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Treatment of Wastewater Effluent with Heavy Metal Pollution Using a Nano Ecological Recycled Concrete

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Abstract: Water pollution exacerbates water stress and poses a great threat to the ecosystem and human health. Construction and demolition waste (CDW) due to rapid urbanization also causes heavy environmental burdens. A major proportion of CDW can be effectively converted into recycled aggregates, which can be reused in many fields, including environment remediation. In this study, a nano ecological recycled concrete (nano-ERC) was produced with recycled aggregates and copper oxide nanoparticles (nCuO) to remove heavy metals (HMs) from a simulated wastewater effluent (SWE) for further treatment. Recycled aggregates were obtained from CDW, thereby simultaneously reducing the treatment cost of the SWE and the environmental burden of solid waste. The adsorption capacity of nano-ERC was presumed to be significantly enhanced by the addition of nCuO due to the unique large surface-to-volume ratio and other properties of NPs. The SWE containing five common HMs, arsenic (As), chromium (Cr), cadmium (Cd), manganese (Mn) and lead (Pb), was filtered through a control ERC and nano-ERCs, and the concentrations of these HMs were determined with ICP-MS in the SWE and the filtrates. Results showed the nano-ERCs could significantly remove these HMs from the SWE compared to the control ERC, due to the enhanced adsorption capacity by nCuO. The relative weighted average removal percentage (RWAR%) was in the range of 53.05–71.83% for nano-ERCs and 39.27–61.65% for control ERC. Except for Cr, concentrations of these HMs in the treated wastewater effluent met the requirements for crop irrigation or scenic water supplementation; the Cr may be removed by multiple filtrations. In conclusion, nano-ERC can serve as a cost-effective approach for the further treatment of wastewater effluent and may be applied more widely in wastewater treatment to help relieve water stress.

Keywords: adsorption; heavy metal pollution; nano ecological; recycled aggregates; water stress

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1. Introduction

Water stress has caused serious risk to the environment and human health due to improper use, environment pollution and climate change [1,2]. According to the World Health Organization, one in three people worldwide still do not have access to safe drinking water [3]. The large amount of water for irrigation has caused a severe water deficit, while water for irrigation is still insufficient or unsafe to secure the production and quality of crops [4]. Polluted water used for irrigation can also contaminate soil and result in long-term adverse effects to the ecosystem and humans [5]. Heavy metal (HM) pollution in the environment has become a big concern because HMs are non-degradable and potentially toxic to the ecosystem and humans [6]. In particular, soil pollution in rice paddies caused by HM-polluted water irrigation is more prominent since rice plants need much more water than other crops through the life cycle; rice plants can also uptake more HMs than other crops, which has resulted in rice contamination and caused serious health concerns worldwide [7]. Although water resources have certain renewability because of the water cycle,

but it takes a long time to regenerate clean water, and the pollution caused by human activities has made the cycle even more complicated and difficult [8]. Thus, it is a must to reuse used water or treated wastewater. For example, in 2019, 75 billion cubic meters of urban wastewater was discharged in China, but less than 15% was reclaimed [9]. China has set goals to increase the amount of reclaimed wastewater to 35% in some regions [9]. To do this, water pollution control or treatment technologies are critical. Current wastewater treatment technologies include physical, chemical and biological processes, for example, flotation, precipitation, oxidation, solvent extraction, evaporation, carbon adsorption, ion exchange, membrane filtration, electrochemistry, biodegradation and phytoremediation [10]. Due to the complex nature of wastewater, especially industrial effluent, a combination of different methods is usually used to meet a specific standard of water quality. Heavy metal removal efficiency can be up to 99.99% by these technologies [11]. However, because of the original high concentrations of HMs and the cost of the technologies, it would be a big investment to improve the treated wastewater quality for direct reuse as irrigation or landscape water. In addition, wastewater treatment consumes about 3% of global electricity [12], which has caused a heavy financial burden to society, and the cost has experienced a steady exponential increase since 2010 [13]. Moreover, wastewater treatment contributes about 1.6% of greenhouse gas emissions [12,14], which is a big obstacle for carbon reduction and neutrality. Among all of the conventional and novel technologies, adsorption is one of the most efficient for removing HMs from wastewater [11]. This study focused on five common HMs, i.e., arsenic (As), cadmium (Cd), chromium (Cr), manganese (Mn) and lead (Pb), co-present in the simulated wastewater effluent (SWE). These HMs are commonly used in many industries, and they are all potential toxicants to the environment and humans [15]. The major source of As in the environment is mining industries due to its co-presence with copper (Cu), Pb and other metals [16]; Cd is commonly used in the battery industry due to its temperature adaptability, rechargeability, long life cycle and low maintenance [17]; Cr is usually discharged into the wastewater from leather, tanning and the textile industry [18]; 90% of Mn is used in the ferroalloy industry, and Pb is commonly used in steel works and others. Although in some stand-alone treatment plants of specific industries, one or more certain HMs may be dominant in the wastewater, a general wastewater treatment plant is a sink for various wastewaters containing multiple HMs, as well as other pollutants.

In addition to the abovementioned water issues, solid waste due to rapid urbanization has also drawn great attention. Several billion tons of construction and demolition waste (CDW) have been generated globally every year, with China as the world's largest producer, generating 2.36 billion tons of CDW per year [19,20]. Consequently, the overall recycling rate of CDW is not ideal, especially in developing countries; for example, in China, the recycling rate of CDW is less than 5% [20]. Despite the overall low recycling rate, the conversion efficiency from CDW to recycled aggregates can be very high. Except for reuse as building materials, recycled concrete demonstrates a porous structure with good adsorption performance [21], which can also be used in environment remediation for ecological purposes. For example, ecological concrete has been used to remove pollutants (e.g., HMs) from urban river, rainfall surface runoff, coastal sediment and constructed wetland [21–25]. To enhance the removal efficiencies of HMs, many types of adsorbent materials, such as fly ash, zeolite and nanoparticles, have been added to the porous structure of concrete [21]. In particular, nanotechnology has been used to manufacture concrete since the beginning of the millennium [26,27]; nano-concrete is defined as concrete inserted with Portland cement micro-particles that are less than 500 nano-meters [26]. Other nanoparticles (NPs), e.g., SiO_2 NPs [28–31], TiO_2 NPs [28], ZnO NPs [29] and Al_2O_3 NPs [30], have been increasingly used in concrete manufacture to enhance concrete's performance, such as to increase strength, decrease shrinkage, prolong durability and improve other mechanical properties [31–34]. However, the concept of "nano ecological recycled concrete (nano-ERC)" was firstly proposed and studied in our research.

In this study, a nano-ERC was manufactured with natural aggregates, recycled aggregates and fly ash, with the addition of copper oxide nanoparticles (nCuO). Recycled

aggregates were obtained from CDW [19]. nCuO was added to improve the adsorption capacity of HMs by the nano-ERC, since nCuO has shown good adsorption performance of HMs from the water system [35–38]. A unique process was adopted to manufacture the nano-ERC to enhance its properties related to ecological use. Specifically, part (25%) of the cement was replaced by ultra-fine fly ash to increase the pore structure and the connectivity of the concrete, thus improving the adsorption efficiency of the concrete. Moreover, ultra-fine fly ash can reduce the use of carbon-emitting cement, which is in accord with the concept of green development. The uniform particle size of recycled aggregates can also help form a good connection inside the concrete. In addition, the mixing process of this concrete was different from other concretes, to better connect the aggregates. Firstly, 50% of the total water amount was added and mixed with all of the aggregates for 30 s to wet their surface. Then the cement, ultra-fine fly ash, the rest of the water and water reducer were added to the wetted aggregate and mixed for another 60 s. After the concrete block was formed in a mold and naturally cured, nCuO was added to generate a nano-ERC. This research aims to study the adsorption capacity of nano-ERC for HMs from a simulated wastewater effluent (SWE) by nano-ERC with tickling filtration. The efficiency and mechanisms of HM removal by the nano-ERC are also discussed. Nano-ERC is presumed to present several advantages: (1) recycled concrete is cheap and easy to obtain access to, (2) NPs can increase the adsorption capacity of concrete for HMs in water, (3) nano-ERC results in many benefits for the sustainable development of society by helping solve the problems of water pollution treatment, water stress and solid waste.

2. Materials and Methods

Chemicals used in the study were purchased from Beijing InnoChem Science & Technology Co., Ltd., Shanghai Titan Scientific Co., Ltd. and Shanghai Hushi Laboratorial Equipment Co., Ltd. in China. Cement, natural aggregates and fly ash were purchased from No. 2 Engineering Company Ltd. of CCCC FIRST Harbor Engineering Company Ltd. in Qingdao, China. Recycled aggregates were obtained from Qingdao Ruijiatai New Environmental Protection Building Materials Co., Ltd., China. A SWE of 0.5 L in triplicate containing arsenic (As) 0.5 mg/L, cadmium (Cd) 0.1 mg/L, chromium (Cr) 1.5 mg/L, manganese (Mn) 2.0 mg/L and lead (Pb) 1.0 mg/L was made with $Na_2HAsO_4{\cdot}7H_2O$ (Sigma-Aldrich, St. Louis, MI, USA, ACS grade), $CdCl_2$ (Innochem, metalsbasis), $K_2Cr_2O_7$ (Hushi, GR), $MnCl_2{\cdot}4H_2O$ (Alfa, metals basis),and Pb $(NO_3)_2$ (Hushi, AR). The pH of the SWE was near 6.0. The concentrations of these HMs were prepared in accordance with the Comprehensive Wastewater Discharge Standard of China (GB8978-1996).

2.1. Procedure of Producing the Nano Ecological Recycled Concrete

A nCuO solution (100 mg/L) was prepared with nCuO powder (Alfa) in pure water and sonicated with a tip sonicator (JY99-IIDN) for 30 min. The other raw materials used for making the concrete are shown in Table 1 [39]. Half of the amount of the water and all of the aggregates were added to the mixer for 30 s to wet the surface of the aggregates. Then cement, ultra-fine fly ash, the rest of the water and water reducer were added and mixed for another 60 s [40]. The mixture was transferred into the cuboid mold (100 mm × 100 mm × 150 mm). A layer of cleaning surfactant was sprayed on the surface within 10–20 min after the concrete was molded. After the concrete block formed with a certain strength, the unsolidified cement slurry was rinsed off from the surface to increase the contact area between the solution and the concrete [41]. The concrete was cured under natural conditions for 28 days to make a control-ERC. To generate the nano-ERC, the cured concrete was then soaked completely in a nCuO solution (100 mg/L) for 48 h and taken out for natural air-drying for another 48 h. A duplicate of nano-ERCs was made in the same way. The procedural description for making ERC/nano-ERC is shown in Figure 1.

Table 1. Raw materials for producing concrete (per cubic meter).

Items	Values
Mass of water (kg)	146.2
2% liquid water reducer (kg)	16.0
Mass of cement (kg)	314.2
Mass of fly ash (kg)	104.8
Mass of recycled aggregates (kg)	785.1
Mass of natural aggregates (kg)	785.1
Scale of aggregate scale (mm)	5–20

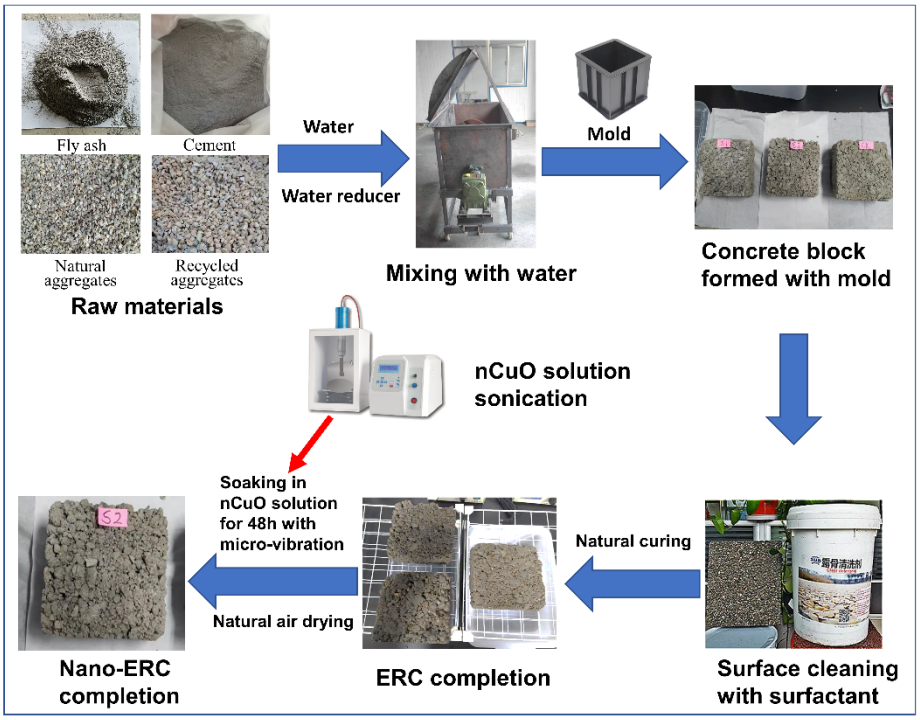

Figure 1. Procedural description of making ecological recycled concrete (ERC)/nano-ERC with recycled aggregates and copper oxide nanoparticles.

2.2. Characterization of the Nano Powder and the Recycled Concrete Aggregates

nCuO characterized by SEM was in the range of 20–60 nm. The hydrodynamic diameter of nCuO in a water suspension was, on average, 147.81 nm, with a polydispersity index (PDI) of 0.13 (Malvern nano-zetasizer). The weights and porosity of the nano-ERC were 1.98–2.77 kg and 26%–27%, respectively (Table 2).

Table 2. Physical parameters of ecological recycled concrete (ERC)/nano-ERC made with recycled aggregates and copper oxide nanoparticles.

Concrete Blocks	Weights (kg)	Porosity
Control ERC	2.77	27%
Nano-ERC 1	1.98	26%
Nano-ERC 2	2.52	26%

2.3. Procedure of Wastewater Effluent Treatment by the Nano-ERC

The concentrations of the five HMs in the three triplicate SWEs were measured with ICP-MS (NexION 1000G). The SWEs were then filtered through the three concretes (control-ERC, nano-ERC1 and nano-ERC2) simultaneously in a tickling filtration procedure (Figure 2). The experiment was an original design. The SWEs were transferred to a clean water bottle with a tap to control the speed of filtration. After all of the solutions were filtered through the concretes, the filtrates were collected, acidified and analyzed with ICP-MS (NexION 1000G) for the concentrations of the five HMs and Cu.

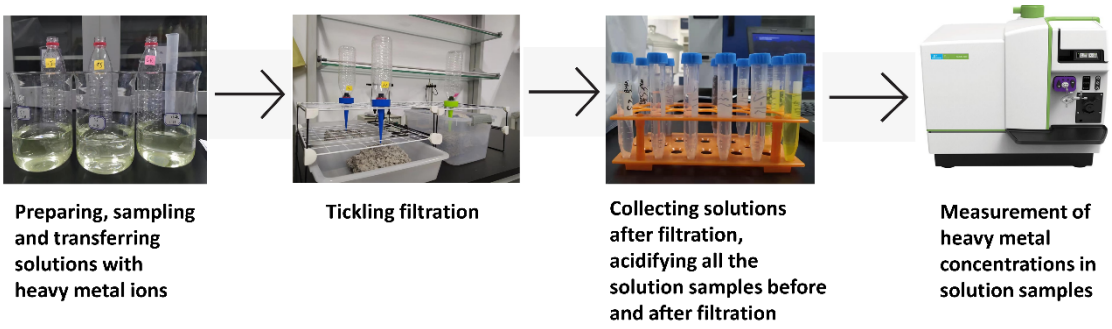

Figure 2. Procedural concept of the tickling filtration experiment with ERC/nano-ERC made with recycled aggregates and copper oxide nanoparticles to remove heavy metals from a simulated wastewater effluent.

2.4. Relative Weighted Average Removal Percentage of Heavy Metals

Considering the water loss by evaporation during the filtration process, a relative weighted average removal percentage (RWAR%) was used to compare the removal efficiency of HMs by the concretes.

$$\text{RWAR\%} = \frac{Conc._{bfi} - Conc._{afi}}{Conc._{bfi}} * \left(1 - \frac{m_i}{m_1 + m_2 + \ldots + m_j}\right) \quad (1)$$

where $Conc._{bfi}$ and $Conc._{afi}$ are HM concentrations before and after filtration through the concrete i; m_i is the mass (g) of concrete i.

2.5. Statistical Analysis

Generalized linear models (GLMs) were used to analyze the effects of heavy metal removal by the ERCs from the SWEs. Data distributions of HM concentrations were defined in the model. Models were validated and accepted with low residual heterogeneity, followed by an analysis of variance (ANOVA). Data were expressed as means ± S.E.M (standard error of measurement). Means were considered significantly different when $p < 0.05$ with the Tukey HSD test. All statistical analyses and figure constructs were performed in R (version 3.6.6) [42].

3. Results and Discussion

3.1. Heavy Metal Concentrations before and after Filtration by ERCs

Concentrations of the five HMs in the SWE were determined before filtration through the ERCs, which were, on average, As 472.31 µg/L, Cd 120.31 µg/L, Cr 1347.63 µg/L, Mn 2209.76 µg/L and Pb 5.34 µg/L, respectively (Table 3, Figure 3). In particular, the measured Pb concentration was much smaller than the nominal concentration, which was due to the chemical reactions between Pb^{2+} and the anions (e.g., Cl^-, $Cr_2O_5^{2-}$) resulting in precipitates. The measured concentrations of the rest of the HMs were approximate to their nominal concentrations in the SWE. After being filtered through the control-ERC and the nano-ERCs, the concentrations of the five HMs were all significantly reduced (Table 3, Figure 3). Except for Cr, the concentrations of the rest of the HMs in the fil-

trates through nano-ERCs met the Standard for Irrigation Water Quality (GB 5084-2021) and Environmental Quality Standards for Surface Water (GB 388-2002) in China (Table 3). Chromium is widely used in textile dyes and mordants, plating, pigments, alloying, water corrosion inhibition, etc., covering many walks of life [43]. It is very difficult to remove Cr from wastewater due to its rapid species transformation between multiple oxidation states varying from −2 to +6, with +3 and +6 as the two most stable states [44]. Notably, Cr in the water system can exist in 11 species include $Cr_2O_7^{2-}$, CrO_4^{2-}, H_2CrO_4, $HCrO_4^-$, Cr $(OH)_2^+$, Cr $(OH)_3$ (aq), Cr $(OH)_4^-$, Cr^{3+}, Cr_2 $(OH)_2^{4+}$, Cr_3 $(OH)_4^{5+}$ and $CrOH^{2+}$ [45]. Therefore, it is challenging to remove Cr from wastewater with a single technique. Up to date physicochemical technology, electrochemical technology and advanced oxidation technology, including membrane filtration, chemical precipitation, adsorption electrocoagulation, electrochemical reduction, photocatalysis, nanotechnology, etc., have been developed to remove Cr from wastewater [45]. The approach proposed by this study can be classified into physicochemical technology, and multiple filtrations or a supply line of filtration can be used to further remove Cr to ensure its concentration meets the abovementioned standards. The copper concentration in the solution before filtration was measured as 1.49 mg/L, which was close to the two standards, GB 5084-2021 and GB 388-2002. However, Cu concentrations in the filtrates were increased to 3.15–38.08 mg/L due to the dissolution of nCuO. Research is needed to stabilize nCuO and decrease its dissolution from the nano-ERC. For example, diethylene glycol (DEG) can be used as a capping agent to stabilize nCuO, as well as to further enhance the adsorption capacity of HMs by nCuO [35]. Geopolymers can be used as an adhesive agent between nCuO and the concrete, as well as increase the adsorption capability of nano-ERC due to its rigid skeleton structure [46]. In addition, the connection between nCuO and the ERC may be strengthened by adding the nCuO earlier together with the cement in the process of making nano-ERC, which needs to be further studied in our future research.

Table 3. Heavy metal (HM) concentrations in the solution before filtration and after filtration through control-ERC and nano-ERCs made with recycled aggregates and copper oxide nanoparticles.

Solution Samples	As (μg/L)	Cd (μg/L)	Cr (μg/L)	Mn (μg/L)	Pb (μg/L)
Solution before filtration	472.31 ± 25.30	120.31 ± 1.44	1347.63 ± 49.32	2209.76 ± 106.25	5.34 ± 2.09
Filtrates through control-ERC	12.15 ± 1.58 *	0.65 ± 0.01 *	492.53 ± 23.66 *	8.05 ± 0.12 *	0.56 ± 0.02 *
Filtrates through nano-ERC1	16.01 ± 0.02 *	1.53 ± 0.04 *	365.07 ± 2.64 *	29.19 ± 1.40 *	0.36 ± 0.18 *
Filtrates through nano-ERC2	0.44 ± 0.01 **	0.13 ± 0.00 **	85.35 ± 2.86 **	1.93 ± 0.33 **	0.28 ± 0.14 **
GB 5084-2021	50-100 [a]	10	100	none	200
GB 388-2002 (V class)	100	10	100	none	100

Note: * indicates significant different from the concentrations of heavy metals in solutions before filtration at the significance level of $\alpha = 0.05$. ** indicates significantly different from the concentrations of heavy metals in solutions before filtration at the significance level of $\alpha = 0.01$. [a] The standard concentrations of arsenic depend on the crops.

However, it is worth noting that water loss due to evaporation could not be ignored in the experimental procedure. By considering the wetted surface of the ERCs and the amount of water evaporation while doing the filtration experiment, the RWAR% was used to calculate the efficiency of HM removal by ERCs with Equation (1). The RWAR% of nano-ERCs was significantly higher than the control-ERC for all of the five HMs (Table 4, Figure 4). Specifically, the RWAR% were 8.31–16.58%, 6.08–16.70%, 35.09–55.92%, 5.92–16.44% and 11.77–22.42% higher for As, Cd, Cr, Mn and Pb by nano-ERCs than by

the control-ERC, respectively. These preliminary results proved that it is feasible to use nano-ERC to increase the removal of the five HMs in the SWE. As two major HM pollutants in the environment, Cr and Pb showed the highest removal percentages by nano-ERC, which lays a critical foundation for the practical application of nano-ERC for environmental remediation. Further research is needed to precisely estimate the amount of HM removal by the ERCs through the improvement of the experimental design to reduce water loss, as well as to ensure the uniformity of the performance of the same type of ERCs.

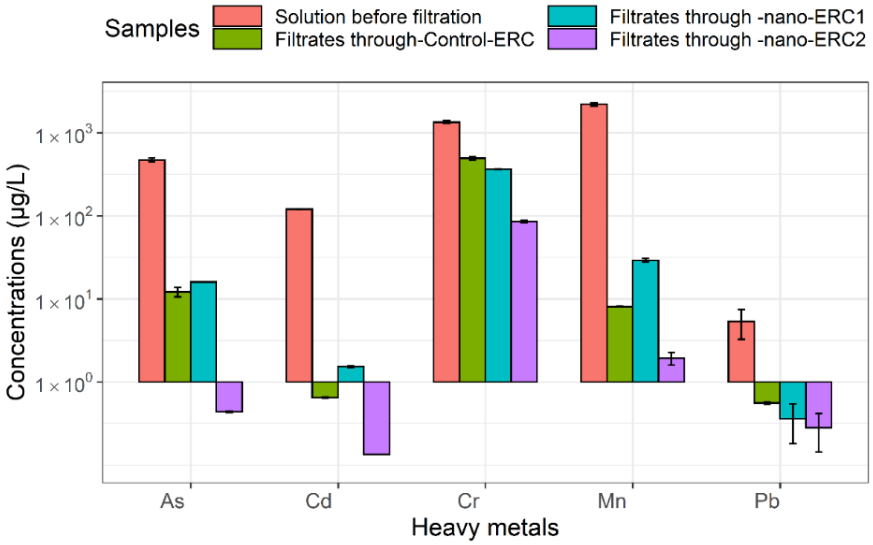

Figure 3. Heavy metal (HM) concentrations before and after filtration through control-ERC/ nano-ERC made with recycled aggregates and copper oxide nanoparticles (concentrations in the y axis were log-transformed in order to make the difference between treatments more visible).

Table 4. Weighted average removal percentage of HMs by control ERC and nano-ERCs made with recycled aggregates and copper oxide nanoparticles.

Sample	As	Cd	Cr	Mn	Pb
Control-ERC	60.29	61.55	39.27	61.66	55.42
Nano-ERC1	70.28	71.83	53.05	71.79	67.85
Nano-ERC2	65.30	65.29	61.22	65.31	61.94

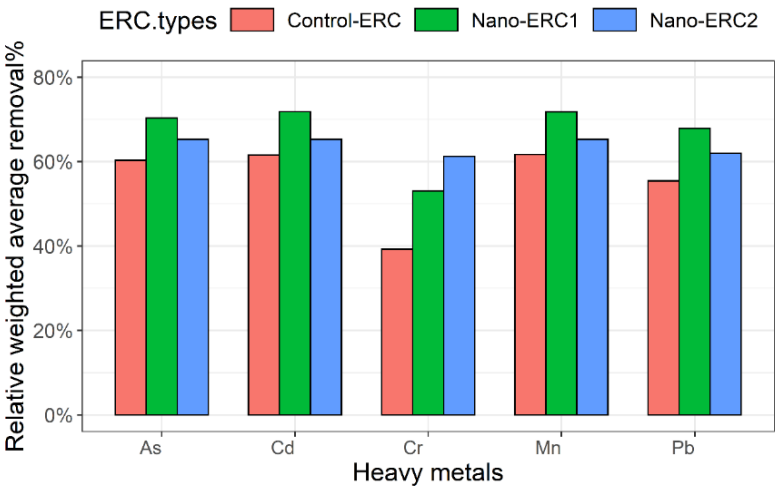

Figure 4. Relative weighted average removal percentage of HMs by control ERC and nano-ERCs made with recycled aggregates and copper oxide nanoparticles.

3.2. Mechanisms of Heavy Metal Removal by Nano-ERC

Concrete itself exhibits good adsorption capability for pollutants from the environment due to its porous structure [19,21]. The addition of nCuO, with good adsorption performance, enhanced the removal efficiencies for HMs such as Pb, nickel (Ni) and Cd, due to its large surface-to-volume ratio [36,38]. The procedural mechanism behind nano-ERC adsorbing HMs is shown in Figure 5. nCuO adhered to the porous surface of ERC during the soaking and drying process and increased the adsorption sites of HMs in the ERC. When the HM-containing solution detoured through the porous paths in the nano-ERC, the HM ions were bound to the adsorption sites of the ERC and nCuO. Consequently, the HM concentrations in the filtrates significantly decreased. However, components in the SWE were relatively simple, while much more complicated components can be co-present and interact in actual wastewater effluents, e.g., nutrients, organic matter, microorganisms, etc. [12]. In particular, organic matter can interact with NPs and form a corona on the NPs [47], which may further improve the adsorption capacity of nano-ERC for HMs. To increase the effect size of HM removal by the nano-ERC, several approaches could be investigated: (1) use capping and adhesive agents to stabilize nCuO and its adhesion to the concrete, (2) improve the porous structure of the concrete and optimize the amount of nCuO adhered in the nano-ERC and (3) adjust the initial pH of the wastewater effluent to ionize the HMs and decrease the precipitation. In addition to HMs being removed from the wastewater, HMs adsorbed by nano-ERC are easier to collect and to be handled together. Nano-ERCs can also be recycled when reaching their adsorption capacity after a certain period of time.

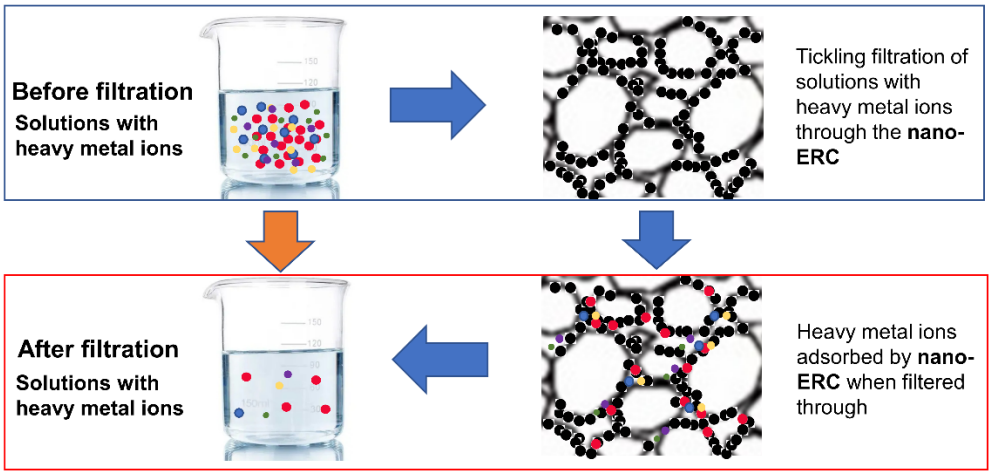

Figure 5. The procedural mechanisms of nano-ERC made with recycled aggregates and copper oxide nanoparticles adsorbing heavy metals (colorful dots in the solution are different heavy metal ions; black dots in the pore of concrete are nCuO).

4. Conclusions

Increasing water demand and declining water quality is forcing us to reuse wastewater. However, the primary obstacles are the high cost of wastewater treatment and the greenhouse gas emissions during the treatment process. This study presents the first trial of manufacturing nano ecological recycled concrete (ERC) with nCuO and ERC and using it to remove five HMs, i.e., As, Mn, Cr, Cd and Pb, from a simulated wastewater effluent (SWE). Nano-ERC has a promising future based on its cost-effective properties of removing HMs and the benefit for reducing the heavy burden of discarded concrete waste due to the rapid urbanization. The approach of using nano-ERC proposed by this present study can serve as a post-treatment method after traditional wastewater treatment or be used in the line of water reuse for irrigation or landscapes. Although the removal efficiencies of these metals by nano-ERC were promising, there are still many issues that need to be addressed,

as reflected in the trial: (1) standardization of the size and physicochemical properties of the ERCs is necessary to optimize their performance, (2) actual wastewater should be used to verify the adsorption performance of the control-ERC and the nano-ERC, (3) the filtration process should be conducted in a relative closed condition to reduce the water loss through evaporation, (4) it is critical to investigate the average mass of nCuO adhered to the mass of ERC and find out the method of determining the optimal ratio of the two components with the best performance of HM removal and (5) the removal capacity of other pollutants by nano-ERC remains to be investigated. In addition to using the treated wastewater directly, nano-ERC may be used in the inlet and outlet of irrigation water (e.g., for paddies) to further remove pollutants in the irrigation water and drainage and, eventually, to reduce the adverse effects to the crops and the surrounding ecosystem, as well as human health. Moreover, nano-ERC may be used for the pre-treatment of wastewater as building materials for wastewater transport pipelines, of which the efficiency needs to be explored. On the whole, three benefits of nano-ERC would be achieved: (1) reducing the burden of wastewater treatment, (2) relieving the stress of dealing with solid waste (i.e., CDW) to the environment and (3) providing a safe irrigation or scenic water supplementation resource. Complementing traditional wastewater treatment technology, nano-ERC may also help to achieve the goal of carbon neutrality by 2050, which is the most urgent mission for the world.

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