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Advances in removal of chromated copper arsenate elements in wood waste, contaminated water and soils

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Chromated Copper Arsenate (CCA) is a water-based mixture of heavy metals widely used as a timber preservative. Despite its efficacy in prolonging the lifespan of treated wood, CCA has become a subject of environmental scrutiny due to the leaching of toxic components into surrounding soil and water. CCA components in soil have been reported with levels as high as 3,300, 2,800 and 2,100 mg/kg for As, Cr and Cu, respectively; way above the recommended levels of 12, 64, 63 mg/kg for agricultural soils. Therefore, the use of CCA as a wood preservative has been restricted in most developed countries. Developing countries, however, continue to utilize CCA treated wood as utility poles. The elements of CCA have potential health risks upon dermal contact with CCA residues from treated structures as well as exposure from contaminated soil and water. There are also concerns about the disposal of CCA treated wood after use, with the current technology of landfilling being unsustainable because of the possibility of CCA leaching into underground water as well as the challenge of limited space for future disposal. Incineration and open burning as a way of disposal produce ash that is highly contaminated and the fumes contribute to air pollution with metals. There is therefore need for sustainable approached for disposal of wood waste. Since the leached elements end up in the environment, several remediation strategies such as chemical methods, bioremediation, phytoremediation and bioadsorption have been reported, as discussed in this review paper, towards sustainable solutions to CCA contamination with some strategies reporting 100% efficiency.

KEYWORDS

copper, chromium, arsenic, CCA, immobilization, bioremediation, biosorption

1 Introduction

1.1 Wood treatment with CCA

Chromated Copper Arsenate (CCA) (As_2CrCuO_9) is a water-based mixture of heavy metals/metalloid used as a wood preservative to increase its durability and protect it from fungal and bacterial degradation as well as attack by insects (Morais et al., 2021). Other names that refer to the same preservative are copper chrome arsenate or copper chromated arsenate. It is composed of 47.5% chromium oxide (CrO₃), 18.5% copper oxide (CuO) and 34% arsenic pentoxide (As_2O_5), which constitutes CCA type C that is commonly applied in many countries since it provides a suitable combination of resistance to leaching as well as



high efficacy (Moghaddam and Mulligan, 2008). Chromium enhances the fixation of copper and arsenic in the wood, copper provides resistance to fungi and bacteria while arsenic provides dual protection by preventing destruction of wood by insects and harsh weather conditions (Matos et al., 2010; Mohajerani et al., 2018). CCA has been in use since 1930s being the preservative of choice for treatment of wood intended for industrial use compared to other non-arsenic and non-chromium preservatives that include alkaline/ ammoniacal copper quaternary (ACQ), copper azole, micronized copper azole, creosote, pentachlorophenol (Penta) (Bolin and Smith, 2013; Jones et al., 2019).

Treatment of wood with CCA involves high-pressure impregnation process at the treatment plants to provide a minimum retention of at least 21 kg/m³. During the fixation, Cr (VI) is reduced to Cr (III), which strongly binds to lignin by forming a complex (Kartal, 2003). The three metals once bound to wood exist in the forms represented in Equation 1 (Abd El-Fatah et al., 2004)

$$4\text{CrO}_3 + 4\text{CuO} + 8\text{H}_3\text{AsO}_4 \rightarrow 4\text{CuHAsO}_4 + 4\text{CrAsO}_4 + 10\text{H}_2\text{O} + 3\text{O}_2$$
(1)

Wood species commonly used due to their durability include eucalyptus, pine, cedar and fir (Arriaga et al., 2023; KPLC, 2014). Wood treated with CCA has a variety of references that include "CCA treated timber," "tanalised timber," "pressure treated timber" and "permapine timber." The treated wood is used in framings, outdoor playground equipment, fence posts, backyard decks, marine structures, garden edging, landscaping, picnic tables, electricity poles and in building (Coles et al., 2014; Hall and Beder, 2005; Morais et al., 2021). Through there are other options for electricity distribution and transmission such as concrete poles and steel pylons, wood treated poles are the ones commonly used due to their relatively low cost and lighter weight making them affordable (Muthike and Ali, 2021).

The use of CCA treated wood in residential areas in countries such as Australia, Canada and United States has been restricted due to the risk of environmental contamination and associated health effects (Hall and Beder, 2005). However, wood preservation by use of CCA continues in developing countries; an activity that takes place at the wood treatment plants. Effluents from these plants find their way into the environment where Cr(III) ions undergo oxidation reactions to form Cr(VI) ions. Cr(VI) compounds are of great concern owing to their high water solubility, permeability through biological membranes and subsequent interaction with proteins (Ambi et al., 2020; Okello et al., 2012; Stern, 2010). Cr(VI) is known to be highly toxic, mutagenic and carcinogenic to humans and animals (Sharma et al., 2022; Vignati et al., 2010). Arsenic is also a known carcinogen (Palma-Lara et al., 2020). Moreover, the toxic effects of CCA have been reported to be more severe than those of its individual constituents. Thus, the presence of these metals in the environment may result in acute and chronic effects in humans, aquatic organisms and the entire terrestrial ecosystem (Shanker et al., 2005; Stern, 2010). Animals and humans are therefore at risk when exposed to arsenic, chromium and copper from the CCA treated wood, dust or contaminated soil/water (Hall and Beder, 2005; Morais et al., 2021).

Because of the environmental impact of CCA, a clear understanding of its toxicity, fate and potential removal strategies is necessary. Therefore, this review focuses on CCA elements in the environment, their toxicity, removal strategies and sustainable alternatives to mitigate adverse effects and safeguard human health and ecosystems for future generations. It comprehensively delves into removal and immobilization of CCA components from a wide range of matrices, including soil, water and wood waste building on earlier reviews that focused on a single matrix such as wood waste (Lopes et al., 2019; Mohammed et al., 2022), or examined other heavy metals rather than specifically addressing the distinct components of CCA (Qasem et al., 2021; Wang et al., 2022; Yeo et al., 2021; Zeng et al., 2024).



1.2 Review methodology

To identify the relevant literature for inclusion in this article, Google scholar was used as the primary database for thorough search. Some of the keywords employed during the search included; "chromium" "copper" "arsenic" "CCA" "bioremediation" "decontamination" and "biosorption". The choice of articles for inclusion was dependent on the titles and abstracts, which were found to be relevant to the subject matter that is, removal of chromium, copper and arsenic from contaminated matrices. The publications cited in this review encompasses journal articles, reports and theses. Data presented in graphical and tabular form in this work was extracted from the abstracts, methodologies, results and discussions of the studies covered. The years of publication considered in this review range between 2000 and 2023. The early years form the basis of our discussion whereas the recent years describe advances, which runs from 2018 up to 2023.

2 Environmental effects of CCA

2.1 Treated wood and associated environmental effects

CCA treated wood has a lifetime of 40 years after which it is disposed of as wood waste since it no longer offers the much needed service (Xing et al., 2020). Besides polluting the environment with CCA elements through leaching during its use, disposing of the wood after use can lead to further environmental contamination (Townsend et al., 2004). Figure 1 shows the various stages that wood undergoes from planting to treatment and final disposal at the end of its service, with most of the stages contributing to environmental pollution.

The continued use of CCA treated wood places, humans and animals at great risk of exposure due to the possible leaching of CCA components into the environment. A study by Mercer and Frostick, (2012) showed that all the three elements of CCA leach from wood. The two leaching experiments employed (batch leaching and lysimeter) showed that freshly treated wood leach more than weathered wood losing 24% arsenic, 6% chromium and 18% copper compared to 0.28%, 0.08% and 1.14% for weathered wood. The concentration of the metal (loids) in leachate from the two types of wood exceeded the environmental quality standards (EQS) of the European Commission Water Framework Directive set at 50, 32 and 28 µg/L for arsenic, chromium and copper, respectively. The World Health Organization (WHO) guideline for Cr, Cu and As in drinking water is 0.05, 2 and 0.01 mg/L, respectively (WHO, 2011). In another study, soil samples near utility poles in Canada reported concentrations of up to 37.5, 65.5, and 38.9 mmol/kg for Cu, Cr, and As, respectively. Levels of Cu, Cr, and As from rainwater runoff of freshly treated poles exposed to natural rain recorded concentrations of 14.0, 77.7 and 55.8 μ mol/L, with chromium and arsenic levels being higher than the recommended levels in drinking water of 15.7, 0.961 and 0.13 µmol/L by Canadian Council of Ministers of the Environment (CCME) (Coles et al., 2014). Leached CCA elements in rainwater runoff eventually drain into water bodies as illustrated in Figure 2.

Soil samples collected 20 cm from utility poles treated with CCA showed high contamination of 5,857, 3,815, and 3,797 mg/kg for As, Cr, and Cu, respectively, which were higher than the acceptable criteria for both industrial and residential soils (Villegas and Zagury, 2023). Another study on soil samples collected near utility poles reported concentrations of As, Cr and Cu at 265, 165 and 360 mg/kg, respectively (Gosselin and Zagury, 2020). Table 1 shows the levels of CCA components in contaminated soils from different parts of the world, with levels in most places exceeding the available guidelines by CCME. The presence of CCA components in soil and water bodies due to runoff paves the way for their uptake by plants and other organisms, eventually finding their way into the food chain.

Disposal of CCA treated wood waste is commonly done through landfilling of the wood itself or through incineration or a waste-toenergy process from which the ash is also landfilled (Choi et al., 2012). Unfortunately, the components can leach out of the treated wood contaminating the surrounding groundwater and soils, thus leading to human and animal exposure. Levels of arsenic and chromium in leachate from landfilled ash were reported to be 1.76 mg/L and 4.8 mg/L, which was 3 and 24 times greater compared to leachate from municipal waste landfills (Jambeck et al., 2007). Combustion and disposal of ash in landfills is of major concern because the process has been reported to convert Cr(III) to Cr(VI), which is more toxic and mobile (Song et al., 2006).



A more worrying scenario is how to handle and reclaim abandoned wood treatment plants due to contamination of such locations and their surrounding environment with heavy metals. Soil collected from such a plant that had been in operation for more than 40 years reported very high concentrations of 2,800, 2,100 and 3,300 mg/kg for Cr, Cu and As, respectively (Beiyuan et al., 2018). Given the environmental risks associated with CCA, there is a great need for sustainable alternatives and strategies for the restoration of these spaces to make them useful again.

2.2 CCA toxicity

CCA has been reported to be toxic to humans and animals upon exposure, with CCA being more severe than the individual elements. Workers in wood treatment plants are majorly exposed to CCA through inhalation, while the normal population is exposed through dermal exposure once in contact with treated wood or ingestion of contaminated soil, food or water. Arsenic exist in three forms, i.e., trivalent arsenite (As (III)), pentavalent arsenate (As(V)) and elemental arsenic (As), with both arsenite and arsenate being toxic (Yeo et al., 2021). According to the United States Environmental Protection Agency (United States EPA), inorganic arsenic is a Group A carcinogen (Morais et al., 2021). Arsenic also affects the central nervous system, the immune system and can lead to fetal mortality. Chromium exists of two forms, i.e., Cr(III) and Cr(VI), with the latter being more toxic and a potential carcinogen to humans. Its high toxicity stems from its oxidative nature corroding the respiratory system and ultimately leading to lung cancer. Though copper is an essential trace element, high levels are detrimental to humans and animals, with diseases related to copper metabolism being reported (Morais et al., 2021).

Since CCA is able to leach into the environment, contaminating soil and exposing humans, especially children, a recent study by Villegas and Zagury (2023) carried out risk characterization of CCA contaminated soil. The study assessed different exposure pathways, including dermal, inhalation and oral ingestion for industrial and residential settings. High hazard index (HI) greater than one was reported for oral and dermal pathways from the residential and industrial scenarios while the inhalation pathway resulted in a lower HI of less than one. The oral pathway contributed the highest carcinogenic risk for As and Cr(VI), with risk values greater than the acceptable value of less than 10^{-4} . These findings point to the need for strategies of preventing environmental pollution to protect humans from possible exposure.

3 Remediation of CCA-contaminated wood waste, soils and water

There are concerns on contamination of the environment by CCA elements through leaching from treated wood. Incineration of CCA treated wood produces ash contaminated with metals, which need to be removed before the ash is placed in landfills. A study by Solo-gabriele et al. (2002) indicated that the contribution of these metals in ash could be as high as 36% by weight for wood with high CCA retention of 40 kg/m³. Open burning has been reported to emit up to 14% of arsenic to the environment (Wasson et al., 2005). On the other hand, landfills take up much space and metals are able to leach to underground water with time. Thus, there is an urgent need

Country	As (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	Sampling points	References
Nigeria	39.55	313.97	200.00	Vicinity of a wood treatment factory	Uwumarongie-Ilori and Okieimen (2010)
Uganda	28.22	365.8	109.72	Random samples from a wood treatment plant	Nakiguli et al. (2020)
Korea	1.53	69.68	41.38	Near boardwalks made of CCA treated wood	Usman et al. (2012)
Sweden	1,320	421	164	Former industrial wood impregnation site (Fine soil particle size fraction)	Kumpiene et al. (2016)
Sweden	1,550	276	238	Former industrial wood impregnation site (Coarse soil particle size fraction)	
New Zealand	3,300	2,800	2,100	Former treatment facility	Beiyuan et al. (2018)
Canada	265	165	360	Near CCA - treated utility poles	Gosselin and Zagury (2020)
United States of America	38	39	51	Soils beneath CCA treated wood structures in schools	Gardner et al. (2013)
China	314.90	133.60	266.35	Soils under CCA-treated boardwalks	Zeng et al. (2023)
Kenya	0.25-0.89	_	_	Soils around wood treatment plant	Opuru et al. (2020)
	0.04–0.26 (mg/L)	_		Water from a river close to the wood treatment plants	
Ghana	8.9	47	89.8	Soils around in-service CCA treated utility poles	Oppong et al. (2021)
Nigeria	22.58	63.09	89.73	Soils around CCA-treated utility poles in a Nigerian University	Adedeji et al. (2023)
Finland	850	1,590	791	Humus soil from an old CCA wood impregnation site	Kilpi-Koski et al. (2019)
Denmark	1,364	540	1,662	Soil from a former wood impregnation site	Frick et al. (2019)
CCME guideline for Industrial soils	12	78	91	_	CCME (2007)
CCME guideline for Agricultural soils	12	64	63	_	CCME (2007)

TABLE 1 Levels of CCA in contaminated soils and water in different parts of the world.

for environmentally sustainable strategies for handling wood waste to reduce environmental contamination. The use of wood waste fiber to produce reinforced polypropylene composites of high tensile strength for use in the construction industry has been reported (Nelson et al., 2023). Pyrolysis has also been explored as a potential alternative for disposal of wood that is out of service (Junk, 2022). Several methods have been proposed for the remediation of CCAcontaminated soils, water and wood waste. These include chemical remediation (Gezer et al., 2006; Janin et al., 2012) bioremediation (Xing et al., 2023), phytoremediation (Li et al., 2022), and physical methods such as soil washing and adsorption (Frighetto et al., 2016) among others as discussed in this section.

3.1 Chemical remediation

Chemical remediation involves the application of chemical amendments, such as iron oxide or calcium hydroxide or any other viable chemical, to immobilize and precipitate heavy metals, reducing their bioavailability and mobility in the environment. Organic acids such as citric, acetic, formic, oxalic, fumaric, tartaric, gluconic, and maleic, as well as mineral acids, such as sulphuric, hydrochloric, nitric, and phosphoric have been utilized (Frighetto et al., 2016). The use of NaOH and citric acid buffer at a pH of 3.5 achieved 100% extraction of copper from wood after 40 days. An optimized method using oxalic acid and metal tolerant bacteria (*Bacillus Licheniformis* CC01) afforded 78% 97% and 93% removal of copper, chromium and arsenic, respectively (Clausen, 2000). Another study which used the same bacteria and oxalic acid achieved very high removal efficiency of 90% copper (CuO), 80% chromium (CrO₃), and 100% arsenic (As₂O₅). Acids convert the CCA elements into their water-soluble form.

Ethylenediaminetetracetic acid (EDTA) as a chelating agent has been widely used to form soluble complexes with metals. A 1% EDTA solution removed 60% copper, 13% chromium, and 25% arsenic from treated chips after 24 h (Kartal, 2003). Oleic acid was able to remove 96% copper, 78% chromium and 96% arsenic at a pH or 2.0 in 2 days. A high pH of 5.0 was found to be ineffective, especially in removal of chromium (Gezer et al., 2006). Leaching of CCA metals by use of sulphuric acid followed by precipitation with calcium hydroxide reported 98% removal of arsenic, 93% chromium and 96% copper from wood (Janin et al., 2012). Electro-removal method based on application of an electric field led to a reduction of 79.5, 87.4, and 81.3% in the mean concentrations of Cu, Cr, and As from treated waste wood. This method has been found to be effective since the electrolytic fluid containing the metals can be reused in production of CCA to provide further protection of wood (José de Castro et al., 2021). The application of super critical fluid extraction (SFE) technology using CO_2 and organophosphorus chelating agents achieved extraction efficiencies of 63.5, 28.6, and 31.3% for Cu, Cr, and As, respectively, from wood waste (Abd El-Fatah et al., 2004). Though chemical methods, have showed a level of efficiency in remediation of wood waste, their disadvantage stems from the fact that chemicals are harmful and they lead to secondary environmental pollution (Xing et al., 2023). Traces of chemicals in decontaminated wood also limits its use in making blended products when mixed with other substances like geopolymer cement (Can and Sivrikaya, 2022; Elmira et al., 2023). As an alternative, bioremediation is hereby discussed.

3.2 Bioremediation

Bioremediation involves the use of microorganisms to eliminate or reduce the concentrations of pollutants. Microorganisms such as fungi generate organic acids that enhance the acidity of the substrate, promoting the solubility of chromium and arsenic, thus facilitating their removal. Oxalic acid, among other organic acids displays a number of properties including chelating ability, reducing agent and ability to create a lower pH for removal of CCA elements. A study using oxalic acid producing fungi *Fomitopsis palustris, Coniophora puteana*, and *Laetiporus sulphureus* reported arsenic removal rates of 100% and 85% for *F. palustris* and *L. sulphureus* (Kartal et al., 2004). Xing et al. (2023) demonstrated *Yarrowia lipolytica* ability to remove 82.8% copper, 43.1% chromium and 63.8% arsenic through the production of oxalic, malic, acetic and citric acids.

Besides bioremediation, bioprocessing methods that take advantage of bioremediation and biodegradation by use of fungi have been explored as alternatives to disposal of CCA wood waste in landfills. Four fungal isolates, Meruliporia incrassata (TFFH-294), Antrodia radiculosa (MJL-630), M. incrassata (Mad-563) and A. radiculosa (FP-90848-T) degraded CCA-treated wood by more than 20% of the original dry weight of the wood (Illman and Yang, 2004). Similarly, another study used five isolates of brown-rot fungi to decay treated wood indicating mass losses of up to 60% and 53% by Crustoderma sp. KUC8611 and F. palustris, respectively. The removal of CrO3 (79%), As2O5 (87%), CuO (50%) by F. palustris was more efficient in CrO3 and As2O5 due to the high production of oxalic acid by the fungi (Choi et al., 2012). Fungi also remove these metals via bio-absorption into their cell structure as well as complexation with low molecular weight proteins (Kartal, 2003). While fungi have been reported to be efficient in removal of metals, they require several days to achieve this efficiency and may also result in transformation of wood constituents (Chang et al., 2012). Additionally, they may not assimilate all the components of CCA (Lopes et al., 2019).

Metal tolerant bacteria have been utilized to remove chromium; a highly stubborn metal in CCA. Three isolates, *Acinetobacter calcoaceticus* FN02, *Aureobacterium esteroaromaticum* VV03, and *Klebsiella oxytoca* CC08 were reported to release 98% of chromium, while *Bacillus licheniformis* CC01 released 93% of copper (Clausen, 2000). A mixed culture of lactic acid bacteria (*Lactobacillus*) *bulgaricus* and *Streptococcus thermophiles*) was able to extract all three metal ions from the CCA treated wood within 4 days at 93% for copper, 86.5% for chromium, and 97.8% for of arsenic. These bacteria were also reported to produce pyruvic acid that aided in extraction of metals (Chang et al., 2012). Earthworms are other microorganisms that have shown potential to remove arsenic from CCA wood waste. *Eisenia fetida*, type of earthworms, were able to bioaccumulate arsenic in their tissues by feeding on CCA wood waste mixed with cow dung leading to their removal (Mohammed et al., 2022). Notwithstanding the promising results associated with bioremediation, long treatment time is a major drawback that should be probed to accelerate the process of acquiring results during research (Costa et al., 2022).

3.3 Pyrolysis

Pyrolysis thermochemical is а (physico-chemical) decomposition of biomass in limited supply or absence of oxygen to yield biochar, bio-oil and gases consisting of CO, CO₂, and H₂ that do not condense easily. Gmar et al. (2022) investigated a combination of pyrolysis and plastic encapsulation as a way of safely disposing contaminated wood. This study reported the highest biochar yield at 300°C with heavy metal retentions of 76, 91% and 83% for As, Cr and Cu, respectively. However, there was notable leaching of heavy metals from the biochars. To prevent leaching, encapsulation of the biochars with high-density polyethylene reduced leaching by 96, 95% and 91% for As, Cr and Cu, respectively. In another study, pyrolysis of CCA-treated wood released 41% of arsenic at 800°C, whereas gasification led to release of 90% arsenic. The release of arsenic was suppressed during pyrolysis by mixing organic sludge rich in iron and calcium with wood treated with CCA (Kato et al., 2021). Maximum recovery of arsenic from the solid state occurred at 475 C during pyrolysis as reported by Junk, (2022). At this temperature, the bio-oil yield was 29 wt% with a 606 ppm arsenic concentration representing a 6.7 wt% of the original arsenic. Pyrolysis reduces As(V) to As(III) as evidenced by the presence of (As2O3) in the bio-oil. Volatilization of arsenic was observed at temperatures above 475 C. It was noted that copper in CCA affects the production of bio-oil during pyrolysis, led to increased char yield and reduced arsenic recovery. Pyrolysis is an already established method for decontamination of CCA-treated wood waste as it involves fairly low capital. Compared to other similar technologies like gasification and incineration, less gaseous emissions are produced during pyrolysis. However, the energy requirements are high and further investigations are necessary to lower the cost of this process. Table 2 summarizes some of the strategies for removal of CCA elements from wood waste.

3.4 Immobilization technology

Immobilization techniques prevent the free movement of contaminants in the matrix and surrounding media through solidification or stabilization. Solidification involves changes in the physical properties of a waste that include compressive strength, permeability and encapsulation, while stabilization

Strategies for removal of CCA elements		Performance %			Conditions	References
		Cr	Cu	As		
Chemical remediation	Mixture of 5% oxalic acid and 0.5 mol/L phosphoric acid	87.4	79.5	81.3	For Cr and Cu: time (60 min), mass of treated wood (5 g) and voltage (25 V) For As: time (20 min), mass (15 g), and voltage (25 V)	José de Castro et al. (2021)
	Glycerol/Choline Chloride Deep Eutectic Solvent (DES)	30	65.5	10	Mass of DES Solution (50 g), time (2 h), temperature 150°C	Can and Sivrikaya (2022)
	0.05 M dilute oxalic acid	92	35	100	wood particle size (1.18–2.34 cm), solid: liquid ratio (10:200 g: mL), temperature (65°C), agitation speed (300 rpm). pH (1.8) for Cr and As, pH (3.2) and Cu	Elmira et al. (2023)
Bioremediation	Yarrowia lipolytica fungus	43.1	82.8	68.3	pH (2.7-3.0), time (21 days), mass of treated wood waste (5 g), optical density (0.05)	Xing et al. (2023)
	Wolfiporia cocos fungus	61	75-78	85	Agitation speed (120 rpm), temperature (27°C), time (10 days),	Costa et al. (2022)
	Antrodia xantha fungus	46	75-78	71	sterile liquid culture medium (100 mL)	
	Fibroporia radiculosa fungus	38	96	59		
Pyrolysis	Stabilization of heavy metals in biochar	91	83	75	Temperature (300°C), mass of CCA-treated wood (100 g), particle size (1.4–2 mm), residence time (1 h)	Gmar et al. (2022)
	Chromium recovery in biochar	Above 80	_	_	Mass of CCA-treated wood (25 g), particle size (780 μ m), heating rate (5°C-15°C min ⁻¹), residence time (30 min), temperature	Junk (2022)
	Arsenic recovery in bio-oil	_	_	28.9	(5/ C for Cr, and 4/5 C for As)	

TABLE 2 Summary of remediation methods for treating CCA-contaminated wood waste.

involves chemical changes of the hazardous constituents in a matrix to a less soluble, mobile or toxic form. Zeolite modified with iron has been shown reduce the amount of arsenic in contaminated soil as evidenced by reduced uptake of arsenic by plants. The arsenic concentration in leaves was reported to have reduced from 36 mg/kg to 15 mg/kg (Alessandro, 2019). A novel electrokinetics method for removal of Cr(VI) in contaminated soils reported 85.50% efficiency (Xu et al., 2017). By incorporating permeable reactive barrier (PRB) consisting of hydrocalumite (CaAl-LDH), the removal efficiency increased to 96.49%. A similar approach for removal of arsenic in contaminated soils involved coupling electro kinetics with PRB consisting of iron-manganese-carbon layered double hydroxide (Fe/Mn/C-LDH). Bamboo was used in the synthesis of the Fe/Mn/C-LDH fillers as the template due to its porous nature. The study established that the rate of removal of arsenic from contaminated soils was influenced by electric field strength, PRB position, moisture content as well as type of PRB filler. Maximum removal rates were 95.71% when the PRB filler was positioned in the middle, where the soil pH ranged between 5 and 8 providing suitable conditions for arsenic adsorption onto the PRB filler Fe/Mn/C-LDH. Moisture content of 35% gave the best removal rates whereas a voltage gradient of 2 V/cm gave the optimal arsenic immobilization (Zhu et al., 2022).

Stabilization of soil contaminated with arsenic by use of acid mine drainage-treated sludge (AMDS), which is a problematic waste in coal mining has been reported. The sludge is composed of iron and calcium minerals with high adsorption capacity for heavy metals (Amanda and Moersidik, 2019). The stabilization efficiency of acid mine drainage sludge was higher than 85% by the use of 3% of AMDS, which was attributed to ligand exchange between the Fe- (oxide) hydroxide and the arsenate to form a new complex, the cation bridge effect between the AMDS surface and the arsenate as well as co-precipitation (Tak et al., 2023). Starch stabilized Fe/Cu nanoparticles (0.04 wt% of starch) are capable of *in situ* stabilization of arsenic and chromium in soil contaminated with CCA. Treatment of contaminated soil with 0.4 g/L nanoparticles at a soil: liquid ratio of 0.1 reduced leachable arsenic in water from 55 to 4.2 μ g/L through immobilization to nanoparticles (Babaee et al., 2017).

3.5 Phytoremediation

Phytoremediation has gained widespread application in the remediation of soils contaminated with heavy metals due to its environmentally friendly nature and associated low cost. This method involves processes such as phytoextraction (involving metals), phytoaccumulation (involving uptake of heavy accumulation translocation and of heavy metals), phytovolatilization (emission of heavy metals to the atmosphere in volatile form) and phytostablization (stabilization of heavy metals in the root zone of plants) (Li et al., 2022; Shah and Daverey, 2020). Phytoaccumulation of arsenic from contaminated soils was conducted using fern (Pteris vittata) as a trap plant. Soil and cow dung mixture (ratio of 1:1) was spiked with arsenic in the form of arsenic trioxide at different concentrations of 0 ppm (A0), 500 ppm (A1), 1,000 ppm (A2) and 2,000 ppm (A3). After 60 days, the fern had accumulated 0.9 ppm, 1769.9 ppm, 15,332.6 ppm and 23,837.2 ppm for A0, A1, A2 and A3, respectively. From the results, the soil seems to have been contaminated with arsenic prior to the experiment since the reported levels were higher than the spiked amount (Uddin et al., 2015). Fern is reported to be a hyperaccumulator, thus an excellent plant for phytoremediation.

The use of maize (Zea mays L.) in phytostabilization of contaminated soils from a CCA wood treatment plant amended with sewage sludge biosolid as an additional stabilizer has been reported. The results showed that phytostabilization using maize was greatly improved by the application of the sewage sludge biosolid through reduced mobility (Nakiguli et al., 2020). In another study, Fomitopsis arundinacea, M. sativa, and S. purpurea were investigated for their potential in phytoaccumulation of pentachlorophenol (PCP) and CCA. The experiment involved mechanical scarification and amendment of the soil with commercial soil to enhance germination of the plants by reducing the severe soil mineralization and compaction. Thereafter, the herbaceous species were sown manually followed by irrigation and were monitored for 4 years. At the end of the experiment, aerial tissues of F. arundinacea, M. sativa, and S. purpurea had accumulated 6.5, 10, and 8.75 mg/kg of copper, respectively, with chromium and arsenic being below detection limit. This was attributed to their interactions with soil particles, which reduces their bioavailability, and competition with other ions like phosphates present in the soil. Also the elements might have been translocated in the roots only and not the other parts of the plant (Yanitch et al., 2020). The use of plants in phytoremediation of contaminated soils is therefore an effective method owing to their ability to accumulate multiple contaminants from soils and also contribute to phytostabilization (Priya et al., 2023). However, this phytoaccumulation has limitations regarding safe disposal of harvested plants after phytoremediation process as well as recovery of the heavy metals. More research is therefore needed on sustainable ways of disposing contaminated plants (Shah and Daverey, 2020).

Some of the sustainable ways that can be considered to avoid reentry of heavy metals into the environment may include; pyrolysis and stabilization with agents like cement to immobilize the contaminants. Another way that can be considered is treatment of the plant biomass with microorganisms, which convert biomass into soluble organic compounds that dissolve the heavy metals, thus reducing the volume occupied by biomass and consequently aiding the management of the waste. In addition, the plants can be used in the production of bio-energy. Moreover, when the harvested plants are mixed with other organic materials and left to decompose, an amendment that is rich in nutrients is produced which can be applied in agriculture (Priva et al., 2023; Singh et al., 2022). For the recovery of metals, suitable chemicals like acids can be used to recover metals when in high concentrations. Table 3 gives a comparison of the performance of the different remediation approaches in contaminated soils from recent studies.

3.6 Adsorption

Adsorption, in the context of remediation, refers to the process by which contaminants in water, air, or soil adhere to the surface of a solid material (adsorbent), effectively removing them from the environment. This occurs due to physical or chemical interactions between the contaminant molecules and the adsorbent surface. Hexavalent chromium usually exists in water/ wastewater as oxyanions such as chromate (CrO_4^{-2}), hydrogen chromate ($HCrO_4^{-}$), and dichromate ($Cr_2O_7^{-2}$), which are mostly toxic, carcinogenic and mutagenic and do not precipitate easily using conventional precipitation methods. Similarly, the predominant forms of arsenate are dihydrogen arsenate $(H_2AsO_4^{-})$, hydrogen arsenate $(HAsO_4^{2-})$ and arsenate ion (AsO_4^{3-}) (Saikia et al., 2011). The removal of heavy metals from contaminated water by use of adsorbents is explained by different adsorption mechanisms such as complexation, electrostatic interactions, ion exchange, redox reactions, physical adsorption and precipitation (Wu et al., 2016). Figure 3 shows some of the adsorbents have been utilized for removal of CCA elements as outlined in the sections that follow.

3.6.1 Inorganic adsorbents

A study by Khaodhiar et al. (2000) reported on removal of copper, arsenate and chromate in a single and multi-solute electrolyte system of NaNO3 using iron-oxide coated sand. At pH above 6.0, 90% of copper was adsorbed. In a binary system of copper and arsenate, the amount of copper adsorbed was shown to increase due to formation of copper arsenate complex as well as the formation of inner-sphere complex that decreases the surface charge of iron oxide. Chromate or even a mixture of chromate and arsenate did not affect adsorption of copper. Highest adsorption of 35% for chromium was achieved at lower pH of 3.0. Copper was shown to influence the adsorption of chromate positively by increasing the surface charge of the iron oxide. Arsenate was also found to adsorb at low pH values with the presence of copper or chromate having no influence on the amount adsorbed. Manganese oxide sorbents have also been evaluated for removal of copper, chromium and arsenic from water reporting appreciable removal. Manganese coated sand was used for removal of chromium, copper and arsenic from a synthetic leachate solution using a sorbent dosage of 20 g/L. For a synthetic leachate solution containing 2, 1 and 0.2 mg/L total chromium, copper and arsenic, respectively, the sorbent reported 70%, 98%, 99% removal of the heavy metals (Wu et al., 2016).

Many other studies have reported on use of various sorbents to a wide range of heavy metals including the elements of CCA. Chitosan coated fly ash (SiO $_2$ - based), showed adsorption capacity of 36.22 mg/g for Cr(III, VI), 28.65 mg/g for Cu(II), and 19.10 mg/ g for As(V). The multiple functional groups in chitosan enhance its adsorption capacity, with Cu(II) achieving the highest removal efficiency of 89.5% using 0.3 g of adsorbent (Adamczuk and Kołodyńska, 2015). In another study, magnetic biochars modified with chitosan showed potential for heavy metal removal owing to chitosan having amine functional group on the surface of the sorbent as well as its large surface area for adsorption. Chitosan combined with magnetic loofah biochar (CMLB) was tested for Cr(VI) and Cu(II) ions removal from wastewater where it exhibited higher adsorption capacity in comparison to the pristine biochar. The magnetic property of this adsorbent enhanced the separation of CMLB from aqueous solutions. The highest adsorption capacity for Cr(III) and Cu(II) was reported as 30.14 mg/g and 54.68 mg/g, respectively using 40% CMLB where equilibrium was attained after 18 h. Also, it was observed that lower pH values (1.0-6.0) favored existence of Cr(VI) in the form HCrO₄⁻ and Cr₂O₇²⁻ which resulted to higher adsorption capacity of the sorbent. Adsorption capacity decreased with increase in pH which led to competition between

Approaches for soil remediation		Heavy metal	Concentration in soil (mg/kg)		Efficiency %	Conditions	References
			Before	After			
Immobilization Oxalate and S,Sethylene-odisuccinic-ad Dithionite-ci (DCB) follow washing Acidified NH followed by 2	Oxalate and EDDS, S,Sethylene-diamine- disuccinic-acid) washing	Cr	2,800	2,688	4	Liquid to soil ratio (10 L/kg), shaking (30 rpm), time (2 h), temperature (20°C \pm 2°C)	Beiyuan et al. (2018)
		Cu	2,100	1,323	37		
		As	3,300	2,772	16		
	Dithionite-citrate-bicarbonate (DCB) followed by 2-h EDDS washing	Cr	2,800	2,150	24	Liquid to-soil ratio (10 L/kg), shaking (30 rpm), time (2 h), temperature (20°C ± 2°C)	
		Cu	2,100	1,302	38		
		As	3,300	1749	47		
	Acidified NH ₂ OH-HCl	Cr	2,800	2,520	10	Liquid to-soil ratio (10 L/kg), shaking (30 rpm), time (2 h), temperature (20°C \pm 2°C)	
	followed by 2-n EDDS washing	Cu	2,100	987	53		
		As	3,300	2,409	27		
Phytoremediation	Phytoremediation using F. arundinacea, M. sativa, and S. purpurea	Cr	97	14	86	Time (4 years)	Yanitch et al. (2020)
		Cu	84	5	94		
		As	7	<5	43		
	Sewage sludge biosolid amendment	Cr	365.80	136.47	63	Time (14 days), temperature (25°C)	Nakiguli et al. (2020)
		Cu	109.72	64.82	41		
		As	28.22	4.98	82		
		Cu	1,662 µg/g	914.10	45		
		As	1,364 µg/g	777.48	43	7	

TABLE 3 Comparative studies for remediation of soils contaminated with chromium, copper and arsenic.

hydroxide complexes and Cr(VI) ions for active sites (Xiao et al., 2019).

Activated alumina powder (AAP) has shown promising results for removal Cr(III) and Cu(II) from industrial waste water. At pH 5, precipitation of Cr and Cu occurred for high metal ion concentration. The percentage removal of Cr(III) and Cu(II) was maximum at pH 4.7 and pH 3, respectively. Results of the study indicated that equilibrium for Cr(III) and Cu(II) was attained at 50 min and 40 min, respectively. The highest removal of Cu(II) from a 0.1 M solution was reported as 99.8% using 10 g of sorbent whereas for Cr(III) it was 99.4% using 2 g of AAP from a 0.01 M solution. Additionally, the effect of particle size on the removal of heavy metal ions was studied where all the particle sizes (125, 250 and 420 mm) resulted to 99% removal of the ions. Cr(III) and Cu(II) adsorption increased as the temperature increased (Rajurkar et al., 2011).

3.6.2 Bioadsorbents

Biomass-derived adsorbents are desirable in that they can be reused several times to remove metals without loss in their efficiency, thus reducing the cost of remediation. Some of the naturally available materials that have been utilized as adsorbents in removal of Cr(III), Cu(II) and As(V) in single and multi-solute system include; activated carbon, corncob, apple and orange peelings, oak and pine bark, biopolymers among many others. Naturally occurring biopolymers (chitin and chitosan) are rich in amine and hydroxyl groups that can form complexes with metal ions. Hassoune et al. (2018) reported 80.64 and 26.25 mg/g maximum uptake for the removal of Cr(VI) from water using gelatin-chestnut (GC) and gelatin-quebracho (GQ) biopolymer adsorbents, respectively. Chitosan, a biopolymer extracted from shrimp shell reported 98.5% and 97.4% adsorption efficiencies for arsenic and chromium, respectively after modification using NaOH and H_2SO_4 that enhanced functional groups increasing the adsorption capacity (Rahman et al., 2023). CCA treated sawdust exposed to a solution containing chitin achieved 74% copper, 62% chromium, and 63% arsenic removal, while chitosan removed 57% copper, 43% chromium, and 30% arsenic.

Azadirachta indica leaf was used to produce activated charcoal for Cu(II) adsorption. In batch experiments, 91.5% removal was achieved at 46°C with a 10 g/L dose. Column experiments optimized at 5 mL/min flow rate, 5 mg/L concentration, and 20 cm bed height showed a maximum adsorption capacity of 185.8 mg/g (Patel, 2020). Ambi et al. (2020) used *Hibiscus sabdariffa* calyces extract, rich in antioxidants, for Cr (VI) removal reporting promising results consistent with Clovis et al. (2020). Use of tea waste has gained ground in adsorption because it is an effective and inexpensive technique. Nagra et al. (2023) used tea waste in column sorption experiment, achieving 92% As(V) removal at pH 5. Tea waste showed competitive adsorption for Cu(II) and Pb(II) from aqueous solutions with a maximum Cu(II) adsorption capacity 37.17 mg/g in single element solutions and 28.41 mg/g in mixed



solutions (Pasgar et al., 2022). Competition between Cr(VI), Cu(II) and As(V) ions during adsorption should therefore be investigated through research for efficiency in their removal. At a at pH 2, Jeyaseelan and Gupta (2016) reported 99% removal efficiency for Cr(VI) using green tea leaves.

Numerous studies have been conducted using water hyacinth, both in its raw form and activated form, to evaluate its capability in treatment of waste water containing heavy metals (Abbas et al., 2021; Ajibade et al., 2013; Gogoi et al., 2017; Lissy and Madhu, 2011; Panneerselvam and Priya, 2023). Water hyacinth (Eichhornia crassipes) shoot powder (WHSP) removed more than 98% of Cu(II) and Cr(VI) from both standard and tannery solutions as reported by (Sarkar et al., 2017). On the other hand, Huynh et al. (2021) reported lower removal efficiencies of 60.8% for As(V) and 60.7% for Cu(II) using water hyacinth grown in water contaminated with 0.5 mg/L arsenic and 5 mg/L after 30 days. Polyflavonoids tannins from the barks of Rhizophora apiculata mangrove, showed adsorption capacity of 8.78 mg/g for Cu (II) at pH 5 (Oo et al., 2009). Furthermore, flavonoids such as quercetin and their derivatives can reduce Cr(VI) to Cr(III) as reported by Okello et al. (2012), therefore their use in the removal of Cr(VI) has an added advantage. Though adsorption is also an alternative in removal of CCA elements, bioremediation stands out as more versatile than the use of adsorbents that require regeneration by use of chemicals (Kartal and Imamura, 2005). During regeneration, if the metals are not captured for recycling in CCA, the process may lead to further environmental degradation. Comparative studies on various biosorbents for removal of CCA components from aqueous solutions are summarized in Table 4.

3.6.3 Adsorbent synthesis using nanotechnology and green synthesis

Nanotechnology has been widely investigated for synthesis of highly efficient adsorbents for heavy metal removal. Iron oxide nanoparticles strongly adsorb arsenic and chromium due to their enhanced metal-binding capacity, improving the overall efficiency of adsorbents (Chowdhury and Yanful, 2010). Boruah et al. (2023) conducted a comprehensive literature review on iron-based nanomaterials for arsenic removal by looking at removal efficiency, factors influencing adsorption, and adsorbent regeneration capacity. The findings showed that magnetite and bimetallic nanomaterials are particularly effective for removing both Arsenic (III) and Arsenic (V). The findings corroborate the work of Parajuli et al. (2020) where magnetite nanoparticles derived from extracts of A. indica adsorbed As(V) successfully in the form of H₂AsO₄through electrostatic attraction at low pH. Additionally, zerovalent iron nanoparticles have been shown to be efficient in removal of chromium (VI) with further benefits in reduction of chromium (VI) to Cr (III) in the presence of organic acids such as citric acid (Yang et al., 2017; Zhou et al., 2018).

Green synthesis, devoid of chemical use, presents a futuristic approach and departure from chemical synthesis in the production of nanoparticles for heavy metal adsorption. Plants contain numerous reducing agents and are considered as the main factory for green synthesis (Shafey, 2020). Coprecipitation of FeCl₃.6H₂O and FeSO₄.7H₂O using *A. indica* leaves extract was applied in the green synthesis of magnetite (Fe₃O₄) nanoparticles (MNPs) in an inert environment, where the molar ratio of the iron salts was 2:1. The maximum

Biosorbent	Heavy metal ion	Maximum adsorption capacity (mg/g)	рН	Removal efficiency (%)	Textural properties	References
Achiote (Bixa Orellana)	Cr(VI)	_	_	82.8	_	Kumar et al. (2022)
	As(III)	_	_	40.42		
Jungle geranium (<i>Ixora Coccinea</i>) leaf powder	Cr(VI)	150.25	2	71	Both amorphous and crystalline characteristics	Bhat et al. (2023)
Water-hyacinth (<i>Eichhornia crassipes</i>) shoot powder	Cr(VI)	_	7 ± 0.2	87.50	_	Shaibur et al. (2022)
	Cu(II)	_	7 ± 0.2	83.35		
Arum (<i>Colocasia sculenta</i>) shoot powder	Cr(VI)	_	7 ± 0.2	77.08		
	Cu(II)	_	7 ± 0.2	75.0		
Moringa (<i>Moringa oleifera</i>) leaves	Cr(VI)	-	4.8-6.2	>90	_	Madhuranthakam et al. (2021)
Onion (Allium Cepa) seeds	Cr(VI)	0.67	7	34.0	Porous surface	Sheikh et al. (2021)
	Cu(II)	1.68	7	99	Porous surface	Sheikh et al. (2021)
Moringa (<i>Moringa oleifera</i>) seeds	Cu(II)	23.3	6	99	BET surface area (18.9 m ² g ⁻¹), Pore diameter (1.6 nm), Zeta potential (-28 ± 0.012)	Tokay and Akpınar (2021)
Coconut (<i>Cocos nucifera</i>) coir	Cu(II)	1.34	6	92	BET surface area (16.7 m ² g ⁻¹), Pore diameter (3.9 nm), Zeta potential (-22 ± 0.5)	Tokay and Akpınar (2021)
Rice (Oryza sativa) husk	Cu(II)	1.56	6	98	BET surface area (8.1 m ² g ⁻¹), Pore diameter (1.5 nm), Zeta potential (-38.2 \pm 0.2)	Tokay and Akpınar (2021)
Golden Shower (Cassia fistula L)	As(III)	1.13	6	91	_	Giri et al. (2022)

TABLE 4 Comparative studies of some bioadsorbents.

adsorption capacity of the MNPs was 62.89 mg/g at pH 2 for As(V) (Parajuli et al., 2020). A comparative study on removal of arsenic from water using iron oxide nanoparticles synthesized through a green procedure utilizing five different leaf extracts is reported by Kamath et al. (2020). The researchers used the following leaves for green synthesis; black tea leaves (Camellia sinensis), oak tree leaves (Quercus viriniana), green tea leaves (C. sinensis), pomegranate leaves (Punica granatum) and eucalyptus leaves (Eucalyptus globulus) reporting adsorption capacities of 18.98, 32.05, 13.70, 11.65 and 39.84 mg/g, respectively. This study introduced unprecedented oak leaves as potential adsorbents in green synthesis with a very high adsorption capacity that can be useful in nanotechnology. Vaseghi et al. (2019) explored a simple method for the removal of chromium and copper from aqueous solutions through a simultaneous process of bio-reduction and adsorption using Eryngium campestre leaf extract. In the study, nanoparticles were produced by reduction of metal ions by the phytochemicals (mainly phenolic acids and flavonoids) found in the leaf extract achieving removal efficiencies of 98.92% and 98.16% for Cu and Cr at pH 7, respectively. Incorporation of green synthesis in nanotechnology therefore provides a synergistic approach in adsorption of Cr(VI), Cu(II) and As(V), presenting a promising avenue for future research.

4 Sustainable alternatives to CCA preservative

Several arsenic free alternatives for CCA have been developed with copper as the primary active component. This is because copper provides the best fungicide protection and exhibits low mammalian toxicity. These alternatives include: acid copper chromate, alkaline copper quat, ammoniacal copper citrate, copper azole, copper dimethyldithio-carbamate, copper HDO, and borates. Since they lack arsenic in their formulation, these alternatives have an advantage of eliminating leaching of arsenic, which is of great concern due to its toxicity in the environment. However, wood treated with these alternatives is 10%-30% more costly than CCA treated wood due to the high cost of the chemicals. In addition, with the exception of acid copper chromate, these alternatives have a propensity of corrosiveness to metal fasteners compared to CCA Leaching of these CCA alternatives has also been reported, with the release of copper being higher or greater than in the CCA chemical. This is attributed to the higher proportion of copper in the CCA alternatives. Nevertheless, presence of copper and co-biocides into the environment is of less concern to environmentalists, since the associated mammalian toxicity is lower as opposed to arsenic in CCA (Lebow, 2004; Solo-gabriele et al., 2004). A study by Belizário et al. (2023) reported Micronized Copper Azol type C (MCA-C), which consists of tebuconazole, propiconazole, and micronized copper to be a potential alternative to CCA. The MCA-C alternative does not contain chromium or arsenic and therefore offers a suitable choice for wood preservative. Other possible CCA alternatives include; creosote, copper naphthenate, pentachlorophenol and ammoniacal copper zinc arsenate (Mankowski et al., 2023).

5 Conclusion and future perspectives

CCA elements in the environment is a major concern due to the associated risk of human and animal exposure. Therefore, the management of CCA waste is in tandem with the spirit of sustainable development goals (SDGs). Several remediation strategies have been highlighted in this review including chemical remediation, bioremediation, pyrolysis, immobilization, phytoremediation and adsorption. Despite the high efficiencies displayed by chemical remediation, it presents a major drawback since it is associated with secondary contaminants. On the other hand, pyrolysis as a way of disposal of wood waste could be a better alternative since it produces waste that is manageable but the energy requirements are too high. Recycling of CCA treated wood in what is referred to as waste-to-energy process as well as conversion of the wood waste to wood chips is a promising route of dealing with CCA treated wood for sustainable development. These processes lead to reduction of space occupied by waste that can be utilized for agriculture. Use of landfills is also greatly reduced thus abating underground water contamination as well production of useful energy that can be converted into different forms.

The use of microorganisms in bioremediation is a viable approach as they do not introduce secondary pollutants to the environment. Additionally, their ability to generate organic acids enhances the solubility of heavy metals, thereby facilitating the removal of the contaminants. However, the efficiency of microorganisms typically manifests after several days, making this method time-intensive. Furthermore, their application may result in the transformation of wood constituents. Bioadsorbents offer an effective and sustainable alternative for the removal of heavy metals from the environment due to their eco-friendly nature and low cost, presenting a promising approach to heavy metal remediation. Their natural abundance ensures a reliable and continuous supply, and their non-toxic properties prevent the introduction of harmful chemicals into the environment, making them a safer and more sustainable option. However, their disposal after use presents a major challenge. Given that each remediation

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technique has its own drawbacks and the increasing need for sustainable solutions, a synergistic approach that integrates multiple strategies for the removal of chromium, copper and arsenic should be the future focus in addressing CCA contamination.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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