wood, and they were present as exchangeable/acid extractable forms in the liquefied CCA-treated wood sludge after liquefaction–precipitation. Based on these results from sequential extraction procedures, it can be concluded that the accessibilities of CCA metals increase markedly by liquefaction–precipitation process.

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