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Pilot-scale investigation of the robustness and efficiency of a copper-based treated wood wastes recycling process

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HIGHLIGHTS

- A leaching process was studied for metals removal from CCA-treated wood wastes.
- This decontamination process was studied at pilot scale (130-L reactor).
- Removals up to 98% of As, 88% of Cr, and 96% of Cu were obtained from wood wastes.
- The produced leachates can be treated by chemical precipitation.

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ABSTRACT

The disposal of metal-bearing treated wood wastes is becoming an environmental challenge. An efficient recycling process based on sulfuric acid leaching has been developed to remove metals from copper-based treated wood chips ($0 < x < 12$ mm). The present study explored the performance and the robustness of this technology in removing metals from copper-based treated wood wastes at a pilot plant scale (130-L reactor tank). After 3× 2 h leaching steps followed by 3× 7 min rinsing steps, up to 97.5% of As, 87.9% of Cr, and 96.1% of Cu were removed from CCA-treated wood wastes with different initial metal loading (>7.3 kg m^{-3}) and more than 94.5% of Cu was removed from ACQ-, CA- and MCQ-treated wood. The treatment of effluents by precipitation–coagulation was highly efficient; allowing removals more than 93% for the As, Cr, and Cu contained in the effluent. The economic analysis included operating costs, indirect costs and revenues related to remediated wood sales. The economic analysis concluded that CCA-treated wood wastes remediation can lead to a benefit of 53.7 US$ t^{-1} or a cost of 35.5 US$ t^{-1} and that ACQ-, CA- and MCQ-treated wood wastes recycling led to benefits ranging from 9.3 to 21.2 US$ t^{-1}.
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1. Introduction

Wood is used extensively worldwide for exterior applications, including utility poles, railway sleepers, timbers, wooden silos, fence posts, decks, picnic tables, etc. [1]. Copper has been successfully used since the 1920s in preservative formulations to protect wood and therefore to extend its service life by 20–50 years [2]. The most common wood preservative used in North America since the 1970s is type-C chromated copper arsenate (CCA-C) formulation [3]. Since January 2004, CCA-treated wood has been voluntarily withdrawn for most residential applications, and it has since been replaced by alternative copper-based treated wood, including alkaline copper quaternary (ACQ-) and copper azole (CA-), both of which have copper present in the form of copper ethanolamine. More recently, the industry has witnessed the introduction of micronized copper quaternary (MCQ-) or micronized copper azole (MCA-), which have copper present as a suspension of copper-carbonate particles.

Although inorganic compounds are very helpful in protecting wood against exterior degradation, the presence of inorganic compounds in preservative formulations can be problematic when

Abbreviations: ACQ, alkaline copper quaternary; CA, copper azole; CCA, chromated copper arsenate; DOC, dissolved organic carbon; ICP-AES, induced coupled plasma-atomic emission spectroscopy; MCA, micronized copper azole; MCQ, micronized copper quaternary; Mm^3, millions of cubic meters; TCLP, toxicity characteristic leaching procedure.

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treated wood reaches its end of life. Large amount of treated wood wastes are expected to be dismantled in the future [4]. In North America, treated wood wastes are most often disposed of in landfills whereas in Europe; treated wood are incinerated in controlled incinerators [5,6]. Landfilling disposal option is of concern because of the potential release of inorganic compounds and the emissions of greenhouse gases [7–9]. Moreover, the potential to dispose of treated wood wastes in landfills in the future is limited, as existing landfill sites are reaching capacity due to the increasing amount of wastes. Increasing fees and stringent regulations related to solid waste landfilling also encourage the creation of new sustainable options that include recycling and value generation [10,11].

The recovery of As, Cr, and Cu from treated wood allows wood waste conversion into value added-products. Over the last decades, metal extraction from treated wood wastes using biological [12–16], physical [16–19], chemical [20–24] or combined methods [25,26] has been widely studied. The thermal remediation processes developed for the management of copper-based treated wood reduce the economic viability of biological or chemical remediation processes in Europe. In North America, biological and chemical remediation processes are encouraged as incineration of CCA-treated wood in controlled conditions is forbidden. Key among the chemical remediation processes developed, are the use of oxidizing agents, chelating agents, organic or inorganic acids [5,22,24]. An efficient leaching process using sulfuric acid to remove metals from treated wood has been identified. This leaching process is highly efficient at solubilizing As, Cr, and Cu from CCA-, ACQ-, CA-, and MCQ-treated samples [22,27]. Further, leaching conditions have been optimized depending on wood preservatives and wood samples in terms of metal removal efficiencies and operating costs at a laboratory scale [28,29].

The effluents produced during the leaching process contain large amount of metals [22,27,28]. Industrial effluents containing metal have been extensively treated by precipitation–coagulation in recent years. The high efficiency, ease of implementation and low cost of this technology explain its wide use for wastewater treatment. Metal can be removed from effluents in the form of metal hydroxides, sulfides, carbonates or phosphates [30]. Previous studies showed that As, Cr and Cu precipitation using ferric chloride and sodium hydroxide allowed more than 98% of the metals to be removed from wood leachates [27,31].

The objective of this study was to assess the performance and the reproducibility of the leaching process for a wide array of CCA-treated wood samples with different initial loading and elapsed time between treatment and remediation and from ACQ-, CA- and MCQ-treated wood wastes at pilot scale. Finally, leachates treatment by precipitation–coagulation was also explored at pilot scale. This work more extensively evaluates the robustness of the process.

2. Materials and methods

2.1. Treated wood wastes preparation and characterization

Different types of CCA-, ACQ-, CA-, and MCQ-treated wood wastes were used in the present study. No damage potentially due to insects or fungi attacks was observed on the different wood samples. CCA-samples came from utility poles, while ACQ-, CA- and MCQ-samples came from 2″ by 6″ treated lumber. Poles were selected from seven loads of discarded poles exposed, in ground contact, to weathering conditions from different time, ranging from 1 to 32 years. The CCA-samples (red and yellow pine) were identified using their year of treatment. The CCA-treated pole sections were cut into thick slices (4.0 cm) using a chain saw. The untreated heartwood was then removed using a saw, except for CCA-1978 and CCA-2005 poles slices, which were chipped directly. The untreated parts were not kept and the treated wood pieces were chipped using a commercial chipper to produce 2–80 mm wood particles. The wood chips were sieved through a 12 mm screen. For ACQ-, CA- and MCQ-samples (southern pine), the treated lumber samples were chipped, and the wood chips were screened through a 12 mm sieve. The availability of metals from wood samples in simulated landfills conditions was estimated using the toxicity characteristic leaching procedure (TCLP) developed for regulatory purpose (EPA method 1311) [32]. According to the American and Canadian regulations, treated wood wastes should be carefully disposed of in lined landfills sites or co-incinerated in order to reduce the dispersion of metals in the Environment. The availability of metals was estimated using only TCLP procedure as the present study focused on the management of treated wood wastes.

2.2. Metals solubilization from treated wood (leaching and rinsing)

A pilot plant scale has been designed to remove metals from CCA-treated wood wastes using a sulfuric acid leaching process [33]. The leaching process involves three leaching steps, followed by three rinsing steps. The leaching and rinsing steps were carried out in a 130-L capacity tank made of 316-stainless steel. A barrier coating (Belzona® 4341, Belzona Company, Quebec, QC, Canada) was applied on the surface of the tank to prevent corrosion due to acid or inorganic attacks. The leaching conditions were optimized (Table 1) based on laboratory scale assays and depended on the wood preservative, initial metal loading and elapsed time between treatment and remediation [28,29]. Depending on the wood sample, the leaching solutions were prepared by diluting between 286 and 1100 mL of sulfuric acid (93%, Chemco Inc., Saint-Augustin, QC, Canada) with 80 L of tap water. The temperature was set at 75 °C using an electric heater plate for CCA-samples and at 20 °C for ACQ-, CA- and MCQ-samples. For CCA-samples, the temperature was fixed at 75 °C to improve the kinetics of chromium removal, which is not necessary for the alternative-treated wood sample (free-chromium samples). Twelve kilograms of wood samples were then added and mixed with the leaching solution at approximately 70 rpm using a mixer and an immersed impeller (23 cm axial diameter). The retention time of the leaching steps was set at 120 min for CCA-samples and 160 min for ACQ-, CA- and MCQ-samples. After the three leaching steps, the wood samples were rinsed three times with 80 L of cold tap water (10 °C). After each leaching and rinsing step, solid-to-liquid separation was carried out by removing most of the liquid using an Elima-Matic Bolted Pump (6.8 $m^3\,h^{-1}$ flow capacity). The wood particles and the remaining water were then transferred into a drainer and allowed to drain in open air.

2.3. Leachates treatment by precipitation–coagulation

The effluents from the leaching steps were treated by precipitation–coagulation in a 136-L capacity settler-tank with a cone-shaped bottom and sludge outlet (PolyAlto tank, Québec, QC, Canada). The addition of a solution of ferric chloride (122 g Fe L^{-1}, Laboratoire Mat, Québec, QC, Canada) enhanced As removal. The amounts of ferric chloride added have been optimized in terms of metal removal efficiencies and operating costs (Table 1). The ferric chloride solution was mixed with the leachate using a mixer and an immersed impeller (12.7 cm axial diameter). A 200 g NaOH L^{-1} solution of sodium hydroxide (97% NaOH, ACS reagent, ACP Chemical, Montréal, QC, Canada) was added under agitation until the pH of the solution reached 7.0–7.2. To adjust the pH of the solution, NaOH was chosen instead of $Ca(OH)_2$ in order to avoid the formation of the instable complex $Ca_3(AsO_4)_2$. Even if $Ca(OH)_2$ is a cheaper raw materials that improve the dewaterability of the sludge, the use of this neutralizing agent in the presence of SO_4^{2-} led to increase

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