

Comparing Conventional and Advanced Approaches for Heavy Metal Removal in Wastewater Treatment: An In-Depth Review Emphasizing Filter-Based Strategies

Jana Ayach, Wassim El Malti, Luminita Duma, Jacques Lalevée, Mohamad

Al Ajami, Hussein Hamad, Akram Hijazi

▶ To cite this version:

Jana Ayach, Wassim El Malti, Luminita Duma, Jacques Lalevée, Mohamad Al Ajami, et al.. Comparing Conventional and Advanced Approaches for Heavy Metal Removal in Wastewater Treatment: An In-Depth Review Emphasizing Filter-Based Strategies. Polymers, 2024, 16 (14), pp.1959. 10.3390/polym16141959. hal-04753607

HAL Id: hal-04753607 https://hal.science/hal-04753607v1

Submitted on 12 Nov 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Comparing Conventional and Advanced Approaches for Heavy Metal Removal in Wastewater Treatment: An In-Depth Review, Emphasizing Filter-Based Strategies

Jana Ayach ^{1,2}, Wassim El Malti ^{3,*}, Luminita Duma ², Jacques Lalevee ^{4*}, Mohamad Al Ajami ¹, Hussein Hamad ¹ and Akram Hijazi ¹

¹Research Platform for Environmental Science (PRASE), Doctoral School of Science and Technology, Lebanese University, Beyrouth, Lebanon;akram.hijazi@ul.edu.lb; hussein.hamad@ul.edu.lb; jana.ayach.1@st.ul.edu.lb; mohamad.r.alajami@hotmail.com

²University of Reims Champagne-Ardenne, CNRS, ICMR UMR 7312, Reims, France; luminita.duma@univ-reims.fr

³College of Health Sciences, American University of the Middle East, Kuwait; wassim.elmalti@aum.edu.kw ⁴CNRS, IS2M UMR 7361, University of Haute-Alsace, Mulhouse, France; Jacque.lalevee@uha.fr

Abstract: Various industrial activities release heavy metal ions into the environment, which repre-14 sent one of the major toxic pollutants owing to their severe effects on the environment, humans, and 15 all living species. Despite several technological advances and breakthroughs, wastewater treatment 16 remains a critical global issue. Traditional techniques are dedicated to extracting heavy metal ions 17 from diverse wastewater origins, encompassing coagulation/flocculation, precipitation, flotation, 18 and ion exchange. Their cost, side toxicity, or ineffectiveness often limit their large-scale use. Due to 19 their adaptable design, simple operation, and reasonable cost, membrane filtration and adsorption 20 have proven their efficiency in removing metals from wastewater. Recently, adsorption-based filters 21 have appeared promising in treating water. Within this range, filters incorporating natural or hybrid 22 adsorbents present an appealing alternative to conventional approaches. This review aims to list 23 and describe the conventional and advanced wastewater treatment methods by comparing their 24 efficiency, cost, and environmental impact. Adsorption-based filters were highlighted due to the 25 significant advantages they can provide. 26

Keywords. Clean Water; Filters; Heavy Metals; SDG6; Wastewater

1. Introduction

Over time, environmental pollution has witnessed a persistent rise, impacting both 30 human existence and the broader ecosystem. Water pollution is growing mainly due to 31 wastewater from various industries dealing with paper and pulp, textiles, chemicals, etc 32 [1]. As of July 2022, a report jointly published by the World Health Organization and the 33 United Nations International Children's Emergency Fund (UNICEF) revealed that ap-34 proximately 2.2 billion individuals remain without access to clean and safe drinking water 35 [2]. Additionally, around 653 million people lack hand-washing facilities in their homes. 36 The makeup of pollutants found in water effluents is contingent upon the industries in 37 the vicinity responsible for generating the wastewater [3]. These effluents often contain 38 harmful substances such as pesticides, dyes, aromatic hydrocarbons, oils, heavy metals, 39 and more, which, when released into the environment, pose detrimental consequences for 40 both humans and wildlife [4–6]. In addition, the continuous industrialization in develop-41 ing countries significantly increased water demand [7]. Among several alternatives to pre-42 serve water resources, developing efficient and sustainable wastewater treatment technol-43 ogies can provide a convenient way to reuse it [1]. 44

29

27 28

1

2

3

4

5

6

7

8

9

10

11

12

Efficient wastewater treatment, encompassing the removal of heavy metals and their 45 responsible utilization, plays a vital role in attaining the United Nations Sustainable De-46 velopment Goal 6 (SDG6) of universal access to drinking water, sanitation, and hygiene 47 by 2030 [8]. Wastewater treatment practices vary from one nation to another, exhibiting 48 significant distinctions, particularly between developed and developing countries. While 49 advanced countries mainly use centralized treatment, many developing countries use de-50 centralized or no wastewater treatment. Although centralized treatment has some ad-51 vantages, it may not be sustainable in the long term because of its high environmental 52 impact related to resource consumption. In contrast, built-up treatment plants can be a 53 sustainable solution for all countries, regardless of their development level. Most im-54 portantly, constructed washes may be a sustainable wastewater treatment for developed 55 and emerging countries [9]. 56

The pollutants generated in the water effluents are usually classified as organic and 57 inorganic contaminants, depending on the types of molecules and their toxicity levels. 58 Heavy metals are highly toxic inorganic pollutants, non-biodegradable, and their accu-59 mulation in wastewater yields a weighty environmental burden [10]. These heavy metals 60 come from multiple industrial activities such as tannery and battery manufacturers, elec-61 troplating, mining, pesticides, and paint industries [11]. Hence, they stand out as the most 62 enduring contaminants within wastewater, and their elevated toxicity can potentially im-63 pact critical bodily functions, including blood composition, brain activity, respiratory sys-64 tem, kidneys, liver, and other vital organs [12]. As a result, extended exposure may lead 65 to the gradual onset of physical, muscular, and neurological degenerative conditions that 66 exhibit similarities to diseases such as Alzheimer's, Parkinson's, muscular dystrophy, and 67 multiple sclerosis [9,10]. As a result, heavy metals significantly harm human health, 68 aquatic ecosystems, and the surrounding environment. Thus, it is becoming crucial to 69 simultaneously reduce these numerous pollution sources, develop effective techniques for 70 eliminating such dangerous pollutants from wastewater, and recycle them for other uses. 71

Several conventional methods, including coagulation, osmosis, ion exchange, and 72 precipitation, are employed to decrease the elevated concentration of various metal ions 73 in wastewater to meet the required regulatory levels [13]. However, many factors often 74 limit these methods, such as cost and toxic environmental side effects [14]. Other relatively 75 advanced techniques, such as adsorption and membrane filtration, have become excellent 76 alternative treatments with improved removal effectiveness and reduced costs and envi-77 ronmental impact [15]. Indeed, the effective removal of heavy metal ions from wastewater 78 necessitates the consideration of several critical factors, including their concentration, spe-79 cific metal types, pH levels, chemical additives, removal efficiency, environmental conse-80 quences, and cost implications. In industrial settings, adsorption is widely acknowledged 81 as one of the most efficient methods for wastewater treatment, effectively reducing the 82 presence of harmful inorganic and organic pollutants in the treated wastewater released 83 as effluents [16]. Heavy metal ions' adsorption results from physicochemical interactions, 84 primarily involving ion exchange or the formation of complexes between the metal ions 85 and the functional groups found on the surface of the adsorbent. Furthermore, cost-effec-86 tive adsorbents derived from natural biomass materials, industrial solid waste, agricul-87 tural byproducts, and biosorbents as source materials emerge as promising alternatives 88 [16,17]. They exhibit enhanced adsorption capabilities after subjecting raw biomass adsor-89 bents to various physical or chemical treatments [18-20]. Nevertheless, natural, synthetic, 90 and hybrid filters (a mix of natural and synthetic) show high adsorption capacity as they 91 efficiently adsorb heavy metals and dye pollutants in aqueous solutions. 92

In this review, we have compiled and compared examples of the most common conventional and relatively advanced wastewater treatment methods used to remove heavy metals. We have described the most relevant features of the removal methods and discussed their performance with respect to the cost, environmental impact, and removal efficiency. In addition, we have focused on advanced techniques, especially membrane 97 filtration and adsorption, and highlighted the advantages of the different filter types and 98 applications. 99

2. Heavy metals: sources and toxicity

Heavy metals in surface water systems can have natural origins or result from human activities. Geological or natural sources encompass volcanic eruptions, the weathering of 102 rocks containing metals, sea salt spray, wildfires, and natural weathering processes that 103 can initiate the release of metals into different environments. Heavy metals can take vari-104 ous forms, including hydroxides, oxides, sulfides, sulfates, phosphates, silicates, and or-105 ganic compounds [21,22]. Furthermore, various industrial manufacturing processes dis-106 charge heavy metals, including Cr, Cd, Ni, Cu, Zn, As, and Pb, at varying toxicity levels. 107 These metals exhibit high water solubility and can be absorbed by aquatic organisms, 108 leading to their accumulation in the human body via the food chain. Exceeding the re-109 quired concentrations of heavy metals when ingested can result in various health disor-110 ders [23]. Heavy metal contamination poses environmental risks due to its carcinogenic, 111 teratogenic, and endocrine-disrupting effects, which can significantly impact children's 112 behavior [24]. 113

Table 1 summarizes the major sources and effects of common heavy metals, exclud-114 ing cadmium, copper, and lead. These latter particular metals are discussed in detail in 115 the following due to their prevalence in water contamination and the significant attention 116 they have received over the decades. 117

Cadmium (Cd), a profoundly toxic metal, is primarily produced as a byproduct of 118 zinc manufacturing. It becomes stored in plants and subsequently ingested by microor-119 ganisms and humans. Exceeding the threshold level (7µg/L) of cadmium can lead to harm-120 ful effects. The metal shows a high transfer rate from soil to plants, resulting in elevated 121 cadmium levels in fruits and vegetables. As a non-essential toxic metal, it significantly 122 disrupts cellular enzymatic systems, leading to oxidative stress and nutrient deficiencies 123 in plants. Additionally, cadmium is associated with hepatotoxicity, causing liver disease, 124 by binding to cysteine-rich proteins like metallothionein. Furthermore, cadmium has an 125 affinity for binding with ligands like glutamate, histamine, and aspartate, potentially 126 causing iron deficiency [25]. 127

Copper (Cu) exists in the environment due to natural and human activities. Anthro-128 pogenic sources of copper include mining, metal and electric production, fungicide use, 129 leather processing, and vehicle brake pads. Also, natural copper contamination sources 130 can arise from forest fires, volcanic eruptions, and windblown dust [26]. Copper is in-131 volved in many enzymatic systems in the organism but becomes toxic at high intake lev-132 els, exceeding 2000 µg/L [27]. Copper is crucial for nutrition and plays a vital role in brain 133 function, but its presence is also associated with the development of Alzheimer's disease. 134 Even small concentrations of copper in drinking water can notably impact individuals' 135 learning and memory [28]. Moreover, prolonged exposure to copper primarily affects the 136 liver, the first site where copper accumulates upon entering the bloodstream. Copper tox-137 icity typically manifests as the development of cirrhosis, accompanied by episodes of he-138 molysis and damage to renal tubules, as well as affecting the brain and other organs. Pro-139 longed and excessive exposure to copper can lead to hepatic necrosis, vascular collapse, 140 coma, and ultimately, fatality [29]. 141

Lead (Pb) primarily enters the environment through various sources such as lead 142 paint, house dust, food, air, soil, pottery, solder, porcelain, and tin. It accumulates in the 143 body and can lead to disruptions in the number of red blood cells and kidney and brain 144 diseases. It poses a significant hazard to young children and pregnant women, affecting 145 physical development and normal mental growth in adults. Drinking water contaminated 146 with lead (threshold of Pb: 25µg/L) harms human health, especially for children consum-147 ing fruit juice and foods prepared with water containing lead [30-32]. 148

149 150

100

| Heavy metal | Major source(s) | Common effects | Reference |
|---------------|-----------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------|-----------|
| Arsenic (As) | Insecticides and pesticides, treated wooden pulps, and smelting metals. | Cancer, skin color changes, Blackfoot disease, diabetes, and others. | [31,32] |
| Zinc (Zn) | Plating, galvanizing, other metal finishing processes, paper and pulp industries, battery industry. | Kidney failure, lung fibrosis and cancer, anemia, vomiting, and others. | [33] |
| Chromium (Cr) | Electroplating, leather tanning, and textile industries. | Dermatitis, kidney and gastric damage, lung cancer, respiratory tract and eyes irritation, and others. | [34,35] |
| Mercury (Hg) | Oil refinery processes, pesticides, and burning coal for power generation | Brain, central nervous system, heart, alimentary tract, kidney, and liver significant damages. | [36] |
| Nickel (Ni) | Electroplating, printing and dyeing, and pharmaceutical and metallurgical industries | Skin irritation, asthma, conjunctivitis, and cancer | [37,38] |
| Bismuth (Bi) | Chemical and pharmaceutical industries | Hypotension, insanity, and renal failure | [39] |

Table 1. Common heavy metal ions in wastewater: sources and effects.

3. Elimination of heavy metals from wastewater: conventional methods

A variety of traditional methods are employed for the extraction of heavy metals from wastewater. These techniques include coagulation and flocculation, chemical precipitation, ion exchange, flotation, electrochemical separation, and evaporation [40]. In the following, we describe the most common and significant conventional practices for wastewater treatment, along with their advantages and disadvantages. 157

3.1. Coagulation and flocculation

Coagulation refers to destabilizing colloids by neutralizing their repulsive forces, 159 while flocculation involves aggregating unstable particles [41]. Traditional coagulants 160 such as aluminum, ferrous sulfate, and ferric chloride are employed to counteract ionic 161 charges. On the other hand, flocculation entails the amalgamation of particles to form 162 larger coagulates. These larger coagulates can be generated using materials like polyalu-163 minum chloride, polyferric sulfate, polyacrylamide, and various polymer flocculants [42]. 164 The coagulation-flocculation process demonstrated both effective performance and 165 straightforwardness in removing heavy metals from wastewater [43]. According to previ-166 ous works, polyethylene-glycol is one of the most practical flocculants [44]. This approach 167 can efficiently eliminate a range of heavy metals, including Cu, Pb, Ni, As, Se, Cr, Sb, and 168 Ag [45]. The removal efficiency for the mentioned heavy metal ions varied between 95% 169

152

and 99%. However, there are several drawbacks associated with the application of floccu-170 lation that can be pinpointed. These include the health hazards posed by inorganic coag-171 ulants, the generation of significant amounts of sludge, limited selectivity for specific met-172 als and inefficiency in addressing emerging pollutants, the potential for increased sewage 173 colors, lower effectiveness when employing natural coagulants, and challenges in scaling 174 up the process [46]. 175

3.2. Precipitation

The chemical precipitation method, also known as coagulation precipitation, is ex-177 tensively utilized in various industries and is regarded as one of the most efficient and well-established techniques. This method involves transforming dissolved metal ions into 179 solid particles, facilitating precipitation (Fig. 1). The coagulant precipitates metal ions in 180many ways, such as changing pH, electro-oxidation potential, or co-precipitation, fol-181 lowed by removing deposits. 182



Figure 1. The precipitation technique for extracting heavy metals from wastewater.

Hydroxide precipitation is commonly utilized due to its cost-effectiveness, simplic-185 ity, and the ability to adjust pH as needed [46]. This is achieved by adding hydroxide to 186 the agitated wastewater, forming insoluble metal hydroxide precipitates. For example, 187 metal ions can react with calcium hydroxide (lime), producing metal hydroxide precipi-188 tates and calcium ions, as illustrated in Eq. 1. 189

$$M^{n+} + Ca(OH)_2 \quad \leftrightarrows \quad M(OH)_{n \ (s)} + Ca^{2+}$$

$$(1) \quad 190$$

In addition, sulfides are widely used to precipitate the metals polluting the 191 wastewater (Eq. 2). 192

$$M^{n+} + S^{2-} \hookrightarrow M_n S_{(S)}$$
(2) 193

Despite its relatively high efficiency, about 95-99%, for most heavy metals present in 194 wastewater, the precipitation method suffers various drawbacks [47]. High pH depend-195 ence and difficulty in removing large sludge volumes from the water are among the most 196 critical limitations [48]. 197

3.3. Ion exchange

The reversible chemical response used to transform unwanted metallic ions into less 199 environmentally harmful ones makes the ion exchange treatment an alternative to the pre-200 viously described methods [49]. A heavy metallic ion is eliminated from the wastewater 201 by bonding to a motionless solid particle as an alternative to the solid particle cation. The 202 ion-alternate technique can put off-target (a few or all) heavy metallic ions, including Pb2+, 203

176

178

184

183

Hg²⁺, Cd²⁺, Ni²⁺, V⁴⁺, V⁵⁺, Cr³⁺, Cr⁴⁺, Cu²⁺, and Zn²⁺ from wastewater. The ion exchange oc-204 curs when the alternate cation of the ion exchanger exchanges the wastewater pollutant 205 cation, as shown in Eq. 3, where M⁻EC⁺ stays for the ion exchanger, EC⁺ the exchange cat-206 ion, WC⁺ the wastewater cation, and M⁻ the stable ion. Na⁺ and H⁺ are usually used as 207 alternate cations [50]. 208

 $M^{-}EC^{+} + WC^{+}$ \Rightarrow M⁻WC⁺ + EC⁺ (3)209

Various types of resins were investigated for cation removal and described in the lit-210 erature, such as Amberlite IRC86 chelating exchange resin [51] and Diaion weak cationic 211 resin CR11 [52]. Both examined resins demonstrated an efficiency of approximately 90% 212 in the removal of trivalent chromium from aqueous solutions [52]. In addition, zeolite ex-213 hibits an outstanding ion exchange capacity of up to 98% due to its inherent negative 214 charge. This negative charge arises from Si⁴⁺ in the center of the tetrahedron, which un-215 dergoes isomorphous replacement with Al³⁺ cations. Furthermore, metal-organic frame-216 works (MOFs) have been extensively studied and proposed as highly promising materials 217 for the ion exchange removal process [53]. Some reported MOFs were used for Cd²⁺, Pb²⁺, 218 and Hg2+ removal with a capacity of 97%, 95%, and 93%, respectively [54], ZIF-8 elimi-219 nated 95% of Cu²⁺[55], and ZIF-67 eliminated 98% of Cr⁶⁺ [56]. Although the ion exchange 220 method has shown promising results, further research is needed to investigate its stability 221 and reusability [44]. 222

3.4. Flotation

Flotation is a gravity separation typically implemented to separate minerals, such as 224 sulfides. This procedure holds substantial importance in the economies of all developed 225 nations [57,58]. Nowadays, it is used extensively for metal ion recovery from industrial 226 wastewater and is applied to a wide range of heavy metal waste streams [59,60]. Several 227 types of flotation separation were discussed in the literature, such as dissolved-air flota-228 tion and ion flotation, which are considered the most significant [61]. 229

The dissolved air flotation consists of applying gas bubbles to the body of 230 wastewater, which will selectively transfer the metal ions to the water's surface, forming 231 a slug that can be easily eliminated. This process helped remove heavy metals, such as Cu, 232 Cd, Fe, Ni, Mo, Mn, and As, from wastewater with good efficiencies [61]. 233

On the other hand, ion flotation was implemented to eliminate Cr³⁺, Pb²⁺, and Cu²⁺ 234 from wastewater in the presence of different surfactants. Removal effectiveness was 235 achieved at 95% for the mentioned metal ions [62]. 236

Despite the simplicity, high selectivity, and high removal efficiencies obtained in flo-237 tation, the process is constrained by the removal rate and ineffectiveness when applied to large dosage amounts of metals in water. In addition, if biosurfactants are not used, the 239 process is constrained by high toxicity [62].

3.5. Electrochemical separation

Electrochemical techniques have recently gained substantial attention and recogni-242 tion as a dependable method for treating industrial wastewater. In contrast to alternative 243 systems, electrochemical systems offer numerous advantages, such as operation under 244 ambient pressure and temperature conditions, dependable performance, and the ability 245 to accommodate fluctuations in influent flow rate and composition. This process is known 246 for its ability to degrade various contaminants, including heavy metals. The fundamental 247 aspect of the electrochemical reaction involves the flow of electric current through an 248 aqueous solution containing metals, a cathode plate, and an insoluble anode (Fig. 2) [63]. 249

Heavy metals are managed through precipitation as hydroxides in electrolytes with 250 a neutral or slightly acidic pH. In this way, the choice of electrode material plays a signif-251 icant part in increasing the capabilities for various types of pollutants. Additionally, the 252 number of ions created by the electrical current will determine the quality of the treated 253

223

238

240

effluent. Therefore, any factor impacting the charge in the process will also affect the pro-254 cess' overall efficiency [63]. 255

Many metal ions, such as Hg2+, Cd2+, Pb2+, and Cu2+, were successfully removed with 256 excellent efficiency using this process. To make this type of treatment viable for industrial 257 applications, it is crucial to address the issue of energy consumption [64]. 258



Figure 2. Electrochemical separation technique for wastewater treatment.

4. Elimination of heavy metals from wastewater: Advanced methods

Over time, the adoption of advanced technologies for removing heavy metals from wastewater has seen substantial growth. In the subsequent sections, we outline and explore some of the more prevalent advanced techniques, such as membrane filtration and 264 adsorption, which include filtration systems. These methods exhibit numerous notable 265 advantages in comparison to traditional approaches. 266

4.1. Membrane filtration

Among the array of wastewater treatment methodologies, membrane filtration is fre-268 quently employed as a separation technique within water purification systems. Mem-269 brane filters can prevent the formation and spread of bacteria and viruses but also remove 270particles like total suspended solids, turbidity, and sediment from water. 271

The attractive nature of membrane filtration to industry and central municipal 272 wastewater recycling lies in its capability to remove suspended solids, organic com-273 pounds, and inorganic contaminants, including heavy metals. The pressure-driven mem-274 brane filtration process is primarily governed by the pressure difference between the feed 275 and permeate sides (Fig. 3) [65]. Various membrane filtration methods can be applied for 276 the extraction of heavy metals from wastewater, depending on the desired particle size 277 retention [15], as illustrated in the filtration spectrum adapted from [66] (Fig. 4). 278

260 261

259

262 263



Figure 4. Filtration spectrum (Source: adapted from [66]).

Reverse osmosis (RO) is a separation process driven by pressure, employing a semi-283 permeable membrane with a pore size of 0.5-1.5 nm. This membrane explicitly allows the 284 passage of molecules smaller than the size of its pores (Fig 5 (a)). The RO process works 285 by exerting pressure values (ranging from 20 to 70 bar) higher than the feed solution's 286 osmotic pressure. This effectively reverses the typical osmotic process. The solute mole-287 cules within the solution generally have a molecular size ranging from 0.25 to 3 nm [67]. 288 The filtration yield was estimated at 95-99 % inorganic salts and charged organic matter 289 removal. In the RO separation process, heavy metal ions present in electroplating 290 wastewater, such as Ni2+, Cr6+, and Cu2+, are effectively removed with an efficiency surpas-291 sing 98.75% [68]. In recent times, the method has been utilized for the purification of in-292 dustrial wastewater derived from mining activities in coaster-field, Victoria-Australia, 293 with averaged extraction efficiency values dependent on the extracted metal: Fe^{3+} (10 %), 294 Zn²⁺ (48 %), As³⁺ (66 %), Ni²⁺ (82 %), and Sb³⁺ (95 %) [44]. RO is a compact method with a 295 high removal rate. Nonetheless, the exerted pressure can result in membrane fouling and 296 deterioration as contaminants accumulate on the surface of the membrane [69]. 297

Conversely, the forward osmosis (FO) procedure entails employing a semi-permea-298 ble membrane to partition the feed solution from the draw solution (Fig. 5 (b)). This ap-299 proach balances selectivity and the flow rate of permeate water [70]. Typically, the draw 300 solution has a higher osmotic pressure than the feed solution. This difference in osmotic 301 pressure induces water movement from the feed solution to the draw solution, leaving 302 behind undesired solutes in the feed solution while treating the water in the draw solution 303 [71]. This method is eco-friendly and efficient, as it does not necessitate hydraulic pressure 304 for the FO process. The devices based on FO are easy to clean, have a low fouling rate, and 305 are widely used in wastewater treatment [63,64], with extraction efficiency reports for Pb, 306 Cr, As, Cd, and Cu as high as 99.5%. In other studies, 98-99.96% removal efficiency was 307

313

achieved in removing Co²⁺, Cu²⁺, and Zn²⁺ from acid mine drainage [72,73]. Despite its 308 relatively effective removal capabilities, FO technology has its limitations. The choices for 309 FO membranes are restricted [74], and the desired solute often needs to be regenerated through methods like membrane distillation [75,76], which can also entail increased energy consumption [72]. 312



Figure 5. Reverse osmosis (a) and Forward osmosis (b) membrane separation methodologies. 314

In contrast, in ultrafiltration (UF), both water and low molecular weight solutes per-315 meate through the membrane, while larger macromolecules exceeding the membrane's 316 pore size are retained. UF typically possesses a pore size range of 0.001-0.1 µm, offering 317 effective removal capabilities for wastewater treatment. Conversely, microfiltration (MF) 318 normally operates with pore sizes that are up to an order of magnitude larger. While 319 wastewater treatment often employs relatively open MF membranes with a pore size of 320 $0.1-1.0 \ \mu$ m, the current trend in water treatment favors using moderately tight MF mem-321 branes with pore sizes ranging from approximately 0.09 to 0.1 μ m [65]. The membrane 322 filtration techniques applied to industrial wastewater can give effluent for disposal or 323 feedstock for reusing [65]. UF membrane technology has been successfully used to elimi-324 nate heavy metals, such as iron, copper, and chromium, with a capacity ranging from 96 325 to 99% [77]. MF is usually used in municipal water treatment applications due to its re-326 moval capacity of viruses and, to some extent, its depth filter function. It also provides a 327 barrier against bacterial and protozoan parasites [65]. 328

Various other membrane types have been documented in the literature, including 329 membranes composed of synthesized metal oxide/polyethersulfone (PES), Al₂O₃/PES, and 330 ZrO₂/PES. These membranes were employed for the removal of metals from wastewater. 331 They were crafted by incorporating PES, Al₂O₃, and ZrO₂ nanoparticles, with particle sizes 332 ranging from 40-50 nm and surface areas measuring between 20-34 m²/g. The membrane 333 fabrication process involved the utilization of the phase inversion method. To prepare the 334 5% w/w metal oxides/PES membranes, the metal oxide nanoparticles were dispersed in a 335 solution of N-methyl pyrrolidone. The effectiveness of these membranes in removing Cr 336 and Pb from both synthetic and natural wastewater was tested. The results showed that 337 the membranes achieved approximately 99% removal for Pb and 88% for Cr [70]. 338

Electrodialysis (ED) is a membrane-based technology widely utilized for seawater 339 desalination. It involves the arrangement of numerous anion exchange membranes (AEM) 340 and cation exchange membranes (CEM) placed alternately between two electrodes (**Fig.** 341 6). The electrolyte solution flows through the electrode compartment, while the feed so-342 lution traverses the adjacent compartment. When a voltage is applied, a reduction reaction 343 occurs at the cathode, during which hydroxide ions are formed. Similarly, within the an-344 ode compartment, an oxidation process occurs, leading to the production of protons. 345 There are slight variations in selectivity for different ions among various membranes used 346 in ED, and the accumulation of divalent ions often leads to membrane fouling. Further-347 more, these membranes tend to be expensive. Additionally, ion-exchange nanofibrous 348 membranes are frequently regarded as a means to enhance the flow and permeability of 349 membranes that selectively filter monovalent and divalent ions [78-80]. 350

Furthermore, ED was used to separate Ni²⁺ and Pb²⁺ from synthetic solutions by a 351 novel heterogeneous ED-CEM (consisting of 2-acrylamido-2-methylpropanesulfonic acid 352 hydrogel and PVC) with extraction efficiencies of 96.9% and 99% for Ni2+ and Pb2+, respec-353 tively. A pilot-scale ED system was employed to extract Cu2+, Ni2+, and small amounts of 354 Cd²⁺, Fe³⁺, Cr⁶⁺, and Zn²⁺ with elimination efficiencies exceeding 90%. ED removes As³⁺ and 355 As⁵⁺ in metallurgical wastewater, and the removal efficiency reaches 91.38% [81]. 356



Figure 6. Electrodialysis membrane separation technique.

Although membrane filtration has significantly progressed in laboratory-scale stud-359 ies, no pilot-scale or large-scale practical applications have been reported. In addition, its 360 main disadvantage is the sludge generation [15]. In contrast, membrane filtration operated 361 through direct pressure can facilitate wastewater treatment with reduced energy con-362 sumption and carbon dioxide emissions. In summary, membrane filtration-based 363 wastewater treatment systems hold the potential to attain net energy gains and additional 364 economic benefits, paving the way for self-sustaining municipal wastewater treatment 365 [65]. 366

4.2. Adsorption

Adsorption is a good weapon in fighting against toxic metals threatening our envi-368 ronment. It is an interfacial physicochemical phenomenon accumulating solute molecules on a solid-liquid or solid-gas interface [82]. On the molecular scale, adsorption can be categorized into chemical and physical, contingent on the underlying interactions guiding 371

357 358

367

the adsorption mechanism. The physical process assumes greater significance when it comes to the removal of heavy metals from wastewater. 373

In the case of physical adsorption, the Van der Walls molecular attractive forces that 374 retain the adsorbent on the surface are generally dominant, and the phenomenon is re-375 versible. The energy involved in the interaction between the adsorbate and the adsorbent 376 shares a similar order of magnitude with the interaction energy between the two. Never-377 theless, it typically exceeds the energy associated with the condensation of the adsorbate 378 (Fig. 7). Chemical adsorption, often referred to as activated adsorption, arises from a 379 chemical bonding or interaction occurring between the solid substrate and the adsorbed 380 substance. 381



Adsorbent surface

Physical adsorption

Chemical adsorption

Figure 7. Physical and chemical adsorption.

The growing ability of adsorption to remove hazardous substances without compromising water quality or generating harmful degradation products has led to its increased utilization over electrochemical, biochemical, or photochemical degradation processes. Additionally, adsorption provides a valuable alternative treatment choice, particularly when the adsorbent is cost-effective and readily obtainable [83]. Various adsorbents have been explored and detailed in existing literature, with these materials being obtainable from biological, organic, or mineral sources.

The significant costs tied to the creation and regeneration of activated carbon inspire 391 researchers to explore alternative avenues. Furthermore, due to low metal adsorption ca-392 pacity, synthetic materials like (E)-2-[(1H-imidazole-4-yl)methylidene]-Hydrazinecar-393 bothioamide ligand (EIMH) often do not provide greater than 80% removal efficiency for 394 heavy metals in wastewater treatment [84]. In recent years, novel adsorbent materials, 395 particularly nanostructured substances like carbon nanotubes, graphene, fullerenes, and 396 similar materials, have emerged. These materials are currently used to effectively adsorb 397 90% of heavy metals in wastewater because of their high specific surface area (500-1500 398 m²/g), large pore volume, and different intermolecular interactions [85]. Moreover, they 399 exhibit non-corrosive properties and possess outstanding mechanical strength, thermal 400 stability, and electrical conductivity. The adsorption performance of these nanostructured 401 materials is generally better than that of classic synthetic adsorbents, such as iron oxide, 402 titanium dioxide, activated carbon, etc. However, their limitations make scientific research 403 challenging regarding functionality and durability, which are essential in environmental 404 applications [86]. The adsorption efficiency can be improved by enhancing the carbon sur-405 face charge by adding surface functional groups such as carboxyl, phenyl, and lactone 406 [87]. Surface modification typically reduces the surface area while increasing the concen-407 tration of surface functional groups. Conversely, increasing the surface area of the adsor-408 bent, dosage of the adsorbent, initial concentration of metal ions, and contact duration 409

tend to enhance adsorption. Multiwalled carbon nanotubes have received significant at-410 tention for effectively removing heavy metals [88]. Their high hydrophobicity and the 411 presence of strong Van der Waals forces have generated considerable interest in their abil-412 ity to swiftly accumulate in aqueous solutions, thus enhancing their adsorption capacity. 413 The complex preparation process of carbon-based adsorbents contributes to their high 414 cost and limits their widespread industrial use. Therefore, it is necessary to continue ex-415 ploring innovative, affordable, and environmentally friendly surface modification tech-416 nologies [88]. 417

Chemical adsorbents encompass metal salt accelerators, activated silica, synthetic 418 polymers, and natural accelerators. Among these, metal salt adsorbents are widely em-419 ployed in sewage treatment plants due to their effectiveness in removing diverse pollu-420 tants like heavy metals and turbidity. The notable advantage of metal salt adsorbents over 421 other types lies in their efficiency in tackling various pollutants. Metal salt adsorbents are 422 readily accessible in the market and can deactivate bacteria. This affordability and availa-423 bility make them popular among users, especially compared to other chemical adsorbents 424 [89]. The metal salt adsorbents commonly utilized are those based on aluminum and iron 425 [90,91]. They show good efficiency (90-99%) for removal of heavy metals like copper, lead, 426 and nickel. In addition, they have the advantage of being less expensive and having a 427 wider pH range. Adsorption has several disadvantages, including the high cost of adsor-428 bent, separation of the adsorbent from metals, and low surface area [90,91]. 429

4.2.1. Filters: classification and applications

The treatment of industrial and domestic wastewater commonly utilizes adsorption-431 based filters due to their advantages over other methods, such as cost reduction and lower 432 energy consumption. However, several factors need to be considered and optimized for 433 optimal performance of these filters. The primary factors include the choice of adsorbent 434 material, which significantly impacts heavy metal removal efficiency. Adsorbent proper-435 ties such as specific surface area, pore size distribution, and functional groups play a cru-436 cial role [92]. Additionally, the characteristics of the heavy metals themselves, including 437 their concentration and the presence of competing ions, must be taken into account [93]. 438 Operating conditions, such as pH, temperature, and contact time, are also critical to ad-439 sorption [94]. Furthermore, the design and configuration of the filters, encompassing pa-440 rameters like porosity, depth, flow rate, and material leakage into the water effluents, re-441 quire careful consideration and improvement [95]. Specific factors such as the regenera-442 tion and reusability of the adsorbents [96], the particle size distribution [97], and the po-443 tential for chemical modifications to enhance adsorption capacity are equally important 444 [98]. The subsequent sections of this review delineate three categories of filters: synthetic, 445 natural, and hybrid, each with unique properties and applications. 446

4.2.1.1. Synthetic filters

Among the many synthetic filters designed and studied in the literature, polymerenhanced filters perform relatively well in removing heavy metals from water. They consist of deploying water-soluble polymeric ligands able to bind metal ions selectively, producing metals-free effluents. **Fig. 8** (a) presents a schematic depiction of a synthetic filter's distinct constituents. To illustrate the different metals that such filters can remove and their filtration efficiency, **Table 2** summarizes various important examples of synthetic filters described in the literature.

455

447

457

Synthetic filter **Targeted metals** Metal ions removal (%) Reference Pb²⁺ 80 PEI [99] Cu^{2+} 94 Cd^{2+} 99 PAN Cu²⁺ 96 [100] Pb²⁺ 95 Cd2+ 90 [101] Synthetic Zeolites Ni²⁺ 85 Pb²⁺ 45 Cu^{2+} 78 [102] Dithiocarbamate resins Hg²⁺ 80 As^{3+} 55 Cr^{3+} 76 As³⁺ 80 85 Metal Oxides Pb²⁺ [103] (e.g., MnO₂, Fe₃O₄) Cd2+ 65 Cr^{3+} 55 96 **MWCNTs** Cd2+ [104]

Table 2. Metal ions elimination by synthetic filters.

458

A filter layer may comprise a semi-interpenetrating polymer network composed of 459 cross-linked polyvinyl alcohol (PVA) as the matrix, with the effective utilization of PEI for 460 the removal of heavy metal ions, including Pb²⁺, Cd²⁺, and Cu²⁺ from aqueous solutions. 461 The PVA chains of the complexing polymer are entrapped in a cross-linked polymeric 462 matrix. The latter is fabricated through exposure to gaseous 1,2-dibromoethane. The ad-463 sorption reactions followed pseudo-first-order kinetics with similar rate constants for the 464 three cations, and the obtained equilibrium constants show the following affinity order: 465 Pb > Cu > Cd [99]. 466

In addition, PAN demonstrates a remarkable ability to effectively adsorb diverse 467 metal ions, rendering it highly appealing for purification purposes. Notably, it is consid-468 ered one of the most cost-effective polymers due to its abundant cyano groups at the ends, 469 which provide ample opportunity for further modification [105,106]. PAN fibers are pro-470 duced through free radical bulk polymerization, utilizing benzoyl peroxide as an initiator. 471 To achieve a bead-free and consistent morphology, a series of experiments were carried 472 out at different potential differences and flow rates. The electrospinning process was op-473 timized at a potential difference of 3 kV cm⁻¹ and a flow rate of 2 mL h⁻¹. At room temper-474 ature, the nitrile groups on the PAN fibers' surface react with H2NOH. Hydroxylamine 475 engages in a nucleophilic addition to the fiber surface, subsequently forming car-476 boximides. The fiber surface is then functionalized to chelate uranyl cations, and the re-477 sulting fiber can remove uranyl ion U(VI) pollutants [107]. By comparing the different 478 PAN fibers, good sorption results of U(VI) ions were obtained (81%). In contrast, only 19% 479 sorption is obtained when the PAN fibers remain unmodified. Most importantly, nano-480 fibers have a high surface area, enhancing their adsorption efficiency and making them 481 ready to link to various chelating agents [108]. 482 Recently, a new filter to remove Cd^{2+} and Pb^{2+} from the water was described in the literature. It consists of fly ash ceramic foams with a 3-D interconnected porous structure and multiwalled carbon nanotubes (MWCNTs) that are used by a combination of carbamate grafting and polydimethylsiloxane coating. The fly ash foam exhibited removal rates of 96.33% and 95.12% for Cd and Pb, respectively. This synthetic filter can save considerable economic effort due to gravity propulsion. It is suitable for front-end wastewater treatment on an industrial scale [104].

In a different investigation, PVA/SA beads were formulated by blending functionalized sodium alginate (SA) with polyvinyl alcohol (PVA), including glutaraldehyde as a cross-linking agent. Zeo/PVA/SA nanocomposite beads were made using zeolite nanoparticles (Zeo) and PVA/SA to remove heavy metals from wastewater with good efficiency: 99.5% (Pb²⁺), 99.2% (Cd²⁺), 98.8% (Sr²⁺), 97.2% (Cu²⁺), 95.6% (Zn²⁺), 93.1% (Ni²⁺), 92.4% (Mn²⁺), and 74.5% (Li²⁺) [109].

Using synthetic adsorbents in filters presents an opportunity to explore the binding 496 mechanisms of heavy metals with different compounds, whether in a specific or non-specific manner. The polymer-based adsorbents are particularly easy to produce and have 498 been extensively used. The manufacture of these synthetic filters demands a substantial 499 amount of energy. Furthermore, they are not environmentally friendly, as they are typically discarded after becoming saturated with pollutants, unlike natural or hybrid filters, 500 which can be reused. 502

Furthermore, the production of synthetic zeolites involves using numerous chemical 503 materials. While industrial waste products like cupola slag and spent fluid cracking cata-504 lysts are employed in synthesizing important zeolites (such as ZSM-5, NaA, and NaX), 505 they pose environmental hazards. Additionally, specific types of municipal solid waste, 506 such as non-recyclable glass and thin-walled aluminum scraps (including aluminum cans 507 and foils), can serve as sources of silicon (Si) and aluminum (Al) for zeolite synthesis. In 508 summary, zeolite stands as an exceptional adsorbent for effectively removing heavy met-509 als from wastewater [110]. 510

4.2.1.2. Natural filters

Natural wastewater treatment systems, including constructed wetlands, biological512sand filters, and other decentralized solutions, are gaining significance as viable alterna-513tives to previously discussed systems (Fig. 8 (b)). This is primarily due to their low establishment costs, ease of use, minimal management requirements, and high efficiency in re-514moving heavy metals from water [111,112].516

In less-developed countries, natural coagulants are commonly employed as point-of-517 use technology due to their ability to be locally produced and easily processed into a us-518 able form. These coagulants are biodegradable and present a cost-efficient substitute for 519 chemical coagulants. Their mode of action involves an adsorption mechanism, followed 520 by either charge neutralization or a polymeric combining effect [113]. Starch and cellulose 521 derivatives, proteinaceous materials, and gums from polysaccharides are significant nat-522 ural products used [88]. For example, Moringa and other native seeds can remove metals 523 from drinking water. Table 3Erreur ! Source du renvoi introuvable. shows that Moringa 524 seeds have higher removal rates of heavy metals than cowpea (Vigna unguiculata), urad 525 (Vigna mungo), peanuts (Arachis hypogaea), and corn (Zea mays). Moringa has a metal 526 removal rate of over 50%, with the highest removal efficiency for copper and lead (90% 527 for both). Beans (black-eyed peas) and peanuts did not have high metal removal efficiency 528 (<15% for Cu and Cd). Corn and urad beans exhibited limited capabilities for the removal 529 of heavy metals. The adsorption of metals using Moringa is restricted to its surface, as 530 Moringa is a cationic polyelectrolyte with a short chain and low molecular weight [114]. 531 Compared to other seeds, Moringa is particularly effective in efficiently removing heavy 532 metals and solids with charges higher than its colloidal surface. Nonetheless, additional 533

research is necessary to gain a deeper understanding of the molecular mechanisms under-534 pinning this effective water purification. This insight will be crucial in refining the design 535 of filters employing Moringa. 536

Table 3. Removal capacities of natural adsorbents.

| Natural Adaptionta | | Ren | ioval capa | city (%) | | Potoronco |
|-----------------------------------------------|------------------|------------------|------------------|------------------|------------------|-----------|
| Natural Ausorbents | Cu ²⁺ | Pb ²⁺ | Cd ²⁺ | Cr ³⁺ | Zn ²⁺ | Kelelence |
| Urad | 30 | 40 | 10 | 20 | 15 | [115] |
| Peanut | 4 | 50 | 8 | 40 | 22 | [116] |
| Bean | 3 | 29 | 10 | 40 | 30 | [117] |
| Corn | 10 | 13 | 40 | 10 | 20 | [118] |
| Moringa | 90 | 90 | 60 | 50 | 55 | [119] |
| Clay Minerals (e.g., Bentonite, Kaolinite) | - | 80 | 70 | 50 | | [120] |
| Zeolites | 90 | 75 | - | | 60 | [121] |
| Peat Moss | 52 | 50 | 70 | | - | [122] |
| Sawdust | 60 | 20 | 30 | 80 | _ | [123] |
| Algae (Biomass) | 50 | 40 | - | 60 | 45 | [124] |

Large amounts of biomass waste, peels, leaves, husks, stems, branches, and pods can 538 be valorized and used in biosorption [125]. Therefore, natural filters using biomass waste 539 adsorbents are green and sustainable filtration devices. For example, natural zeolites have 540 high ion exchange and adsorption performance due to their particular structure. The ion 541 exchange capacity is influenced by various factors, such as the structure of the framework, 542 the size and shape of the ions, the charge density of the anionic framework, the ionic 543 charge, and the concentration of the external electrolyte solution [126,127]. 544

Given the numerous advantages, the development and utilization of natural coagu-545 lants have promising prospects as an environmentally friendly and sustainable technol-546 ogy for wastewater treatment [128]. Without a doubt, the drawbacks of employing natural 547 adsorbents primarily stem from the characteristics of the adsorbents themselves. Factors 548 like seasonal variations and storage duration can impact natural adsorbents' production 549 and consistent availability [129–131]. 550

4.2.1.3. Hybrid filters

Although natural filters are environmentally sustainable and broadly applied in wa-552 ter filtration, their affinity for heavy metals remains moderate, and this finding makes the 553 raw materials less attractive. Improved heavy metal retention from wastewater has been 554 obtained for filters that include modified natural products. Such filtration devices are 555 called hybrid filters. Given their setup composition, they are environmentally sustainable. 556 Fig. 8 (c) shows a schematic representation of a hybrid filter. 557

Two exemplary hybrid filters will be discussed in this context, specifically, those 558 based on beetroot and limestone. Beetroot fibers, subjected to chemical modification using 559 sodium dodecyl sulfate (SDS), an anionic surfactant, were prepared as representative ex-560 amples. They proved their efficiency in cleaning water contaminated by organic and inor-561 ganic compounds and softening hard water (Table 4). In this specific system, Rahman et 562 al. pinpointed the factors influencing wastewater purification efficacy, which include fiber 563

551

particle size, initial pollutant concentration, and wastewater pH. The findings indicate that 564 the purification efficiency increases as the fiber size decreases (from millimeters to mi-565 crometers). The optimal pH for adsorption was determined to be in the range of 6.0-6.5. 566 Next, the modified filter's dual function was investigated by assessing its ability to remove 567 heavy metals and soften hard water through its chemical functionalities' positive and neg-568 ative charges. Hybrid filters based on beetroot fibers were able to remove various pollu-569 tants: metals (lead, zinc, nickel, copper), total solid dissolved, and methylene blue. They 570 exhibited high efficiency when particle size decreased due to increased contact surface 571 area at pH values from 6.0 to 6.5. The maximum removal was 70 mg/g of filter for heavy 572 metals like Pb, Cu, and Zn [132]. 573

Table 4. Metal ions elimination by a hybrid filter.

| Hybrid filter | Targeted metals | Metal ions removal (%) | Reference |
|----------------------------------------|--------------------|------------------------|-----------|
| | Pb ²⁺ | 100 | |
| | Zn^{2+} | 99 | |
| Beetroot fibers and SDS | Ni ²⁺ | 99 | [132–135] |
| | Cu ²⁺ | 99 | |
| Limestone and activated carbon | Fe ²⁺ | 100 | [136–138] |
| Clay-EDTA | Pb ²⁺ | 95 | [139] |
| Chitosan Modified with Thiol Groups | Hg ²⁺ | 90 | [140] |

The second hybrid filter example is based on limestone, a material with sedimentary 575 rock origin consisting mainly of CaCO₃, which in turn comes from calcite and then arag-576 onite. The chemical composition of limestone renders it an appropriate material for water 577 and wastewater treatment, as these elements possess the capability to adsorb a diverse 578 range of pollutants. While limestone is an eco-friendly substance extensively utilized in 579 water filtration, it exhibits a moderate affinity for heavy metals. Consequently, endeavors 580 have been dedicated to developing an environmentally friendly filtration system by com-581 bining limestone and activated carbon to eliminate iron from wastewater. A filter com-582 posed of activated carbon and limestone in a 1:1 ratio successfully removed a 10 mg/l iron 583 solution. Utilizing 1 kg of this filter, it only took 95 minutes, at pH 6, to completely elimi-584 nate iron ions from the contaminated solution [136]. 585

Furthermore, hybrid inorganic/natural adsorbents primarily based on alumina and 586 4-amino antipyrine have been implemented to remove Pb, Cu, and Cr from industrial 587 wastewater. The hybrid adsorbents have been effectively implemented for the selective 588 elimination of the heavy metals mentioned above, with recuperation values between 92 589 and 98%. Hybrid alumina adsorbents are known for their exceptional thermal decomposition resistance and a sturdy balance that approaches acid-leaching conditions [141]. They were recently replaced by natural adsorbents, representing a safe, more eco-friendly, 592 and energetically cheaper alternative [142] 593

Furthermore, modified zeolite can be easily obtained through basic or acidic treat-594 ment and surfactant modification. These alterations expand the range of potential appli-595 cations for zeolites in wastewater treatment. As a result, surfactant-modified zeolites have 596 the capability to not only adsorb anionic species but also undergo cation exchange with 597 metal cations. This is due to the large size of the modifying agents' cations, preventing 598 them from entering the inner channels. [110]. 599

574



Figure 8. (a) synthetic filter, (b) natural filter, and (c) hybrid filter (Source: This work).

4.2.1.4. Adsorbents regeneration/reuse

Spent adsorbents require regeneration after reaching their saturation point with con-603 taminants to restore their adsorption capacity and facilitate reuse. The reusability of spent 604 adsorbents depends on their recoverability, decontamination efficacy, and regeneration 605 potential [143]. High recovery and regeneration capabilities are highly desirable for com-606 mercial and industrial applications, as they can significantly reduce the overall cost asso-607 ciated with adsorbent production [144]. Although the regeneration process can be cycled 608 multiple times, regenerated adsorbents typically show a decrease in adsorption capacity 609 compared to fresh ones [145]. Selecting the most appropriate regeneration technique is 610 crucial for maximizing contaminant desorption efficiency. Several factors influence the 611 feasibility of industrial-scale application, including the type of adsorbent, contaminant, 612 adsorbent stability, toxicity, and the cost and energy demands of the regeneration process. 613

A diverse range of approaches can be employed to recover and regenerate spent adsorbents. These include magnetic separation [146], thermal desorption [147], solvent regeneration [148], microwave irradiation [149], supercritical fluid regeneration [150], and advanced oxidation processes [151].

- Magnetic separation leverages the magnetic properties of adsorbents to separate 618 them from the wastewater after they have adsorbed heavy metals. By applying an external 619 magnetic field, the magnetic adsorbents can be easily retrieved from the solution, facili-620 tating their regeneration and reuse. The primary advantages are efficiency and simplicity, 621 allowing for quick recovery of adsorbents, reducing the need for complex filtration meth-622 ods, and lowering operational costs while minimizing secondary pollution [152,153]. 623 However, the stability of the magnetic properties over multiple regeneration cycles needs 624 to be ensured. 625

- Thermal desorption involves heating the adsorbent to volatilize the adsorbed metals, which are then captured and treated separately. Regeneration temperatures typically 627

range from 200-800°C [147]. Thermal desorption can achieve high regeneration efficiencies, often exceeding 90%, making it suitable for large-scale industrial applications [152]. It is effective for various adsorbents, including activated carbons, zeolites, and metal-organic frameworks (MOFs), but has drawbacks like high energy requirements and potential degradation of adsorbents over multiple cycles.

- Solvent regeneration involves using various solvents to desorb metals from the ad-633 sorbent surface, effectively restoring its adsorption capacity. Common solvents include 634 acids (e.g., hydrochloric acid, nitric acid), alkalis (e.g., sodium hydroxide), and organic 635 solvents (e.g., ethanol, methanol)[148,152,154]. This method is particularly effective for 636 adsorbents with a strong affinity for certain heavy metals and is relatively cost-effective. 637 However, it poses environmental risks due to the disposal of spent solvents and can lead 638 to partial degradation of the adsorbent material over time. Recent advancements focus on 639 developing eco-friendly solvents and integrating solvent recovery systems to enhance sus-640 tainability [152,154]. 641

Microwave irradiation heats the adsorbent using microwave energy, volatilizing the
adsorbed contaminants and allowing for efficient desorption and regeneration. Studies
have shown that microwave-assisted regeneration can recover up to 90% of the adsorptive
capacity of activated carbon used in wastewater treatment [152,155]. This method is highly
effective at targeting specific adsorbed species and is energy-efficient and environmental impact [149].

- Supercritical fluid regeneration uses supercritical fluids, typically supercritical CO₂, 649 which operate under conditions above their critical temperature and pressure. This 650 method achieves high efficiency in desorbing heavy metals from adsorbents and can 651 achieve metal recovery rates exceeding 90%, making it highly effective for industrial ap-652 plications. Supercritical CO₂ is non-toxic, non-flammable, and relatively low-cost. The 653 process minimizes waste and environmental impact and preserves the structural integrity 654 of the adsorbent material over multiple regeneration cycles. However, it requires high in-655 itial setup costs and precise control of operational parameters to optimize regeneration 656 efficiency [156]. 657

Advanced oxidation processes (AOPs) generate highly reactive species, such as hydroxyl radicals, which degrade contaminants adsorbed onto the adsorbents, restoring
their adsorption capacity [157]. AOPs can regenerate adsorbents without significant material degradation, allowing for multiple reuse cycles and reducing operational costs.
These processes target a wide range of heavy metals, ensuring comprehensive removal
and recovery. However, they require high initial setup costs and precise control of operational parameters [157].

5. Comparison between filters and other methods

The need for cost-effective technologies to remove heavy metals from contaminated 666 soil and water is rising. The conventional and advanced metal removal methods described 667 in this review are summarized in Fig. 9. The conventional methods have become insuffi-668 cient to meet the current strict regulatory effluent restrictions and are sometimes expen-669 sive [128]. Adsorption is a commonly employed method for extracting toxic metals from 670 aquatic environments, achieving an average removal rate of around 99% through the use 671 of inexpensive adsorbents, including agricultural waste and activated carbon derived 672 from agricultural waste [158,159]. Therefore, there is a great demand for alternative, cost-673 effective technologies. Selecting a specific treatment method relies on various factors, in-674 cluding the waste's type and concentration, the effluent's variability, the desired remedi-675 ation level, and economic and energy considerations [40]. For example, filters use less en-676 ergy than osmosis pumps, which remains true when compared to pressurized nano and 677 ultra-filtration phenomena [68]. The pros and cons of different techniques for addressing 678 heavy metals in wastewater are outlined in Table 5. 679





Wastewater Treatment methods Conventional

Figure 9. Summary of the conventional and advanced wastewater treatment methods described in 681 this review.

Table 5. The primary benefits and drawbacks of diverse traditional and advanced approaches, in-683 cluding filtration methods, for eliminating heavy metals from wastewater. 684

| Treatment met | hod | Advantages | Disadvantages | Reference |
|---------------------------------|---------------|-----------------------------|----------------------------|------------|
| Coogulation on | d A | Low cost: | Sludge generation; | |
| Coagulation an | | Low cost, | The extra operational cost | [160] |
| flocculation or | precipitation | Simple operation | of sludge disposal | |
| | | Removal of metals and | | |
| Ion analysis | | organic pollutants | Long duration time; | [1/1] |
| ion exchange | \land | simultaneously; | Limited application | [101] |
| | | Less harmful byproduct | | |
| Membrane filtration, | | Small space requirement; | High operational cost due | |
| forward and reverses osmosis | | Low pressure; | to mombrane fouling | [160] |
| | | High separation selectivity | to memorane rouning | |
| | | Availability; | | |
| | | High efficiency; | Hard separation of the | |
| Adsorption | | Less expensive than other | adsorbent from metals | [162][163] |
| | | techniques; | | |
| | | Works in a wide pH range | | |
| Adsorption | Synthetic | High officionay | High operate consumption | [164] |
| by Filters | filters | r ngh eniciency | ringh energy consumption | [104] |

680

| | Natural | Low-cost; | Madarata offician av | [16] |
|--|-------------------|------------------------------|---------------------------|-------|
| | filters | Eco-friendly | Moderate efficiency | [165] |
| | Hybrid filters | High separation selectivity; | 50% harmful effect on the | |
| | | Economical; | | [166] |
| | | 50% Eco friendly | environment | |

6. Conclusion and perspectives

Ensuring the well-being of both human health and the environment entails the vital mission of eradicating heavy metals from wastewater. A plethora of treatment ap-proaches, encompassing adsorption, membrane filtration, coagulation and flocculation, chemical precipitation, ion exchange, flotation, electrochemical processes, and filtration, have been the subject of extensive research and have proven their effectiveness in the re-moval of heavy metals from wastewater. Each method has advantages and limitations, and further research and development are needed to optimize efficiency, cost-effective-ness, and environmental sustainability. The integration of multiple treatment methods and the development of hybrid systems can enhance the overall effectiveness of heavy metal removal. Additionally, using advanced materials, such as nanostructured materials, natural coagulants, and modified natural products, shows promise in improving heavy metal removal efficiency.

Natural filters are cheaper and more eco-friendly than hybrid filters but are less effi-cient than synthetic ones in removing organic and inorganic pollutants. To limit the im-pact of such filters on the environment, efforts should be invested to develop efficient bi-omass-based biodegradable filters for water treatment. A sustainable and economically viable strategy for designing such filters is the use of what is currently considered as bio-mass-waste. Coupling between various sorts of natural and hybrid filter treatment could also be a promising alternative. Furthermore, adopting sustainable wastewater treatment practices and promoting public awareness and education are essential in addressing global water pollution. By continuing to advance and implement efficient and sustainable wastewater treatment technologies, we can mitigate the harmful effects of heavy metals, protect water resources, and ensure a cleaner and safer environment for future genera-tions.

730

References

| 1. | Carolin, C.F.; Kumar, P.S.; Saravanan, A.; Joshiba, G.J.; Naushad, Mu. Efficient Techniques for the Removal of Toxic Heavy | 731 |
|-----|-----------------------------------------------------------------------------------------------------------------------------------|-----|
| | Metals from Aquatic Environment: A Review. Journal of Environmental Chemical Engineering 2017, 5, 2782-2799, | 732 |
| | doi:10.1016/j.jece.2017.05.029. | 733 |
| 2. | bica, J.; Chatterley, C.; Dönmez, A.; Johnston, R.; Mitis, F.; Slaymaker, T. Progress on Household Drinking Water, Sanitation and | 734 |
| | <i>Hygiene</i> 2000-2022; United state, 2023; | 735 |
| 3. | Carstea, E.M.; Bridgeman, J.; Baker, A.; Reynolds, D.M. Fluorescence Spectroscopy for Wastewater Monitoring: A Review. | 736 |
| | Water Research 2016, 95, 205–219, doi:10.1016/j.watres.2016.03.021. | 737 |
| 4. | John, M.J.; Anandjiwala, R.D. Recent Developments in Chemical Modification and Characterization of Natural Fiber- | 738 |
| | Reinforced Composites. Polym. Compos. 2008, 29, 187–207, doi:10.1002/pc.20461. | 739 |
| 5. | Wong, Y.C.; Szeto, Y.S.; Cheung, W.H.; McKay, G. Adsorption of Acid Dyes on Chitosan-Equilibrium Isotherm Analyses. | 740 |
| | Process Biochemistry 2004 , 39, 695–704, doi:10.1016/S0032-9592(03)00152-3. | 741 |
| 6. | Xu, P.; Zeng, G.M.; Huang, D.L.; Feng, C.L.; Hu, S.; Zhao, M.H.; Lai, C.; Wei, Z.; Huang, C.; Xie, G.X.; et al. Use of Iron Oxide | 742 |
| | Nanomaterials in Wastewater Treatment: A Review. Science of The Total Environment 2012, 424, 1-10, | 743 |
| | doi:10.1016/j.scitotenv.2012.02.023. | 744 |
| 7. | Yenkie, K.M. Integrating the Three E's in Wastewater Treatment: Efficient Design, Economic Viability, and Environmental | 745 |
| | Sustainability. Current Opinion in Chemical Engineering 2019, 26, 131–138, doi:10.1016/j.coche.2019.09.002. | 746 |
| 8. | António Guterres Water and Sanitation - United Nations Sustainable Development Available online: | 747 |
| | https://www.un.org/sustainabledevelopment/water-and-sanitation/ (accessed on 7 July 2023). | 748 |
| 9. | Gallego-Schmid, A.; Tarpani, R.R.Z. Life Cycle Assessment of Wastewater Treatment in Developing Countries: A Review. | 749 |
| | Water Research 2019 , 153, 63–79, doi:10.1016/j.watres.2019.01.010. | 750 |
| 10. | Raval, N.P. Adsorptive Removal of Nickel(II) Ions from Aqueous Environment: A Review. Journal of Environmental | 751 |
| | Management 2016 , 20. | 752 |
| 11. | Francisco RPeligro, L.M.; Ivana Pavlovic; Ricardo Rojas; Cristobalina Barriga Surface Charge Properties of and Cu(II) | 753 |
| | Adsorption by Spores of the Marine Bacillus Sp. Strain SG-1. Appl. Environ. Microbiol. 2016, 64, 1123-1129, | 754 |
| | doi:10.1128/AEM.64.3.1123-1129.1998. | 755 |
| 12. | Meena, A.K.; Mishra, G.K.; Rai, P.K.; Rajagopal, C.; Nagar, P.N. Removal of Heavy Metal Ions from Aqueous Solutions Using | 756 |
| | Carbon Aerogel as an Adsorbent. Journal of Hazardous Materials 2005, 122, 161–170, doi:10.1016/j.jhazmat.2005.03.024. | 757 |
| 13. | Zinicovscaia, I. Conventional Methods of Wastewater Treatment. In Cyanobacteria for Bioremediation of Wastewaters; | 758 |
| | Zinicovscaia, I., Cepoi, L., Eds.; Springer International Publishing: Cham, 2016; pp. 17–25 ISBN 978-3-319-26749-4. | 759 |
| 14. | Alikaj, M.; Brahushi, F. Heavy Metals Assessment in the Macrophytes of Viroi Lake. 2017. | 760 |
| 15. | Barakat, M.A. New Trends in Removing Heavy Metals from Industrial Wastewater. Arabian Journal of Chemistry 2011, 4, 361- | 761 |
| | 377, doi:10.1016/j.arabjc.2010.07.019. | 762 |
| 16. | Elkady, M.; Shokry, H.; Hamad, H. New Activated Carbon from Mine Coal for Adsorption of Dye in Simulated Water or | 763 |
| | Multiple Heavy Metals in Real Wastewater. Materials 2020, 13, 2498, doi:10.3390/ma13112498. | 764 |
| 17. | Rehman, M.S.U.; Kim, I.; Han, JI. Adsorption of Methylene Blue Dye from Aqueous Solution by Sugar Extracted Spent Rice | 765 |
| | Biomass. Carbohydrate Polymers 2012, 90, 1314–1322, doi:10.1016/j.carbpol.2012.06.078. | 766 |
| 18. | El malti, W.; Hijazi, A.; Khalil, Z.A.; Yaghi, Z.; Medlej, M.K.; Reda, M. Comparative Study of the Elimination of Copper, | 767 |
| | Cadmium, and Methylene Blue from Water by Adsorption on the Citrus Sinensis Peel and Its Activated Carbon. RSC Adv. | 768 |
| | 2022 , <i>12</i> , 10186–10197, doi:10.1039/D1RA08997H. | 769 |
| 19. | Wassim El Malti, W.E.; Hamieh, M.; Noaman, A.; El-Dine, R.N.; Hijazi, A.; Al-Khatib, W. Polyurethane Loaded with | 770 |
| | Vegetable Activated Carbon for Heavy Metals Removal from Water. Journal of Ecological Engineering 2021, 12. | 771 |

| 20. | Jiang, Z.; Ho, SH.; Wang, X.; Li, Y.; Wang, C. Application of Biodegradable Cellulose-Based Biomass Materials in | 772 |
|-----|----------------------------------------------------------------------------------------------------------------------------------|-----|
| | Wastewater Treatment. Environmental Pollution 2021, 290, 118087, doi:10.1016/j.envpol.2021.118087. | 773 |
| 21. | N. Herawati , S. Suzuki, N.; Hayashi, K.; Rivai, I.F. Cadmium, Copper, and Zinc Levels in Rice and Soil of Japan, Indonesia, | 774 |
| | and China by Soil Type. Bulletin of Environmental Contamination and Toxicology 2000, 64, 33–39, doi:10.1007/s001289910006. | 775 |
| 22. | Kabata-Pendias, A. Soil-Plant Transfer of Trace Elements-an Environmental Issue. Geoderma 2004, 122, 143-149, | 776 |
| | doi:10.1016/j.geoderma.2004.01.004. | 777 |
| 23. | Babel, S.; Kurniawan, T.A. Cr(VI) Removal from Synthetic Wastewater Using Coconut Shell Charcoal and Commercial | 778 |
| | Activated Carbon Modified with Oxidizing Agents and/or Chitosan. Chemosphere 2004, 54, 951–967, | 779 |
| | doi:10.1016/j.chemosphere.2003.10.001. | 780 |
| 24. | Ali, H.; Khan, E.; Sajad, M.A. Phytoremediation of Heavy Metals-Concepts and Applications. Chemosphere 2013, 91, 869- | 781 |
| | 881, doi:10.1016/j.chemosphere.2013.01.075. | 782 |
| 25. | Igiri, B.E.; Okoduwa, S.I.R.; Idoko, G.O.; Akabuogu, E.P.; Adeyi, A.O.; Ejiogu, I.K. Toxicity and Bioremediation of Heavy | 783 |
| | Metals Contaminated Ecosystem from Tannery Wastewater: A Review. Journal of Toxicology 2018, 2018, 1-16, | 784 |
| | doi:10.1155/2018/2568038. | 785 |
| 26. | Ab Hamid, N.H.; bin Mohd Tahir, M.I.H.; Chowdhury, A.; Nordin, A.H.; Alshaikh, A.A.; Suid, M.A.; Nazaruddin, N. 'Izzah; | 786 |
| | Nozaizeli, N.D.; Sharma, S.; Rushdan, A.I. The Current State-Of-Art of Copper Removal from Wastewater: A Review. Water | 787 |
| | 2022 , <i>14</i> , 3086, doi:10.3390/w14193086. | 788 |
| 27. | Fewtrell, L.; Kay, D.; MacGill, S. A Review of the Science behind Drinking Water Standards for Copper. International Journal | 789 |
| | of Environmental Health Research 2001, 11, 161–167, doi:10.1080/09603120020047573. | 790 |
| 28. | Schweitzer, L.; Noblet, J. Water Contamination and Pollution. In Green Chemistry; Elsevier, 2018; pp. 261–290 ISBN 978-0-12- | 791 |
| | 809270-5. | 792 |
| 29. | Al-Saydeh, S.A.; El-Naas, M.H.; Zaidi, S.J. Copper Removal from Industrial Wastewater: A Comprehensive Review. Journal | 793 |
| | of Industrial and Engineering Chemistry 2017, 56, 35–44, doi:10.1016/j.jiec.2017.07.026. | 794 |
| 30. | Badr, E.A.E.; Agrama, A.A.E.; Badr, S.A.E. Heavy Metals in Drinking Water and Human Health, Egypt. Nutrition & Food | 795 |
| | Science 2011 , 41, 210–217, doi:10.1108/00346651111132484. | 796 |
| 31. | Sun, L.; Lu, M.; Li, Q.; Jiang, H.; Yin, S. Research Progress of Arsenic Removal from Wastewater. IOP Conf. Ser.: Earth Environ. | 797 |
| | <i>Sci.</i> 2019 , <i>218</i> , 012142, doi:10.1088/1755-1315/218/1/012142. | 798 |
| 32. | Lu, Z.; Qi, X.; Zhu, X.; Li, X.; Li, K.; Wang, H. Highly Effective Remediation of High-Arsenic Wastewater Using Red Mud | 799 |
| | through Formation of AlAsO4@silicate Precipitate. Environmental Pollution 2021, 287, 117484, | 800 |
| | doi:10.1016/j.envpol.2021.117484. | 801 |
| 33. | Mansoorian, H.J.; Mahvi, A.H.; Jafari, A.J. Removal of Lead and Zinc from Battery Industry Wastewater Using | 802 |
| | Electrocoagulation Process: Influence of Direct and Alternating Current by Using Iron and Stainless Steel Rod Electrodes. | 803 |
| | Separation and Purification Technology 2014, 135, 165–175, doi:10.1016/j.seppur.2014.08.012. | 804 |
| 34. | Mitra, S.; Sarkar, A.; Sen, S. Removal of Chromium from Industrial Effluents Using Nanotechnology: A Review. Nanotechnol. | 805 |
| | <i>Environ. Eng.</i> 2017 , <i>2</i> , 11, doi:10.1007/s41204-017-0022-y. | 806 |
| 35. | Prasad, S.; Yadav, K.K.; Kumar, S.; Gupta, N.; Cabral-Pinto, M.M.S.; Rezania, S.; Radwan, N.; Alam, J. Chromium | 807 |
| | Contamination and Effect on Environmental Health and Its Remediation: A Sustainable Approaches. Journal of Environmental | 808 |
| | Management 2021 , 285, 112174, doi:10.1016/j.jenvman.2021.112174. | 809 |
| 36. | Chang, J.; Peng, D.; Deng, S.; Chen, J.; Duan, C. Efficient Treatment of Mercury(II)-Containing Wastewater in Aerated | 810 |
| | Constructed Wetland Microcosms Packed with Biochar. Chemosphere 2022, 290, 133302, | 811 |
| | doi:https://doi.org/10.1016/j.chemosphere.2021.133302. | 812 |
| | | |

| 37. | Djaenudin; Widyarani; Hariyadi, H.R.; Wulan, D.R.; Cahyaningsih, S. Removal of Nickel Ion from Electroplating Wastewater | 813 |
|-----|-------------------------------------------------------------------------------------------------------------------------------|-----|
| | Using Double Chamber Electrodeposition Cell (DCEC) Reactor Partitioned with Water Hyacinth (Eichhornia Crassipes) | 814 |
| | Leaves. IOP Conf. Ser.: Earth Environ. Sci. 2017, 60, 012020, doi:10.1088/1755-1315/60/1/012020. | 815 |
| 38. | Li, N.; Yuan, M.; Lu, S.; Xiong, X.; Xie, Z.; Liu, Y.; Guan, W. Highly Effective Removal of Nickel Ions from Wastewater by | 816 |
| | Calcium-Iron Layered Double Hydroxide. Front. Chem. 2023, 10, 1089690, doi:10.3389/fchem.2022.1089690. | 817 |
| 39. | Abbas, A.; Chen, L.; Liao, Y.; Wu, Z.; Yu, Y.; Yang, J. Removal of Bismuth Ion from Aqueous Solution by Pulverized Eggshells. | 818 |
| | DWT 2021 , 213, 395–405, doi:10.5004/dwt.2021.26724. | 819 |
| 40. | Wang, J.; Chen, C. Biosorbents for Heavy Metals Removal and Their Future. Biotechnology Advances 2009, 27, 195-226, | 820 |
| | doi:10.1016/j.biotechadv.2008.11.002. | 821 |
| 41. | Ibarra-Rodríguez, D.; Lizardi-Mendoza, J.; López-Maldonado, E.A.; Oropeza-Guzmán, M.T. Capacity of 'Nopal' Pectin as a | 822 |
| | Dual Coagulant-Flocculant Agent for Heavy Metals Removal. Chemical Engineering Journal 2017, 323, 19–28, | 823 |
| | doi:10.1016/j.cej.2017.04.087. | 824 |
| 42. | Chang, Q.; Zhang, M.; Wang, J. Removal of Cu2+ and Turbidity from Wastewater by Mercaptoacetyl Chitosan. Journal of | 825 |
| | Hazardous Materials 2009 , 169, 621–625, doi:10.1016/j.jhazmat.2009.03.144. | 826 |
| 43. | Sakhi, D.; Rakhila, Y.; Elmchaouri, A.; Abouri, M.; Souabi, S.; Jada, A. Optimization of Coagulation Flocculation Process for | 827 |
| | the Removal of Heavy Metals from Real Textile Wastewater. In Advanced Intelligent Systems for Sustainable Development | 828 |
| | (AI2SD'2018); Ezziyyani, M., Ed.; Advances in Intelligent Systems and Computing; Springer International Publishing: Cham, | 829 |
| | 2019; Vol. 913, pp. 257–266 ISBN 978-3-030-11880-8. | 830 |
| 44. | Samaei, S.M.; Gato-Trinidad, S.; Altaee, A. Performance Evaluation of Reverse Osmosis Process in the Post-Treatment of | 831 |
| | Mining Wastewaters: Case Study of Costerfield Mining Operations, Victoria, Australia. Journal of Water Process Engineering | 832 |
| | 2020 , 34, 101116, doi:10.1016/j.jwpe.2019.101116. | 833 |
| 45. | Teh, C.Y.; Budiman, P.M.; Shak, K.P.Y.; Wu, T.Y. Recent Advancement of Coagulation-Flocculation and Its Application in | 834 |
| | Wastewater Treatment. Ind. Eng. Chem. Res. 2016, 55, 4363–4389, doi:10.1021/acs.iecr.5b04703. | 835 |
| 46. | Yadav, M.; Gupta, R.; Sharma, R.K. Green and Sustainable Pathways for Wastewater Purification. In Advances in Water | 836 |
| | Purification Techniques; Elsevier, 2019; pp. 355–383 ISBN 978-0-12-814790-0. | 837 |
| 47. | Qasem, N.A.A.; Mohammed, R.H.; Lawal, D.U. Removal of Heavy Metal Ions from Wastewater: A Comprehensive and | 838 |
| | Critical Review. npj Clean Water 2021, 4, 36, doi:10.1038/s41545-021-00127-0. | 839 |
| 48. | Kumar, M.; Nandi, M.; Pakshirajan, K. Recent Advances in Heavy Metal Recovery from Wastewater by Biogenic Sulfide | 840 |
| | Precipitation. Journal of Environmental Management 2021, 278, 111555, doi:10.1016/j.jenvman.2020.111555. | 841 |
| 49. | Dabrowski, A.; Hubicki, Z.; Podkościelny, P.; Robens, E. Selective Removal of the Heavy Metal Ions from Waters and | 842 |
| | Industrial Wastewaters by Ion-Exchange Method. Chemosphere 2004, 56, 91–106, doi:10.1016/j.chemosphere.2004.03.006. | 843 |
| 50. | Tenório, J.A.S.; Espinosa, D.C.R. Treatment of Chromium Plating Process Effluents with Ion Exchange Resins. Waste | 844 |
| | Management 2001, 21, 637–642, doi:10.1016/S0956-053X(00)00118-5. | 845 |
| 51. | Kang, SY.; Lee, JU.; Moon, SH.; Kim, KW. Competitive Adsorption Characteristics of Co2+, Ni2+, and Cr3+ by IRN-77 | 846 |
| | Cation Exchange Resin in Synthesized Wastewater. Chemosphere 2004, 56, 141–147, doi:10.1016/j.chemosphere.2004.02.004. | 847 |
| 52. | Cavaco, S.A.; Fernandes, S.; Quina, M.M.; Ferreira, L.M. Removal of Chromium from Electroplating Industry Effluents by | 848 |
| | Ion Exchange Resins. Journal of Hazardous Materials 2007, 144, 634–638, doi:10.1016/j.jhazmat.2007.01.087. | 849 |
| 53. | Kobielska, P.A.; Howarth, A.J.; Farha, O.K.; Nayak, S. Metal-Organic Frameworks for Heavy Metal Removal from Water. | 850 |
| | Coordination Chemistry Reviews 2018, 358, 92–107, doi:10.1016/j.ccr.2017.12.010. | 851 |
| 54. | Fang, QR.; Yuan, DQ.; Sculley, J.; Li, JR.; Han, ZB.; Zhou, HC. Functional Mesoporous Metal-Organic Frameworks for | 852 |
| | the Capture of Heavy Metal Ions and Size-Selective Catalysis. Inorg. Chem. 2010, 49, 11637-11642, doi:10.1021/ic101935f. | 853 |

- Zhang, Y.; Xie, Z.; Wang, Z.; Feng, X.; Wang, Y.; Wu, A. Unveiling the Adsorption Mechanism of Zeolitic Imidazolate
 Framework-8 with High Efficiency for Removal of Copper Ions from Aqueous Solutions. *Dalton Trans.* 2016, 45, 12653–12660,
 doi:10.1039/C6DT01827K.
- Li, X.; Gao, X.; Ai, L.; Jiang, J. Mechanistic Insight into the Interaction and Adsorption of Cr(VI) with Zeolitic Imidazolate
 Framework-67 Microcrystals from Aqueous Solution. *Chemical Engineering Journal* 2015, 274, 238–246, 858
 doi:10.1016/j.cej.2015.03.127.
- Shah, R. Ligational, Potentiometric and Floatation Studies on Cu(II) Complexes of Hydrazones Derived from p and o-Vanillin
 Condensed with Diketo Hydrazide. *Journal of Molecular Liquids* 2016, 220, 939–953, doi:10.1016/j.molliq.2016.04.047.
- Dehghani, M.H.; Karimi, B.; Rajaei, M.S. The Effect of Aeration on Advanced Coagulation, Flotation and Advanced Oxidation
 Processes for Color Removal from Wastewater. *Journal of Molecular Liquids* 2016, 223, 75–80, doi:10.1016/j.molliq.2016.08.019.
 863
- 59. Elehinafe, F.B.; Agboola, O.; Vershima, A.D.; Bamigboye, G.O. Insights on the Advanced Separation Processes in Water
 864 Pollution Analyses and Wastewater Treatment A Review. South African Journal of Chemical Engineering 2022, 42, 188–200, 865 doi:10.1016/j.sajce.2022.08.004.
- Wang, G.; Nguyen, A.V.; Mitra, S.; Joshi, J.B.; Jameson, G.J.; Evans, G.M. A Review of the Mechanisms and Models of Bubble Particle Detachment in Froth Flotation. *Separation and Purification Technology* 2016, 170, 155–172, 868
 doi:10.1016/j.seppur.2016.06.041.
- Deliyanni, E.A.; Kyzas, G.Z.; Matis, K.A. Various Flotation Techniques for Metal Ions Removal. *Journal of Molecular Liquids* 870
 2017, 225, 260–264, doi:10.1016/j.molliq.2016.11.069.
- Jia, K.; Yi, Y.; Ma, W.; Cao, Y.; Li, G.; Liu, S.; Wang, T.; An, N. Ion Flotation of Heavy Metal Ions by Using Biodegradable 872 Biosurfactant as Collector: Application and Removal Mechanism. *Minerals Engineering* 2022, 176, 107338, 873 doi:10.1016/j.mineng.2021.107338.
- Ajiboye, T.O.; Oyewo, O.A.; Onwudiwe, D.C. Simultaneous Removal of Organics and Heavy Metals from Industrial 875
 Wastewater: A Review. *Chemosphere* 2021, 262, 128379, doi:10.1016/j.chemosphere.2020.128379.
- 64. Jin, W.; Fu, Y.; Hu, M.; Wang, S.; Liu, Z. Highly Efficient SnS-Decorated Bi2O3 Nanosheets for Simultaneous Electrochemical 877
 Detection and Removal of Cd(II) and Pb(II). *Journal of Electroanalytical Chemistry* 2020, 856, 113744, 878
 doi:10.1016/j.jelechem.2019.113744.
- Hube, S.; Eskafi, M.; Hrafnkelsdóttir, K.F.; Bjarnadóttir, B.; Bjarnadóttir, M.Á.; Axelsdóttir, S.; Wu, B. Direct Membrane
 Filtration for Wastewater Treatment and Resource Recovery: A Review. *Science of The Total Environment* 2020, 710, 136375,
 doi:10.1016/j.scitotenv.2019.136375.
- Amy, G.; Ghaffour, N.; Li, Z.; Francis, L.; Linares, R.V.; Missimer, T.; Lattemann, S. Membrane-Based Seawater Desalination:
 Present and Future Prospects. *Desalination* 2017, 401, 16–21, doi:10.1016/j.desal.2016.10.002.
- 67. Tijing, L.D.; Woo, Y.C.; Choi, J.-S.; Lee, S.; Kim, S.-H.; Shon, H.K. Fouling and Its Control in Membrane Distillation A Review.
 885 *Journal of Membrane Science* 2015, 475, 215–244, doi:10.1016/j.memsci.2014.09.042.
 886
- 68. Ozaki, H.; Sharma, K.; Saktaywin, W. Performance of an Ultra-Low-Pressure Reverse Osmosis Membrane (ULPROM) for
 887
 Separating Heavy Metal: Effects of Interference Parameters. *Desalination* 2002, 144, 287–294, doi:10.1016/S0011-9164(02)00329 6.
- Kajitvichyanukul, P.; Hung, Y.-T.; Wang, L.K. Membrane Technologies for Oil–Water Separation. In *Membrane and* 890 *Desalination Technologies*; Wang, L.K., Chen, J.P., Hung, Y.-T., Shammas, N.K., Eds.; Humana Press: Totowa, NJ, 2011; pp. 891 639–668 ISBN 978-1-58829-940-6.
- Abdullah, N.; Yusof, N.; Lau, W.J.; Jaafar, J.; Ismail, A.F. Recent Trends of Heavy Metal Removal from Water/Wastewater by
 Membrane Technologies. *Journal of Industrial and Engineering Chemistry* 2019, *76*, 17–38, doi:10.1016/j.jiec.2019.03.029.
 894

| 71. | He, M.; Wang, L.; Lv, Y.; Wang, X.; Zhu, J.; Zhang, Y.; Liu, T. Novel Polydopamine/Metal Organic Framework Thin Film | 895 |
|-----|---------------------------------------------------------------------------------------------------------------------------------|-----|
| | Nanocomposite Forward Osmosis Membrane for Salt Rejection and Heavy Metal Removal. Chemical Engineering Journal 2020, | 896 |
| | 389, 124452, doi:10.1016/j.cej.2020.124452. | 897 |
| 72. | Cui, Y.; Ge, Q.; Liu, XY.; Chung, TS. Novel Forward Osmosis Process to Effectively Remove Heavy Metal Ions. Journal of | 898 |
| | Membrane Science 2014, 467, 188–194, doi:10.1016/j.memsci.2014.05.034. | 899 |
| 73. | Abdullah, N.; Tajuddin, M.H.; Yusof, N. Forward Osmosis (FO) for Removal of Heavy Metals. In Nanotechnology in Water and | 900 |
| | Wastewater Treatment; Elsevier, 2019; pp. 177–204 ISBN 978-0-12-813902-8. | 901 |
| 74. | Cui, Y.; Wang, H.; Wang, H.; Chung, TS. Micro-Morphology and Formation of Layer-by-Layer Membranes and Their | 902 |
| | Performance in Osmotically Driven Processes. Chemical Engineering Science 2013, 101, 13–26, doi:10.1016/j.ces.2013.06.011. | 903 |
| 75. | Ge, Q.; Ling, M.; Chung, TS. Draw Solutions for Forward Osmosis Processes: Developments, Challenges, and Prospects for | 904 |
| | the Future. <i>Journal of Membrane Science</i> 2013, 442, 225–237, doi:10.1016/j.memsci.2013.03.046. | 905 |
| 76. | Chung, TS.; Zhang, S.; Wang, K.Y.; Su, J.; Ling, M.M. Forward Osmosis Processes: Yesterday, Today and Tomorrow. | 906 |
| | Desalination 2012, 287, 78–81, doi:10.1016/j.desal.2010.12.019. | 907 |
| 77. | Groves, G.R.; Buckley, C.A.; Cox, J.M.; Kirk, A.; Macmillan, C.D.; Simpson, M.J. Dynamic Membrane Ultrafiltration and | 908 |
| | Hyperfiltration for the Treatment of Industrial Effluents for Water Reuse. Desalination 1983, 47, 305–312, doi:10.1016/0011- | 909 |
| | 9164(83)87085-4. | 910 |
| 78. | Zheng, G. Nanofiber Membranes by Multi-Jet Electrospinning Arranged as Arc-Array with Sheath Gas for Electrodialysis | 911 |
| | Applications. <i>Materials and Design</i> 2020 . | 912 |
| 79. | Swanckaert, B.; Geltmeyer, J.; Rabaey, K.; De Buysser, K.; Bonin, L.; De Clerck, K. A Review on Ion-Exchange Nanofiber | 913 |
| | Membranes: Properties, Structure and Application in Electrochemical (Waste)Water Treatment. Separation and Purification | 914 |
| | <i>Technology</i> 2022 , 287, 120529, doi:10.1016/j.seppur.2022.120529. | 915 |
| 80. | Vickers, N.J. Animal Communication: When I'm Calling You, Will You Answer Too? Current Biology 2017, 27, R713-R715, | 916 |
| | doi:10.1016/j.cub.2017.05.064. | 917 |
| 81. | Min, K.J.; Kim, J.H.; Park, K.Y. Characteristics of Heavy Metal Separation and Determination of Limiting Current Density in | 918 |
| | a Pilot-Scale Electrodialysis Process for Plating Wastewater Treatment. Science of The Total Environment 2021, 757, 143762, | 919 |
| | doi:10.1016/j.scitotenv.2020.143762. | 920 |
| 82. | Nazir, A.; Khan, K.; Maan, A.; Zia, R.; Giorno, L.; Schroën, K. Membrane Separation Technology for the Recovery of | 921 |
| | Nutraceuticals from Food Industrial Streams. Trends in Food Science & Technology 2019, 86, 426–438, | 922 |
| | doi:10.1016/j.tifs.2019.02.049. | 923 |
| 83. | Yang, X.; Wan, Y.; Zheng, Y.; He, F.; Yu, Z.; Huang, J.; Wang, H.; Ok, Y.S.; Jiang, Y.; Gao, B. Surface Functional Groups of | 924 |
| | Carbon-Based Adsorbents and Their Roles in the Removal of Heavy Metals from Aqueous Solutions: A Critical Review. | 925 |
| | Chemical Engineering Journal 2019 , 366, 608–621, doi:10.1016/j.cej.2019.02.119. | 926 |
| 84. | Houari, B.; Louhibi, S.; Tizaoui, K.; Boukli-hacene, L.; Benguella, B.; Roisnel, T.; Dorcet, V. New Synthetic Material Removing | 927 |
| | Heavy Metals from Aqueous Solutions and Wastewater. Arabian Journal of Chemistry 2019, 12, 5040-5048, | 928 |
| | doi:10.1016/j.arabjc.2016.11.010. | 929 |
| 85. | Di Natale, F.; Gargiulo, V.; Alfè, M. Adsorption of Heavy Metals on Silica-Supported Hydrophilic Carbonaceous | 930 |
| | Nanoparticles (SHNPs). Journal of Hazardous Materials 2020, 393, 122374, doi:10.1016/j.jhazmat.2020.122374. | 931 |
| 86. | Chai, W.S.; Cheun, J.Y.; Kumar, P.S.; Mubashir, M.; Majeed, Z.; Banat, F.; Ho, SH.; Show, P.L. A Review on Conventional | 932 |
| | and Novel Materials towards Heavy Metal Adsorption in Wastewater Treatment Application. Journal of Cleaner Production | 933 |
| | 2021 , 296, 126589, doi:10.1016/j.jclepro.2021.126589. | 934 |
| 87. | Demiral, İ.; Samdan, C.; Demiral, H. Enrichment of the surface functional groups of activated carbon by modification method. | 935 |
| | Surfaces and Interfaces 2021 , 22, 100873, doi:10.1016/j.surfin.2020.100873. | 936 |

| 88. | Burakov, A.E.; Galunin, E.V.; Burakova, I.V.; Kucherova, A.E.; Agarwal, S.; Tkachev, A.G.; Gupta, V.K. Adsorption of Heavy | 937 |
|------|---------------------------------------------------------------------------------------------------------------------------------------|-----|
| | Metals on Conventional and Nanostructured Materials for Wastewater Treatment Purposes: A Review. Ecotoxicology and | 938 |
| | Environmental Safety 2018, 148, 702–712, doi:10.1016/j.ecoenv.2017.11.034. | 939 |
| 89. | Crini, G.; Lichtfouse, E. Advantages and Disadvantages of Techniques Used for Wastewater Treatment. Environ Chem Lett | 940 |
| | 2019 , 17, 145–155, doi:10.1007/s10311-018-0785-9. | 941 |
| 90. | Jagaba, A.H.; Kutty, S.R.M.; Hayder, G.; Latiff, A.A.A.; Aziz, N.A.A.; Umaru, I.; Ghaleb, A.A.S.; Abubakar, S.; Lawal, I.M.; | 942 |
| | Nasara, M.A. Sustainable Use of Natural and Chemical Coagulants for Contaminants Removal from Palm Oil Mill Effluent: | 943 |
| | A Comparative Analysis. Ain Shams Engineering Journal 2020, 11, 951–960, doi:10.1016/j.asej.2020.01.018. | 944 |
| 91. | Badawi, A.K.; Zaher, K. Hybrid Treatment System for Real Textile Wastewater Remediation Based on | 945 |
| | Coagulation/Flocculation, Adsorption and Filtration Processes: Performance and Economic Evaluation. Journal of Water | 946 |
| | Process Engineering 2021 , 40, 101963, doi:10.1016/j.jwpe.2021.101963. | 947 |
| 92. | Foo, K.Y.; Hameed, B.H. Insights into the Modeling of Adsorption Isotherm Systems. Chemical Engineering Journal 2010, 156, | 948 |
| | 2–10, doi:https://doi.org/10.1016/j.cej.2009.09.013. | 949 |
| 93. | Wu, FC.; Tseng, RL.; Juang, RS. Initial Behavior of Intraparticle Diffusion Model Used in the Description of Adsorption | 950 |
| | Kinetics. Chemical Engineering Journal 2009, 153, 1–8, doi:https://doi.org/10.1016/j.cej.2009.04.042. | 951 |
| 94. | Mohan, D.; Pittman, C.U. Arsenic Removal from Water/Wastewater Using Adsorbents-A Critical Review. Journal of | 952 |
| | <i>Hazardous Materials</i> 2007 , 142, 1–53, doi:https://doi.org/10.1016/j.jhazmat.2007.01.006. | 953 |
| 95. | Bhatnagar, A.; Sillanpää, M. Utilization of Agro-Industrial and Municipal Waste Materials as Potential Adsorbents for Water | 954 |
| | Treatment—A Review. Chemical Engineering Journal 2010, 157, 277–296, doi:https://doi.org/10.1016/j.cej.2010.01.007. | 955 |
| 96. | Singh, K.K.; Rastogi, R.; Hasan, S.H. Removal of Cr(VI) from Wastewater Using Rice Bran. Journal of Colloid and Interface | 956 |
| | Science 2005, 290, 61–68, doi:https://doi.org/10.1016/j.jcis.2005.04.011. | 957 |
| 97. | Mohan, S.V.; Karthikeyan, J. Removal of Lignin and Tannin Colour from Aqueous Solution by Adsorption onto Activated | 958 |
| | Charcoal. Environmental Pollution 1997, 97, 183–187, doi:https://doi.org/10.1016/S0269-7491(97)00025-0. | 959 |
| 98. | Gupta, V.K.; Ali, I.; Saleh, T.A.; Nayak, A.; Agarwal, S. Chemical Treatment Technologies for Waste-Water Recycling-an | 960 |
| | Overview. <i>RSC Adv.</i> 2012 , <i>2</i> , 6380–6388, doi:10.1039/C2RA20340E. | 961 |
| 99. | Bessbousse, H.; Rhlalou, T.; Verchère, JF.; Lebrun, L. Removal of Heavy Metal Ions from Aqueous Solutions by Filtration | 962 |
| | with a Novel Complexing Membrane Containing Poly(Ethyleneimine) in a Poly(Vinyl Alcohol) Matrix. Journal of Membrane | 963 |
| | <i>Science</i> 2008 , 307, 249–259, doi:10.1016/j.memsci.2007.09.027. | 964 |
| 100. | Li, P.; Jiang, H.; Barr, A.; Ren, Z.; Gao, R.; Wang, H.; Fan, W.; Zhu, M.; Xu, G.; Li, J. Reusable Polyacrylonitrile-Sulfur Extractor | 965 |
| | of Heavy Metal Ions from Wastewater. Adv Funct Materials 2021, 31, 2105845, doi:10.1002/adfm.202105845. | 966 |
| 101. | Velarde, L.; Nabavi, M.S.; Escalera, E.; Antti, ML.; Akhtar, F. Adsorption of Heavy Metals on Natural Zeolites: A Review. | 967 |
| | <i>Chemosphere</i> 2023 , 328, 138508, doi:10.1016/j.chemosphere.2023.138508. | 968 |
| 102. | Pereao, O.; Bode-Aluko, C.; Laatikainen, K.; Nechaev, A.; Petrik, L. Morphology, Modification and Characterisation of | 969 |
| | Electrospun Polymer Nanofiber Adsorbent Material Used in Metal Ion Removal. J Polym Environ 2019, 27, 1843–1860, | 970 |
| | doi:10.1007/s10924-019-01497-w. | 971 |
| 103. | Adsorbents for Water and Wastewater Treatment and Resource Recovery; Santos, S., Botelho, C., Pintor, A., Eds.; MDPI, 2022; ISBN | 972 |
| | 978-3-0365-4721-3. | 973 |
| 104. | Zhou, W.; Wu, P.; Zhang, L.; Zhu, D.; Zhao, X.; Cai, Y. Heavy Metal Ions and Particulate Pollutants Can Be Effectively | 974 |
| | Removed by a Gravity-Driven Ceramic Foam Filter Optimized by Carbon Nanotube Implantation. Journal of Hazardous | 975 |
| | <i>Materials</i> 2022 , <i>421</i> , 126721, doi:10.1016/j.jhazmat.2021.126721. | 976 |

| 105. | Li, X.; Zhang, C.; Zhao, R.; Lu, X.; Xu, X.; Jia, X.; Wang, C.; Li, L. Efficient Adsorption of Gold Ions from Aqueous Systems | 977 |
|------|-------------------------------------------------------------------------------------------------------------------------------|------|
| | with Thioamide-Group Chelating Nanofiber Membranes. Chemical Engineering Journal 2013, 229, 420–428, | 978 |
| | doi:10.1016/j.cej.2013.06.022. | 979 |
| 106. | Meng, J.; Cao, J.; Xu, R.; Wang, Z.; Sun, R. Hyperbranched Grafting Enabling Simultaneous Enhancement of the Boric Acid | 980 |
| | Uptake and the Adsorption Rate of a Complexing Membrane. J. Mater. Chem. A 2016, 4, 11656-11665, | 981 |
| | doi:10.1039/C6TA02348G. | 982 |
| 107. | Horzum, N.; Shahwan, T.; Parlak, O.; Demir, M.M. Synthesis of Amidoximated Polyacrylonitrile Fibers and Its Application | 983 |
| | for Sorption of Aqueous Uranyl Ions under Continuous Flow. Chemical Engineering Journal 2012, 213, 41-49, | 984 |
| | doi:10.1016/j.cej.2012.09.114. | 985 |
| 108. | Fu, F.; Wang, Q. Removal of Heavy Metal Ions from Wastewaters: A Review. Journal of Environmental Management 2011, 92, | 986 |
| | 407–418, doi:10.1016/j.jenvman.2010.11.011. | 987 |
| 109. | Isawi, H. Using Zeolite/Polyvinyl Alcohol/Sodium Alginate Nanocomposite Beads for Removal of Some Heavy Metals from | 988 |
| | Wastewater. Arabian Journal of Chemistry 2020 , 13, 5691–5716, doi:10.1016/j.arabjc.2020.04.009. | 989 |
| 110. | Yuna, Z. Review of the Natural, Modified, and Synthetic Zeolites for Heavy Metals Removal from Wastewater. Environmental | 990 |
| | Engineering Science 2016 , 33, 443–454, doi:10.1089/ees.2015.0166. | 991 |
| 111. | Ahluwalia, S.S.; Goyal, D. Microbial and Plant Derived Biomass for Removal of Heavy Metals from Wastewater. Bioresource | 992 |
| | <i>Technology</i> 2007 , <i>98</i> , 2243–2257, doi:10.1016/j.biortech.2005.12.006. | 993 |
| 112. | Shreya; Verma, A.K.; Dash, A.K.; Bhunia, P.; Dash, R.R. Removal of Surfactants in Greywater Using Low-Cost Natural | 994 |
| | Adsorbents: A Review. Surfaces and Interfaces 2021, 27, 101532, doi:10.1016/j.surfin.2021.101532. | 995 |
| 113. | Ahmad, A.; Kurniawan, S.B.; Abdullah, S.R.S.; Othman, A.R.; Hasan, H.A. Exploring the Extraction Methods for Plant-Based | 996 |
| | Coagulants and Their Future Approaches. Science of The Total Environment 2022, 818, 151668, | 997 |
| | doi:10.1016/j.scitotenv.2021.151668. | 998 |
| 114. | Ueda Yamaguchi, N.; Cusioli, L.F.; Quesada, H.B.; Camargo Ferreira, M.E.; Fagundes-Klen, M.R.; Salcedo Vieira, A.M.; | 999 |
| | Gomes, R.G.; Vieira, M.F.; Bergamasco, R. A Review of Moringa Oleifera Seeds in Water Treatment: Trends and Future | 1000 |
| | Challenges. Process Safety and Environmental Protection 2021, 147, 405-420, doi:10.1016/j.psep.2020.09.044. | 1001 |
| 115. | Raghuwanshi, P.K.; Mandloi, M.; Sharma, A.J.; Malviya, H.S.; Chaudhari, S. Improving Filtrate Quality Using Agrobased | 1002 |
| | Materials as Coagulant Aid. Water Quality Research Journal 2002, 37, 745–756, doi:10.2166/wqrj.2002.050. | 1003 |
| 116. | Nand, V. Water Purification Using Moringa Oleifera and Other Locally Available Seeds in Fiji for Heavy Metal Removal. | 1004 |
| | 2012, 2, 125–129, doi:https://doi.org/10.5696/2156-9614-11.31.210911. | 1005 |
| 117. | Etorki, A.M.; El-Rais, M.; Mahabbis, M.T.; Moussa, N.M. Removal of Some Heavy Metals from Wastewater by Using of Fava | 1006 |
| | Beans. AJAC 2014, 05, 225–234, doi:10.4236/ajac.2014.54028. | 1007 |
| 118. | Zvinowanda, C.M.; Okonkwo, J.O.; Shabalala, P.N.; Agyei, N.M. A Novel Adsorbent for Heavy Metal Remediation in | 1008 |
| | Aqueous Environments. Int. J. Environ. Sci. Technol. 2009, 6, 425-434, doi:10.1007/BF03326081. | 1009 |
| 119. | Muyibi, S.A.; Mohd. Noor, M.J.M.; Leong, T.K.; Loon, L.H. Effects of Oil Extraction from Moringa Oleifera Seeds On | 1010 |
| | Coagulation Of Turbid Water. International Journal of Environmental Studies 2002, 59, 243–254, doi:10.1080/00207230210924. | 1011 |
| 120. | Wang, F.Y.; Wang, H.; Ma, J.W. Adsorption of Cadmium (II) Ions from Aqueous Solution by a New Low-Cost Adsorbent- | 1012 |
| | Bamboo Charcoal. Journal of Hazardous Materials 2010, 177, 300–306, doi:10.1016/j.jhazmat.2009.12.032. | 1013 |
| 121. | Zhang, H.; Zhao, D.; Chen, L.; Yu, X.J. Investigation of Cu(II) Adsorption from Aqueous Solutions by NKF-6 Zeolite. Water | 1014 |
| | Science and Technology 2011 , 63, 395–402, doi:10.2166/wst.2011.233. | 1015 |
| 122. | Sen Gupta, B.; Curran, M.; Hasan, S.; Ghosh, T.K. Adsorption Characteristics of Cu and Ni on Irish Peat Moss. Journal of | 1016 |
| | Environmental Management 2009 , 90, 954–960, doi:10.1016/j.jenvman.2008.02.012. | 1017 |

| 123. | Meez, E.; Rahdar, A.; Kyzas, G.Z. Sawdust for the Removal of Heavy Metals from Water: A Review. <i>Molecules</i> 2021 , <i>26</i> , 4318, doi:10.3390/molecules26144318. | 1018 1019 |
|------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------|
| 124 | Caliceti M · Argese F · Sfriso A · Pavoni B Heavy Metal Contamination in the Seaweeds of the Venice Lagoon <i>Chemosphere</i> | 1020 |
| 121. | 2002 , <i>47</i> , 443–454, doi:10.1016/S0045-6535(01)00292-2. | 1020 |
| 125. | Bilal, M.; Rasheed, T.; Sosa-Hernández, J.; Raza, A.; Nabeel, F.; Iqbal, H. Biosorption: An Interplay between Marine Algae | 1022 |
| | and Potentially Toxic Elements – A Review. Marine Drugs 2018, 16, 65, doi:10.3390/md16020065. | 1023 |
| 126. | Alpat, S.K.; Özbayrak, Ö.; Alpat, Ş.; Akçay, H. The Adsorption Kinetics and Removal of Cationic Dye, Toluidine Blue O, from | 1024 |
| | Aqueous Solution with Turkish Zeolite. Journal of Hazardous Materials 2008, 151, 213-220, doi:10.1016/j.jhazmat.2007.05.071. | 1025 |
| 127. | Kallo, D. Applications of Natural Zeolites in Water and Wastewater Treatment. Reviews in Mineralogy and Geochemistry 2001, | 1026 |
| | 45, 519–550, doi:10.2138/rmg.2001.45.15. | 1027 |
| 128. | Bahrodin, M.B.; Zaidi, N.S.; Hussein, N.; Sillanpää, M.; Prasetyo, D.D.; Syafiuddin, A. Recent Advances on Coagulation- | 1028 |
| | Based Treatment of Wastewater: Transition from Chemical to Natural Coagulant. <i>Curr Pollution Rep</i> 2021, 7, 379–391, doi:10.1007/s40726-021-00191-7 | 1029 1030 |
| 129 | Vimer A : Dame B Panava Seed Extract as Coagulant for Potable Water Treatment in the Case of Tulte River for the | 1031 |
| 127. | Community of Yekuset District Ethiopia Environmental Challences 2021 4, 100198, doi:10.1016/i.envc.2021.100198 | 1032 |
| 130 | Merenstein II.: Bennett II Bridging Patterns of Neurocognitive Aging across the Older Adult Lifespan Neuroscience & | 1033 |
| 100. | Biobehavioral Reviews 2022, 135, 104594, doi:10.1016/i.neubiorev.2022.104594 | 1034 |
| 131 | Zhang, Q : Zhang, F : Ni, Y : Kokot, S. Effects of Aluminum on Amyloid-Beta Aggregation in the Context of Alzheimer's | 1035 |
| | Disease. <i>Arabian Journal of Chemistry</i> 2019 , <i>12</i> , 2897–2904, doi:10.1016/j.arabic.2015.06.019. | 1036 |
| 132. | Rahman, N.S.A.; Yhaya, M.F.; Azahari, B.; Ismail, W.R. Utilisation of Natural Cellulose Fibres in Wastewater Treatment. | 1037 |
| | <i>Cellulose</i> 2018 , <i>25</i> , 4887–4903, doi:10.1007/s10570-018-1935-8. | 1038 |
| 133. | Rima, J.; Rahme, K.; Assaker, K. The Use of Modified Beetroot Fibers by Sodium Dodecyl Sulfate (SDS) Cleaning Water | 1039 |
| | Contaminated by Organic and Inorganic Compounds. JFR 2014, 3, 19, doi:10.5539/jfr.v3n5p19. | 1040 |
| 134. | Rima, J.; Assaker, K. B-Cyclodextrin Polyurethanes Copolymerised with Beetroot Fibers (Bio-Polymer), for the Removal of | 1041 |
| | Organic and Inorganic Contaminants from Water. JFR 2013, 2, 150, doi:10.5539/jfr.v2n1p150. | 1042 |
| 135. | Rima, J.; Ghauch, A.; Ghaouch, M.; Martin-Bouyer, M. Cleaning of Water Contaminated by Heavy Metals Using Beetroot | 1043 |
| | Fibers as Biofilter. Toxicological & Environmental Chemistry 2000, 75, 89–97, doi:10.1080/02772240009358895. | 1044 |
| 136. | Abdulla, G.; Kareem, M.M.; Hashim, K.S.; Muradov, M.; Kot, P.; Mubarak, H.A.; Abdellatif, M.; Abdulhadi, B. Removal of | 1045 |
| | Iron from Wastewater Using a Hybrid Filter. IOP Conf. Ser.: Mater. Sci. Eng. 2020, 888, 012035, doi:10.1088/1757- | 1046 |
| | 899X/888/1/012035. | 1047 |
| 137. | Hussain, S.; Aziz, H.A.; Isa, M.H.; Ahmad, A.; Van Leeuwen, J.; Zou, L.; Beecham, S.; Umar, M. Orthophosphate Removal | 1048 |
| | from Domestic Wastewater Using Limestone and Granular Activated Carbon. Desalination 2011, 271, 265–272, | 1049 |
| | doi:10.1016/j.desal.2010.12.046. | 1050 |
| 138. | Bakar, A.A.A.; Razak, N.F.; Akbar, N.A.; Daud, N.M.; Ali, K.A.M. Removal of Cr (III) from Industrial Wastewater Using | 1051 |
| | Coconut Shell Carbon and Limestone as Adsorbent. IOP Conf. Ser.: Earth Environ. Sci. 2021, 646, 012063, doi:10.1088/1755- | 1052 |
| | 1315/646/1/012063. | 1053 |
| 139. | Li, Z.; Pan, Z.; Wang, Y. Preparation of Ternary Amino-Functionalized Magnetic Nano-Sized Illite-Smectite Clay for | 1054 |
| | Adsorption of Pb(II) Ions in Aqueous Solution. Environ Sci Pollut Res 2020, 27, 11683–11696, doi:10.1007/s11356-020-07766-9. | 1055 |
| 140. | He, H.; Meng, X.; Yue, Q.; Yin, W.; Gao, Y.; Fang, P.; Shen, L. Thiol-Ene Click Chemistry Synthesis of a Novel Magnetic | 1056 |
| | Mesoporous Silica/Chitosan Composite for Selective Hg(II) Capture and High Catalytic Activity of Spent Hg(II) Adsorbent. | 1057 |
| | <i>Chemical Engineering Journal</i> 2021 , 405, 126743, doi:10.1016/j.cej.2020.126743. | 1058 |
| | | |

| 141. | Mahmoud, M.E.; Hafez, O.F.; Alrefaay, A.; Osman, M.M. Performance Evaluation of Hybrid Inorganic/Organic Adsorbents | 1059 |
|------|--------------------------------------------------------------------------------------------------------------------------------|------|
| | in Removal and Preconcentration of Heavy Metals from Drinking and Industrial Waste Water. Desalination 2010, 253, 9–15, | 1060 |
| | doi:10.1016/j.desal.2009.11.044. | 1061 |
| 142. | Chen, S.; Yuan, Z.; Hanigan, D.; Westerhoff, P.; Zhao, H.; Ni, J. Coagulation Behaviors of New Covalently Bound Hybrid | 1062 |
| | Coagulants (CBHyC) in Surface Water Treatment. Separation and Purification Technology 2018, 192, 322-328, | 1063 |
| | doi:10.1016/j.seppur.2017.10.036. | 1064 |
| 143. | Yang, J.; Shen, M.; Wen, H.; Luo, Y.; Huang, R.; Rong, L.; Xie, J. Recent Advance in Delivery System and Tissue Engineering | 1065 |
| | Applications of Chondroitin Sulfate. Carbohydrate Polymers 2020, 230, 115650, doi:10.1016/j.carbpol.2019.115650. | 1066 |
| 144. | Gupta, V.K.; Agarwal, S.; Ahmad, R.; Mirza, A.; Mittal, J. Sequestration of Toxic Congo Red Dye from Aqueous Solution | 1067 |
| | Using Ecofriendly Guar Gum/ Activated Carbon Nanocomposite. International Journal of Biological Macromolecules 2020, 158, | 1068 |
| | 1310–1318, doi:10.1016/j.ijbiomac.2020.05.025. | 1069 |
| 145. | Harikishore Kumar Reddy, D.; Vijayaraghavan, K.; Kim, J.A.; Yun, YS. Valorisation of Post-Sorption Materials: | 1070 |
| | Opportunities, Strategies, and Challenges. Advances in Colloid and Interface Science 2017, 242, 35-58, | 1071 |
| | doi:10.1016/j.cis.2016.12.002. | 1072 |
| 146. | Tamjidi, S.; Esmaeili, H.; Kamyab Moghadas, B. Application of Magnetic Adsorbents for Removal of Heavy Metals from | 1073 |
| | Wastewater: A Review Study. Mater. Res. Express 2019, 6, 102004, doi:10.1088/2053-1591/ab3ffb. | 1074 |
| 147. | Hwang, S.Y.; Lee, G.B.; Kim, J.H.; Hong, B.U.; Park, J.E. Pre-Treatment Methods for Regeneration of Spent Activated Carbon. | 1075 |
| | Molecules 2020, 25, 4561, doi:10.3390/molecules25194561. | 1076 |
| 148. | Jiang, D.; Chu, B.; Amano, Y.; Machida, M. Removal and Recovery of Phosphate from Water by Mg-Laden Biochar: Batch | 1077 |
| | and Column Studies. Colloids and Surfaces A: Physicochemical and Engineering Aspects 2018, 558, 429-437, | 1078 |
| | doi:10.1016/j.colsurfa.2018.09.016. | 1079 |
| 149. | Zhang, X.N.; Mao, G.Y.; Jiao, Y.B.; Shang, Y.; Han, R.P. Adsorption of Anionic Dye on Magnesium Hydroxide-Coated | 1080 |
| | Pyrolytic Bio-Char and Reuse by Microwave Irradiation. Int. J. Environ. Sci. Technol. 2014, 11, 1439–1448, doi:10.1007/s13762- | 1081 |
| | 013-0338-5. | 1082 |
| 150. | Momina, M.; Shahadat, M.; Isamil, S. Regeneration Performance of Clay-Based Adsorbents for the Removal of Industrial | 1083 |
| | Dyes: A Review. RSC Adv. 2018, 8, 24571–24587, doi:10.1039/C8RA04290J. | 1084 |
| 151. | Acevedo-García, V.; Rosales, E.; Puga, A.; Pazos, M.; Sanromán, M.A. Synthesis and Use of Efficient Adsorbents under the | 1085 |
| | Principles of Circular Economy: Waste Valorisation and Electroadvanced Oxidation Process Regeneration. Separation and | 1086 |
| | Purification Technology 2020 , 242, 116796, doi:10.1016/j.seppur.2020.116796. | 1087 |
| 152. | Lata, S.; Singh, P.K.; Samadder, S.R. Regeneration of Adsorbents and Recovery of Heavy Metals: A Review. International | 1088 |
| | Journal of Environmental Science and Technology 2015 , 12, 1461–1478, doi:10.1007/s13762-014-0714-9. | 1089 |
| 153. | Ruthiraan, M.; Mubarak, N.M.; Abdullah, E.C.; Khalid, M.; Nizamuddin, S.; Walvekar, R.; Karri, R.R. An Overview of | 1090 |
| | Magnetic Material: Preparation and Adsorption Removal of Heavy Metals from Wastewater. In Magnetic Nanostructures : | 1091 |
| | Environmental and Agricultural Applications; Abd-Elsalam, K.A., Mohamed, M.A., Prasad, R., Eds.; Springer International | 1092 |
| | Publishing: Cham, 2019; pp. 131–159 ISBN 978-3-030-16439-3. | 1093 |
| 154. | Younas, F.; Mustafa, A.; Farooqi, Z.U.R.; Wang, X.; Younas, S.; Mohy-Ud-Din, W.; Ashir Hameed, M.; Mohsin Abrar, M.; | 1094 |
| | Maitlo, A.A.; Noreen, S.; et al. Current and Emerging Adsorbent Technologies for Wastewater Treatment: Trends, Limitations, | 1095 |
| | and Environmental Implications. Water 2021, 13, doi:10.3390/w13020215. | 1096 |
| 155. | El Messaoudi, N.; El Khomri, M.; El Mouden, A.; Bouich, A.; Jada, A.; Lacherai, A.; Iqbal, H.M.N.; Mulla, S.I.; Kumar, V.; | 1097 |
| | Américo-Pinheiro, J.H.P. Regeneration and Reusability of Non-Conventional Low-Cost Adsorbents to Remove Dyes from | 1098 |
| | Wastewaters in Multiple Consecutive Adsorption-Desorption Cycles: A Review. Biomass Conversion and Biorefinery 2022, | 1099 |
| | doi:10.1007/s13399-022-03604-9. | 1100 |
| | | |

| 156. | Tiwari, A.K.; Pal, S.L.; Srivastava, N.; Shah, M.; Ahmad, I.; Alshahrani, M.Y.; Pal, D.B. Bioadsorbent and Adsorbent-Based | 1101 | |
|--------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------|--|
| | Heavy Metal Removal Technologies from Wastewater: New Insight. Biomass Conversion and Biorefinery 2023, 13, 13335–13356, | 1102 | |
| | doi:10.1007/s13399-022-02343-1. | 1103 | |
| 157. | Bello, M.M.; Raman, A.A.A. Synergy of Adsorption and Advanced Oxidation Processes in Recalcitrant Wastewater | 1104 | |
| | Treatment. Environmental Chemistry Letters 2019, 17, 1125–1142, doi:10.1007/s10311-018-00842-0. | 1105 | |
| 158. | Ahalya, N.; Kanamadi, R.D.; Ramachandra, T.V. Biosorption of Chromium (VI) from Aqueous Solutions by the Husk of | 1106 | |
| | Bengal Gram (Cicer Arientinum). Electron. J. Biotechnol. 2005, 8, 258–264, doi:10.2225/vol8-issue3-fulltext-10. | 1107 | |
| 159. | Wang, W.; Wang, R.; Zhang, C.; Lu, S.; Liu, T. Synthesis, Characterization and Self-Assembly Behavior in Water as | 1108 | |
| | Fluorescent Sensors of Cationic Water-Soluble Conjugated Polyfluorene-b-Poly(N-Isopropylacrylamide) Diblock | 1109 | |
| | Copolymers. Polymer 2009, 50, 1236–1245, doi:10.1016/j.polymer.2008.12.026. | 1110 | |
| 160. | Kurniawan, T.A.; Chan, G.Y.S.; Lo, WH.; Babel, S. Physico-Chemical Treatment Techniques for Wastewater Laden with | 1111 | |
| | Heavy Metals. Chemical Engineering Journal 2006, 118, 83–98, doi:10.1016/j.cej.2006.01.015. | 1112 | |
| 161. | Barakat, M. Removal of Toxic Cyanide and Cu(II) Ions from Water by Illuminated TiO2 Catalyst. Applied Catalysis B: | 1113 | |
| | Environmental 2004, 53, 13–20, doi:10.1016/j.apcatb.2004.05.003. | 1114 | |
| 162. | Chakraborty, R.; Asthana, A.; Singh, A.K.; Jain, B.; Susan, A.B.H. Adsorption of Heavy Metal Ions by Various Low-Cost | 1115 | |
| | Adsorbents: A Review. International Journal of Environmental Analytical Chemistry 2022, 102, 342–379, | 1116 | |
| | doi:10.1080/03067319.2020.1722811. | 1117 | |
| 163. | Adsorption Processes for Water Treatment and Purification; Bonilla-Petriciolet, A., Mendoza-Castillo, D.I., Reynel-Ávila, H.E., | 1118 | |
| | Eds.; Springer International Publishing: Cham, 2017; ISBN 978-3-319-58135-4. | 1119 | |
| 164. | Abdullah, R.; Abustan, I.; Ibrahim, A.N.M. Wastewater Treatment Using Bentonite, the Combinations of Bentonite- Zeolite, | 1120 | |
| | Bentonite-Alum, and Bentonite-Limestone as Adsorbent and Coagulant. 2013, 4, 13. | 1121 | |
| 165. | Zheng, R.; Chen, Y.; Wang, J.; Song, J.; Li, XM.; He, T. Preparation of Omniphobic PVDF Membrane with Hierarchical | 1122 | |
| | Structure for Treating Saline Oily Wastewater Using Direct Contact Membrane Distillation. Journal of Membrane Science 2018, | 1123 | |
| | 555, 197–205, doi:10.1016/j.memsci.2018.03.041. | 1124 | |
| 166. | Boulaadjoul, S.; Zemmouri, H.; Bendjama, Z.; Drouiche, N. A Novel Use of Moringa Oleifera Seed Powder in Enhancing the | 1125 | |
| | Primary Treatment of Paper Mill Effluent. Chemosphere 2018, 206, 142–149, doi:10.1016/j.chemosphere.2018.04.123. | 1126 | |
| | | 1127 | |
| Discla | Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(a) and contained in all publications are solely those of the individual author (b) and (on the aditor(c)) disclaim groups with the sole of the individual author (c) and contained in all publications are solely those of the individual author (c) and contained in all publications are solely those of the individual author (c) and contained in all publications are solely those of the individual author (c) and contained in all publications are solely those of the individual author (c) and contained in all publications are solely those of the individual author (c) and contained in all publications are solely those of the individual author (c) and (c) and (c) and (c) and (c) are solely the solely and (c) are solely the solely and (c) are solely the solely are solely and (c) are solely are solely are solely are solely at the solely are solely at the solely are solely are solely at the solely are solely are solely at the solely at th | | |

thor(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to1129people or property resulting from any ideas, methods, instructions or products referred to in the content.1130