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Sustainable Approaches for Wastewater Treatment: An Analysis of Sludge-Based Materials for Heavy Metal Removal from Wastewater by Adsorption

R. M. A. S. Dhananjana Rajakaruna [1], B. G. N. Sewwandi [1,*], Mohamed M. M. Najim [2], Mirza Barjees Baig [3], Bader Alhafi Alotaibi [4] and Abou Traore [5]

[1] Department of Zoology and Environmental Management, Faculty of Science, University of Kelaniya, Kelaniya 11600, Sri Lanka; sdrajakaruna97@gmail.com
[2] Faculty of Agriculture, Sultan Sharif Ali Islamic University, Kampus Sinaut, Km 33, Jalan Tutong, Tutong TB1741, Brunei; najim.mujithaba@unissa.edu.bn
[3] Prince Sultan Institute for Environmental, Water & Desert Research, King Saud University, Riyadh 11451, Saudi Arabia; mbbaig@ksu.edu.sa
[4] Department of Agricultural Extension and Rural Society, College of Food and Agriculture Sciences, King Saud University, Riyadh 11451, Saudi Arabia; balhafi@ksu.edu.sa
[5] Department of Community Sustainability, College of Agriculture and Natural Resources, Michigan State University, 328 Natural Resources Building, East Lansing, MI 48824, USA; traoreab@msu.edu
* Correspondence: sewwandidh@kln.ac.lk

Abstract: A comprehensive study incorporating results from different findings related to heavy metal removals from wastewater using sludge as an absorbent will assist researchers and practitioners in planning wastewater treatment processes. This study aims to provide a comprehensive foundation on the potential of using sludge-based materials to remove heavy metals from wastewater based on recent studies. The physicochemical properties of sludge and the nature of metal ions have significantly contributed to the adsorption of heavy metals into sludge-based materials. Many researchers found the effects of pH, temperature, initial heavy metal concentrations, contact time, and adsorbent dose on the adsorption characteristics of heavy metals into sludge-based materials. Isotherm, kinetic, and thermodynamic studies have explained the mechanism of heavy metal adsorption by sludge-based materials. The effectiveness of regeneration of sludge-based adsorbents has been investigated by some researchers, providing an environmentally friendly solution to remove heavy metals from wastewater. It was found that less attention has been paid to metal recovery and recycling of sludge-based adsorbents, which indicates the need for future studies to enhance the reusability of sludge in wastewater treatment. Moreover, many studies have been conducted as lab-scale experiments on heavy metal adsorption from aqueous solutions using sludge-based adsorbents, leaving a research gap for future studies to focus on the removal of heavy metals from actual wastewater at field scale.

Keywords: adsorption mechanisms; isotherm models; kinetic models; regeneration; sludge; wastewater treatment

Citation: Rajakaruna, R.M.A.S.D.; Sewwandi, B.G.N.; Najim, M.M.M.; Baig, M.B.; Alotaibi, B.A.; Traore, A. Sustainable Approaches for Wastewater Treatment: An Analysis of Sludge-Based Materials for Heavy Metal Removal from Wastewater by Adsorption. *Sustainability* **2023**, *15*, 14937. https://doi.org/10.3390/su152014937

Academic Editors: Andreas Angelakis and Agostina Chiavola

Received: 13 July 2023
Revised: 11 October 2023
Accepted: 12 October 2023
Published: 16 October 2023

1. Introduction

The demand for water has been increasing with the increase in human population over time, which has led to acute stress on global freshwater resources [1]. Water stored in reservoirs, lakes, rivers, streams, etc., is used for human consumption, such as agricultural, industrial, recreational, and domestic purposes, and these processes generate polluted water [2]. Water pollution leads to the deterioration of both the mental and physical health of people [3,4]. Water pollution can occur from both point sources and non-point sources. Point source pollution is mainly caused by discharges from industrial facilities and municipal wastewater treatment plants, while major non-point sources are agricultural runoff, land disturbances, and storm drainage [5–7]. Different types of water pollution

sources add a wide range of pollutants to water, which can be organic and inorganic pollutants, radioactive pollutants, pharmaceuticals, pathogens, microplastics, heavy metals, and other pollutants [8–11]. Among all these water pollutants, heavy metals play a major role in water pollution, leading to many environmental and health problems [12].

Heavy metals are released to water bodies by various activities, such as industrial, mining, and agricultural activities, and many of these activities induce the distribution of heavy metals in ecosystems [13–16]. Ecosystem health is severely affected by heavy metal pollution of water due to its bioaccumulation potential along food chains, high toxicity, non-biodegradability, and persistence in the environment [17–19]. The accumulation of heavy metals with a high degree of toxicity, such as Pb^{2+}, Hg^{2+}, Cd^{2+}, and Ni^{2+}, can cause adverse effects on the metabolic processes of the human body [20]. They can cause multiple organ damage and DNA damage, even at lower levels of exposure [21]. Both the USEPA and the International Agency for Research on Cancer have identified several heavy metals as human carcinogens [22]. Some of them are Pb^{2+}, Hg^{2+}, Cd^{2+} and Ni^{2+} [23]. Since heavy metals are highly toxic, non-biodegradable, and harmful, even in small concentrations, it is crucial to establish suitable wastewater treatment methods for efficient removal of those from the environment.

Many wastewater treatment methods, such as ion exchange, chemical precipitation, reverse osmosis, coagulation, membrane filtration, flotation, and electrochemical treatment, are used for the removal of heavy metals from wastewater [24–30]. It is important to select the most appropriate wastewater treatment method, which depends on the cost required for treatment, the characteristics of the wastewater, and the applicability of the process [26]. Nevertheless, these methods have several drawbacks, such as inadequate removal, poor efficiency, high operation and maintenance cost, sensitive operating conditions, high energy requirements, and expensive disposal methods [31,32]. In adsorption, heavy metals (adsorbate) move from wastewater into the solid material (adsorbent) [12]. The chemical and physical properties of the adsorbent influence the efficiency of the heavy metal removal process from wastewater [33]. Adsorption is a widely used wastewater treatment method for heavy metal removal owing to its high efficiency, cost-effectiveness, eco-friendliness, and ability to produce well-treated effluent [34–36].

Conventional adsorbents such as activated carbons are well known for their efficiency in removing heavy metals from wastewater. Activated carbon is used in various water treatment facilities, including drinking water treatment plants and industrial and municipal wastewater treatment plants [34,37,38]. Despite the high heavy metal removal efficiency, universal application of conventional adsorbents is limited due to high cost, signifying the need for economical adsorbents efficient enough to remove heavy metals from wastewater [39,40].

Low-cost, non-conventional adsorbents are viable alternatives to conventional, expensive adsorbents for the removal of heavy metals from wastewater [41]. The efficiency of low-cost adsorbents can vary with the characteristics and composition of the material, the extent of surface modification, and the concentration of heavy metals in wastewater [42,43]. Numerous materials, such as agricultural waste, food waste, industrial by-products, and locally available natural materials, have been used as low-cost adsorbents [41,44–47]. One such material is sludge from different sources, which is readily abundant in large amounts, environmentally friendly, and sustainable, and is proven for heavy metal removal from wastewater [48,49].

Sludge is an adhesive, watery material produced during wastewater treatment processes [50]. It is a by-product of many industrial, agricultural, and municipal wastewater treatment processes and drinking water treatment processes. Most of the sludge is disposed of in landfills as the predominant sludge disposal method without taking proper use of the sludge while aggravating the solid waste disposal problem in some developed and developing countries [32,51–53]. Sludge is treated by several physical and chemical methods before the final disposal, which is labor-consuming and costly [54]. The available uses of sludge are limited to a few options, such as fertilizers, soil conditioning, metal recovery,

brick and ceramic production, and production of biogas, biodiesel, and electricity [55–57]. For example, Zhao et al. [58] reviewed sewage sludge-based biochar being utilized in soil remediation, carbon emission reduction, and water pollutant removal. Among all these uses, utilizing sludge for wastewater treatment is a novel concept that proved to be effective. Conversion of sludge into adsorbents is a sustainable alternative to the excessive production of sludge and expensive conventional adsorbents [32]. For instance, Kumar et al. [59] identified industrial by-products such as sludge are suitable for heavy metal adsorption. Furthermore, an emerging interest in employing sludge for heavy metal removal from wastewater can be observed according to recent studies [60–63].

Therefore, sludge-based adsorbents could be a promising technology to remove heavy metals from wastewater of different origins. Accordingly, this paper aims to discuss (1) the potential of using various sludge-based adsorbents for heavy metal removal from wastewater, (2) the effects of different factors on the adsorption of heavy metals into sludge-based adsorbents, (3) the mechanisms of adsorption of heavy metals into sludge-based adsorbents and (4) the methods to regenerate sludge-based adsorbents for heavy metal removal from wastewater.

2. Sludge-Based Adsorbents for Heavy Metal Removal from Wastewater

Sludge, a by-product of several anthropogenic activities, causes many solid waste problems due to excessive loads and lack of substantial uses. For instance, the sewage sludge generation in Southeast Asian countries is expected to increase from 24 to 40 million tons per year by 2050 due to increases in urban populations and wastewater discharge [64]. The total sludge production from 2007 to 2013 in China increased by 13%, leading to a dry-solid production of 6.25 million tons [65]. Nevertheless, there is a potential for using different types of sludge to remove heavy metals from wastewater by adsorption, as found by recent studies. Industrial sludge is generated as a by-product from various industrial activities in large quantities, such as the electroplating industry [60,61], the petrochemical industry [66], the metal recovery industry [67], etc. Drinking water treatment plant sludge produced during the treatment of both groundwater [68] and surface water [62] has also been applied as an adsorbent to remove heavy metals from wastewater. In addition, agricultural sludge from the palm oil industry [69] and sludge from wastewater treatment plants of animal farms [63] have been used to prepare adsorbents to remove heavy metals from wastewater. Municipal sewage sludge [70,71] has also been used in recent studies to remove heavy metals from wastewater. Therefore, studies have shown that the adsorption of heavy metals into agricultural sludge, industrial sludge, sewage sludge, and drinking water treatment plant sludge can be a promising, eco-friendly, and low-cost solution to remove heavy metals from wastewater and a solution for the solid waste problem [46,72–74].

2.1. Industrial Sludge

Electroplating sludge is an industrial sludge generated by the alkaline precipitation of electroplating wastewater. It consists of metals such as Ni^{2+}, Zn^{2+}, and Cu^{2+} in high concentrations that can effectively be recovered using bioleaching and electrodialysis [75,76]. Calcined electroplating sludge has more efficiently adsorbed Cu^{2+} in water than raw electroplating sludge, with a maximum adsorption capacity of 92 mg g^{-1} and 76.34 mg g^{-1}, respectively, by both surface adsorption and intraparticle diffusion [60]. Electroplating sludge modified by a facile calcination method has shown a removal efficiency of 87% of total Ni in nickel-containing electroplating wastewater [61]. Wajima [77] found that sulfur-impregnated paper sludge pyrolyzed at 400 °C removed 99.6% of Ni^{2+} at pH 7 and 100% of Pb^{2+} at pH 6. Sludge produced by wastewater treatment of LCD panel-display industries contains CeO_2, CaO, and SiO_2. Hong et al. [78] found that LCD sludge contains 26.48% CeO_2, 23.92% CaO, and 6.86% SiO_2 that could adsorb Cd^{2+}, Cu^{2+}, Pb^{2+}, and Ni^{2+} with maximum adsorption capacities of 1.29, 8.50, 8.47, and 3.02 mg g^{-1}, respectively, which are dominated by chemisorption. Oily sludge is produced in large quantities during crude oil production, storage, and refining processes [79]. For instance, over 5 million tons of oily

sludge are discharged annually in China [80]. Oily sludge of the petrochemical industry pyrolyzed at 750 °C has effectively removed Cr^{6+} with a maximum removal efficiency of 93.5% owing to the presence of a low-valence sulfur compound [81]. Cd^{2+}, Pb^{2+}, and Cu^{2+} in water have been adsorbed by pyrolyzed oily sludge of the petrochemical industry with a removal efficiency of 99.3%, 98.9%, and 99.4% at pH 9.79, 9.12 and 8.45, respectively, due to the formation of metal hydroxides and sulfides on the surface of the adsorbent [66]. Mining waste is another industrial sludge source that turns the mining industry toward waste-free production by using it for wastewater treatment in the same industry [82]. Borax sludge produced by sieving in the boron production process has been identified as a mixture of dolomite and tincalconite, which adsorbs Cr^{3+} from water and has best followed the Temkin isotherm model and pseudo-second-order model [83]. A study on the removal of heavy metals by iron ore sludge found that it can be used as a promising adsorbent for the simultaneous removal of heavy metals (As^{2+}, Mn^{2+}, Zn^{2+}, Cd^{2+}, and Pb^{2+}) in water, which was described as chemical adsorption by kinetic studies [67]. Red mud is an industrial by-product from the alkaline refining process that has been used for adsorption owing to the small particle size and large surface area [84]. Bai et al. found that red mud effectively removes Pb^{2+}, Cd^{2+}, and Cu^{2+}, with a maximum removal efficiency of 94.5%, 92.8%, and 78.1%, respectively [85].

2.2. Drinking Water Treatment Plant Sludge

Drinking water treatment plant sludge is generated during the coagulation and flocculation process of both surface water and groundwater treatment [86]. The common coagulating agents used in drinking water treatment are aluminum and iron salts [87]. Polyaluminum chloride coagulated surface water treatment sludge has been used as an effective adsorbent for removing Pb^{2+} and Cu^{2+} in synthetic stormwater, with a maximum adsorption capacity of 224.4 mg g^{-1} and 89 mg g^{-1}, respectively [88]. Fe and Al salt-flocculated surface water treatment sludge has adsorbed Co^{2+} from water with a maximum adsorption capacity of 17.31 mg g^{-1} by a spontaneous endothermic process that is favorable at high temperatures [89]. Maximum removal efficiencies of 97.4% and 96.6% have been obtained for Zn^{2+} and Cu^{2+} by alum sludge from a drinking water treatment plant at pH 6 [90]. Adsorption of Cu^{2+} into surface water treatment sludge modified by calcination has been well described by Freundlich and pseudo-first-order models [91]. Abo-El-Enein et al. [92] used surface water treatment sludge incinerated at 500 °C for the removal of Pb^{2+}, Cd^{2+} and Ni^{2+} and found that the metals are adsorbed in the order of $Pb^{2+} > Cd^{2+} > Ni^{2+}$. H_3PO_4-treated alginate gel-encapsulated surface water treatment sludge has effectively removed Cd^{2+} in aqueous solutions with a maximum adsorption capacity of 30 mg g^{-1} [93]. Ghorpade and Ahammed [94] used surface water treatment plant sludge coagulated by polyaluminum chloride and anionic polyacrylamide to remove Cu^{2+}, Co^{2+}, Cr^{6+}, Hg^{2+}, Pb^{2+}, and Zn^{2+} in aqueous solutions and in electroplating wastewater, where complete removal of Cu^{2+}, Hg^{2+}, and Zn^{2+} was obtained from column tests of real electroplating wastewater. Adsorption of Cd^{2+}, Cu^{2+}, and Zn^{2+} into surface water treatment sludge granulated with clay has best fitted the pseudo-second-order model, indicating chemical adsorption [62]. Groundwater treatment sludge has been used as an adsorbent to remove Cu^{2+} and Pb^{2+} in aqueous solutions [95]. Magnetized groundwater treatment sludge has effectively adsorbed Cu^{2+} in aqueous solutions following the pseudo-second-order and Langmuir isotherm models with a maximum adsorption capacity of 73.1 mg g^{-1}. The major adsorption mechanism has been cation exchange by H^+ and Na^+ on the sludge surface with Cu^{2+} in aqueous solutions [68]. Pyrolyzed ferric chloride coagulated groundwater treatment sludge was used to remove Cd^{2+}, Cu^{2+}, Ni^{2+}, and Pb^{2+} spiked in deionized water and desalination concentrate [96]. Kan et al. [97] used groundwater treatment sludge coated on silica sand to remove Cr^{6+} in water, which showed an adsorption capacity of 0.27 mg g^{-1} at pH 4.

2.3. Agricultural Sludge

Palm oil mill sludge has been used as an adsorbent for heavy metal removal from wastewater [98]. Pyrolyzed palm oil mill sludge at 400 °C has best removed Cu^{2+} and Cd^{2+} from the water, which has best fitted Langmuir and pseudo-second-order models, with maximum adsorption capacities of 48.8 and 46.2 mg g^{-1}, respectively [99]. Raw palm oil sludge has also been used to adsorb Cd^{2+} in aqueous solutions at pH 5.8 and recorded a maximum adsorption capacity of 18.49 mg g^{-1} in 60 min, which has been well described by Freundlich and pseudo-second-order models [69].

Sludge from wastewater treatment plants of animal husbandry has also been used to prepare adsorbents to remove heavy metals from wastewater [100]. Coagulated sludge pyrolyzed at 500 °C has shown a maximum adsorption capacity of 450.58 mg g^{-1} for Pb^{2+} at pH 5.6. The main adsorption mechanisms have been ion exchange and electrostatic interaction [100]. Coagulated sludge pyrolyzed at 500 °C has also adsorbed Cd^{2+} from aqueous solutions with a maximum adsorption capacity of 139.28 mg g^{-1} following the Langmuir isotherm model. The reasons for the higher adsorption have been the presence of metallic oxides, phosphate, and organic functional groups and the formation of new binding sites by pyrolysis [63]. Apart from that thiourea, treated pyrolyzed sludge effectively adsorbed Pb^{2+} in water with a maximum adsorption capacity of 143.13 mg g^{-1}, owing to the presence of various functional groups, such as –COO, –C=S, –C–NH_2, and R–SH [101].

2.4. Sewage Sludge

Sewage sludge is the residue produced by municipal wastewater treatment plants, especially from settling tanks. Sewage sludge-based adsorbents have been produced by carbonization, physical activation, and chemical activation [102]. Xue et al. [103] used pyrolyzed sewage sludge to remove Pb^{2+} and Cd^{2+} in aqueous solutions and achieved maximum adsorption capacities of 116.2 mg g^{-1} and 97.3 mg g^{-1}, respectively, at pH 7 and temperature of 20 °C. A study by Wang et al. [104] showed an affinity towards incinerated sewage sludge of Cu^{2+}, Cd^{2+}, and Zn^{2+} in the order of $Cu^{2+} > Cd^{2+} > Zn^{2+}$ at pH 6. The adsorption process has been best described by the Freundlich isotherm model, with corresponding adsorption capacities of 0.13 mmol g^{-1}, 0.11 mmol g^{-1}, and 0.06 mmol g^{-1} for Cu^{2+}, Cd^{2+}, and Zn^{2+}, respectively. Furthermore, cation exchange has been the major adsorption mechanism for all heavy metals [104]. Ho et al. [105] suggested electrostatic attraction, precipitation, surface complexation, and ion exchange as possible adsorption mechanisms for Pb^{2+} removal by anaerobically digested pyrolyzed municipal sewage sludge, which has shown a maximum adsorption capacity of 51.20 mg g^{-1}. The adsorption has best fitted the Langmuir isotherm model and pseudo-second-order model.

Ferric-activated pyrolyzed municipal sewage sludge has effectively removed Pb^{2+} in aqueous solutions with a maximum adsorption capacity of 42.96 mg g^{-1} at pH 5. The adsorption process has been well described by the Freundlich isotherm model and pseudo-second-order model. Yang et al. [106] observed that CaO-modified sewage sludge reached 99.74% removal efficiency for Cd^{2+} at pH 5 and 40 °C with a 1 g L^{-1} adsorbent dose within 90 min. A study on polyethyleneimine-modified sewage sludge exhibited a maximum adsorption capacity of 86.96 mg g^{-1} for Cr^{6+} at pH 5 and 25 °C, where the dominant adsorption mechanism was electrostatic interactions [107]. Li et al. [108] utilized thiol-functionalized pyrolyzed sewage sludge to remove heavy metals from aqueous solutions. The order of adsorption was $Pb^{2+} > Cd^{2+} > Cu^{2+} > Ni^{2+}$. The maximum adsorption capacity of each metal obtained at pH 5 resulted in 238.1 mg g^{-1} for Pb^{2+}, 96.2 mg g^{-1} for Cd^{2+}, 87.7 mg g^{-1} for Cu^{2+}, and 52.4 mg g^{-1} for Ni^{2+}. Nekooghadirli et al. [109] found that both Ni^{2+} and Pb^{2+} adsorption into $ZnCl_2$-treated sewage sludge best followed the Langmuir isotherm model, with a maximum adsorption capacity of 88.76 mg g^{-1} for Pb^{2+} and 74.06 mg g^{-1} for Ni^{2+} at 25 °C. They also observed that both metals show endothermic adsorption processes based on thermodynamic studies. Khosravi et al. [110] coated sewage sludge on TiO_2/ZnO nanofibers and hydrolyzed it as a practical modification to remove Ni^{2+} and Cu^{2+} from aqueous solutions, with maximum adsorption

capacities of 282.3 mg g^{-1} and 298.1 mg g^{-1} for Ni^{2+} and Cu^{2+}, respectively. Up to five regeneration cycles by 0.1 M HCl and over 90% removal efficiency of both metals were observed, indicating the accelerated regeneration capacity of the adsorbent.

KOH-treated pyrolyzed sewage sludge has been reported as having a higher adsorption capacity (57.48 mg g^{-1}) for Pb^{2+} when compared to CH_3COOK activation (47.59 mg g^{-1}) and CO_2 activation (22.40 mg g^{-1}). Physical adsorption has been dominant in Pb^{2+} adsorption, while chemical adsorption has been enhanced by oxygen-containing functional groups such as C=O [111]. A study by dos Reis et al. [112] on KOH-treated pyrolyzed sewage sludge found a maximum adsorption capacity of 31.85 mg g^{-1} for Cu^{2+} in aqueous solutions at pH 6 and temperature of 25 °C, which is relatively higher than the maximum adsorption capacity of $ZnCl_2$-activated sewage sludge (19.79 mg g^{-1}) and raw sewage sludge (3.513 mg g^{-1}). In addition, the same study found that $ZnCl_2$-activated sewage sludge recovers 98.9% of Cu^{2+}, while KOH-treated sewage sludge recovers 95.5% of Cu^{2+} after desorption by 1.5 mol L^{-1} HNO_3 [112]. A study on urea NaOH-modified carbonized sewage sludge effectively removed Cr^{6+} in water with a maximum adsorption capacity of 15.3 mg g^{-1} at 2.5 pH and temperature of 25 °C. The adsorption best fitted with the Langmuir isotherm model and pseudo-second-order model. The study also found that over 95% removal efficiency of Cr^{6+} could be obtained by regenerating the adsorbent with NaOH until five regeneration cycles [113].

Pyrolyzed sewage sludge-based biochar has been washed by HCl and HF, followed by CH_3COOK activation. The results of the study suggested that HF activation enhanced Pb^{2+} adsorption better than HCl activation [114]. The highest adsorption capacity was obtained by HF and CH_3COOK-activated biochar (49.47 mg g^{-1}), extremely higher than that of untreated biochar (7.56 mg g^{-1}) [114]. H_2SO_4-treated sewage sludge has been used to remove Cd^{2+} in aqueous solutions. The results showed that the adsorption process best fitted the pseudo-second-order model and Langmuir isotherm model, with a maximum adsorption capacity of 56.2 mg g^{-1} for Cd^{2+} [115]. Li et al. [71] showed that ion exchange has been the main adsorption mechanism of HNO_3-treated pyrolyzed sewage sludge where heavy metals with greater electronegativity are more attracted. The study also observed that the adsorbent removes Cu^{2+}, Zn^{2+}, and Al^{3+} from spiked natural acid rock drainage, with removal efficiencies of 98.9%, 42.6%, and 34.6%, respectively.

3. Physicochemical Properties of Sludge-Based Adsorbents

Physicochemical properties of sludge enhance the adsorption of heavy metals into sludge-based adsorbents. Treatment strategies and modifying agents utilized for the preparation of sludge-based adsorbents have a major impact on tailoring the physicochemical properties of the adsorbent [58,116]. For instance, sewage sludge-based adsorbents show up to 1800 m^2/g surface area due to chemical activation by alkali metal hydroxides such as KOH [102]. Studies have shown that sludge surface structure is usually irregular and amorphous (no clearly defined shape) [90,94]. Surface functional groups of sludge contribute to the adsorption of heavy metals such as nitrogen-containing (–NH– and $–NH_2$) and oxygen-containing functional groups (–COO–, –C=O, and –OH) that are abundant among sludge-based adsorbents such as sewage sludge, electroplating sludge, and drinking water treatment plant sludge [61,63,89,117,118]. Other functional groups, such as –C=C–, C=S, and S^{2-}, are also available in sludge [66,101,105]. The functional groups play an important role in the adsorption of heavy metal into sludge. For instance, drinking water treatment plant sludge has many functional groups, such as Al–OH, Fe–OH, and –OH bonds, contributing to the adsorption of heavy metals. Strong covalent bonds were observed between Fe–O and Al–O groups, with Co^{2+} forming inner- or outer-sphere complexes. –OH groups also contributed to the adsorption of Co^{2+} [89]. Oxygen functional groups such as –COO–, –OH, and C=C in sludge-activated carbon contribute to adsorbing Pb^{2+} from aqueous solutions [114]. Pyrolyzed sludge included oxygen-containing functional groups such as –C–OH, –COOH, –OH, Al–O, and Si–O, which may participate in the adsorption of Cd^{2+} by performing complexation reactions on the sludge surface [63]. Pb^{2+} adsorption into

amino-functionalized magnetic sludge biochar was performed by electrostatic interactions between Pb^{2+} with O–H, C=O, –NH– and $–NH_2$ on sludge, surface complexation between Pb^{2+} with functional groups forming –COO–Pb and –O–Pb complexes and precipitation of $Pb(OH)_2$ and $PbCO_3$ into the adsorbent surface [119]. Pyrolyzed oily sludge of the petrochemical industry adsorbed Cd^{2+}, Pb^{2+}, and Cu^{2+} in water by surface precipitation. It was induced by –OH and S^{2-} groups forming hydroxides and sulfides of heavy metals on the surface of sludge [66]. Adsorption of Pb^{2+} into anaerobically digested pyrolyzed sewage sludge suggested that the adsorption mechanism was governed by precipitation as phosphate and carbonate on the surface, ion exchange with Ca^{2+}, $Mg^{2+}$and K^{+}, electrostatic interactions, and surface complexation with –C=C–, –C=O, –COO–, and –OH functional groups [105]. KOH-treated pyrolyzed sewage sludge chemically adsorbed Pb^{2+} by forming mainly $PbCO_3$ by the oxygen-containing functional groups such as –C=O [111]. Pb^{2+} adsorption into thiourea-modified pyrolyzed sludge was carried out by coordination with –COO–, –C=S, and $–C–NH_2$ groups and ion exchange with R–SH and $–RSO_3^{-}$ groups [101]. Chemical adsorption of Pb^{2+} into carbonized sewage sludge was governed by collateral cooperation, electrostatic interactions, and ion exchange of functional groups such as –COO–, –C–N, and N–C–O [118]. Calcinated electroplating sludge adsorbed Ni^{2+} via electrostatic attraction and cation exchange caused by –COOH, –C–OH, and –OH and surface complexation between Ni^{2+} and –COO– [61]. Cu^{2+} adsorption into diethylenetriaminepentaacetic acid–chitosan-modified sewage sludge was conducted by surface complexation with amino and carboxyl functional groups of sludge [120]. Chemical adsorption of Cr^{6+}, Cu^{2+}, Se^{2+}, and Pb^{2+} into pyrolyzed sewage sludge–chitosan composite was carried out by various functional groups, such as $–NH_2$, –NH–, N–C=O, –C=O, and –OH [118].

4. Factors Affecting the Adsorption Process

Adsorption of heavy metals into sludge-based adsorbents depends on many factors, such as pH, temperature, initial heavy metal concentration, contact time, and adsorbent dose [12]. Studies have been carried out to check the effects of these factors on the adsorption process.

4.1. Effect of pH on Adsorption of Heavy Metals

The pH of the medium influences the adsorption process by affecting the adsorbent, adsorbate, and competition by H^{+} [12]. The dissociation of functional groups in adsorbents, ionization, and solubility of heavy metal ions depends on the pH of the medium [88,91]. The electric charge density on the adsorbent surface depends on the solution pH, and it improves or diminishes the electrostatic interactions between the sludge surface and metal ions [62].

In the acidic medium, the high competition caused by H^{+} reduces the binding of metal ions into adsorbents [67,109,114]. Adsorption of Cu^{2+} into calcined drinking water treatment sludge increased and maximized at pH 6.6 due to the reduction of H^{+} in the medium with the increase in pH from 2.8 to 11 at 20 °C by 0.25 g L^{-1} of adsorbent [91]. Goh et al. [99] found that both Cu^{2+} and Cd^{2+} ions were less favorably adsorbed by pyrolyzed palm oil sludge in the acidic medium in the range of pH 4 to 10 at 25 °C due to high competition of H^{+} ions in the medium, which are more mobile and more preferably adsorbed by sludge than metal ions. The maximum adsorption of Cu^{2+} (50 mg g^{-1}) and Cd^{2+} (49 mg g^{-1}) was pH 10 in that study. With the increase in pH from 3 to 8, Jiao et al. [89] observed that the removal of Co^{2+} by drinking water treatment sludge was lower in the acidic medium (pH 3) due to high H^{+} concentration and the maximum adsorption of 16.54 mg g^{-1} Co^{2+} was observed at pH 8. Pb^{2+} removal by ferric-activated pyrolyzed sewage sludge increasing from pH 2–5, with the highest removal efficiency of 98.46% of Pb^{2+} at pH 5, indicating the reduction of electrostatic repulsion between Pb^{2+} and positively charged adsorbent surface in the acidic medium [106]. Adsorption of Mn^{2+}, Pb^{2+}, Zn^{2+}, and Cd^{2+} into iron ore sludge increased with the reduction of competitive H^{+} ions in the medium, where the electrostatic attraction was the primary mechanism of adsorption and maximized adsorption at pH 8 [67].

pH at point of zero charge (pH_{PZC}) has been used to describe the effect of pH on the sludge-based adsorbent surface. pH_{PZC} is the pH of the medium at which the adsorbent surface charge is zero [12]. When the pH of the solution is greater than the pH_{PZC} of the adsorbent, the surface of the adsorbent is negatively charged, providing favorable conditions for cation adsorption [67,110]. The surface of the sludge-based adsorbents is negatively charged by the increase in pH [89,109,110]. An increase in pH values till optimum pH values of Ni^{2+} (pH 5) and Cu^{2+} (pH 6) increased the adsorption capacity of sewage sludge carbon-coated ZnO/TiO_2 nanofibers due to an increase in negatively charged ions on the adsorbent surface in a pH range of 2–8 at 25 °C [110]. The formation of $Cu(OH)_2$ and $Ni(OH)_2$ at higher pH values reduced the adsorption capacity of Cu^{2+} and Ni^{2+} into sewage sludge carbon-coated ZnO/TiO_2 nanofibers. In a basic medium with high pH values, the precipitation of metal ions reduces the adsorption capacity of adsorbents [89,91,99,119]. Duan and Fedler [88] observed that removing Pb^{2+} and Cu^{2+} from stormwater by drinking water treatment sludge at pH values greater than 6 was mainly due to precipitation. The formation of $Pb(OH)^+$ and $Pb(OH)_2$ caused the reduction of Pb^{2+} adsorption in the basic medium [119].

Nevertheless, the adsorption of Cr^{6+} into NaOH and urea-treated carbonized sewage sludge has been maximum in the acidic medium (pH 1–2.5) in a range of pH 1 to 8 at 25 °C due to the protonation of the adsorbent surface, making it positively charged at low pH, which enhances the electrostatic interaction of chromate ions [113]. A study by Wang et al. [107] found that Cr^{6+} adsorption into polyethyleneimine-modified sewage sludge has also decreased with the increase in pH from 3 to 8 due to changes in the adsorbent and Cr^{6+}. With the increase in pH, surface protonation is reduced, leading to a lessening in the electrostatic interactions between adsorbent and anions of Cr^{6+} such as $HCrO_4^-$, CrO_4^{2-}, and $Cr_2O_7^{2-}$. In the basic medium, anions of Cr^{6+} adsorption were reduced by the electrostatic repulsive effect of negatively charged sludge.

4.2. Effect of Temperature on Adsorption

An increase in the diffusion rate of metal ions and the creation of new binding sites by breaking of bonds on the adsorbent surface can be influenced by rising solution temperature [88,121]. Zhang et al. [114] found that pyrolyzed HF-treated sewage sludge increased the adsorption capacity of Pb^{2+} by 1.2 times with an increase in temperature from 15 °C to 45 °C. Both the adsorption capacity and removal efficiency of Cu^{2+} and Pb^{2+} into drinking water treatment sludge always increased with the increase in temperature from 20 °C to 40 °C at different initial concentrations of Cu^{2+} (60–140 mg L^{-1}) and Pb^{2+} (120–380 mg L^{-1}) because of the higher mass transfer rate and higher reaction rate induced by the increased temperature, indicating the adsorption process is endothermic [88]. Shahin et al. [91] found that Cu^{2+} removal efficiency by calcined drinking water treatment sludge also increased with an increase in temperature from 20 °C to 80 °C by 84% to 90% at pH 4.8 by the increase in mobility of Cu^{2+} and the number of molecules that have sufficient energy to bind with active sites. An increase in adsorption with the increase in solution temperature also indicates that the particular adsorption process is endothermic, which is favorable at higher temperatures [88,91]. Therefore, wastewater treatment by adsorption could be conveniently carried out at room temperature in tropical countries where monthly mean air temperatures vary between 24 °C and 27 °C throughout the year [122].

Adsorption Thermodynamics

Adsorption thermodynamics are used to determine the feasibility of the adsorption process [12]. Negative Gibbs free-energy values indicate a spontaneous and feasible adsorption process [69]. Positive enthalpy values suggest that the adsorption is an endothermic process [88,99]. Negative enthalpy changes indicate the adsorption process is exothermic [69]. Endothermic processes absorb energy, while exothermic processes release energy into the environment. A positive entropy value indicates an increase in randomness of the

solid–solution interface [99,110,123]. Gibbs free energy, enthalpy, and entropy are some of such thermodynamic parameters.

Adsorption thermodynamic parameters can be calculated using Equations (1)–(4).

$$K_d = \frac{C_o - C_e}{C_e} \times \frac{V}{m} \quad (1)$$

$$G^O = -RTlnK_d \quad (2)$$

$$\Delta G^O = \Delta H^o - T\Delta S^o \quad (3)$$

$$lnK_d = \frac{S^o}{R} - \frac{H^o}{RT} \quad (4)$$

where ΔG^O is Gibbs free energy (J/mol), ΔH^o is enthalpy (J/mol), and ΔS^o is entropy (J/mol/K), K_d is the distribution coefficient (L/g), R is the universal gas constant (8.314 J/mol/K), and T is the adsorption temperature (K) [88,123].

Adsorption of Pb^{2+} and Cu^{2+} into drinking water treatment sludge under acidic conditions and temperatures of 20 °C, 30 °C, and 40 °C indicate a spontaneous and feasible adsorption process [88]. A study conducted by Ong et al. [123] investigated that Ni^{2+} adsorption into recovered MnO_2 from groundwater treatment sludge was spontaneous, thermodynamically favorable, and endothermic. When the adsorption process is endothermic, adsorption can be enhanced by increasing temperature. Khosravi et al. [110] found that the adsorption of Ni^{2+} and Cu^{2+} into hydrolyzed sewage sludge-coated nanofibers is a spontaneous endothermic process where the randomness of the solid–solution interface was increased during adsorption. Pyrolyzed palm oil mill sludge adsorption processes of both Cu^{2+} and Cd^{2+} were spontaneous, thermodynamically feasible endothermic processes [99]. Nekooghadirli et al. [109] studied Pb^{2+} and Ni^{2+} adsorption into $ZnCl_2$-activated pyrolyzed sewage sludge and found a favorable adsorption at higher temperatures that was an endothermic process. Increased randomness at the adsorbent–adsorbate interface during the adsorption was indicated by the positive entropy values, which means an increase in disorder. Adsorption of Cd^{2+} into palm oil sludge was a spontaneous, exothermic, and feasible adsorption process [69]. These results indicate the feasibility of using sludge as an adsorbent for heavy metal removal from wastewater. The sludge-based adsorbents that showed an endothermic adsorption process can be effectively used in wastewater treatment at high temperatures (20 °C to 40 °C) [88,99,109,110].

4.3. Effect of Initial Metal Ion Concentration on Adsorption

Adsorption of heavy metals into drinking water treatment sludge increases with the initial heavy metal concentration [88]. With the increase in initial Cd^{2+} concentration from 1 to 5 mg L^{-1} at pH 6, the adsorption capacity of Cd^{2+} into H_2SO_4-treated sewage sludge increased up to a level after which adsorption capacity decreased, indicating the saturation of binding sites in sewage sludge with Cd^{2+} ions [115]. A study by Shahin et al. [91] found that the adsorption of Cu^{2+} into calcined drinking water treatment sludge was directly proportional to the initial Cu^{2+} concentration, shown by the increase in adsorption capacity increasing from 1.90 to 35.5 mg g^{-1} with an increase in initial concentration from 50 to 1000 mg L^{-1}. It may be induced by the enhancement of Cu^{2+} mass transfer. In this study, a low concentration of Cu^{2+} (50 mg L^{-1}) reached the maximum adsorption into calcined drinking water treatment sludge instantaneously. Ong et al. [123] observed an increase in the Ni^{2+} adsorption capacity of MnO_2 recovered from groundwater treatment sludge with an increase in initial Ni^{2+} concentration (10-100 mg L^{-1}), which was reduced to a slight increase at higher Ni^{2+} concentration (100–200 mg L^{-1}) due to electrostatic repulsion between Ni^{2+} bound on the binding sites and Ni^{2+} in aqueous solutions. Higher Cr^{6+} concentrations ranging from 20 to 50 mg L^{-1} caused higher adsorption of Cr^{6+} into

groundwater treatment sludge coated on silica sand than lower Cr^{6+} concentrations from 1 to 10 mg L^{-1} [97]. Therefore, it can be concluded that the adsorption capacity of sludge-based adsorbents increases with the increase in the initial concentration of heavy metals due to the abundance of heavy metals in aqueous solutions. Nevertheless, at some point, the adsorption of heavy metals reaches maximum adsorption capacity, after which the adsorption process is stabilized due to the saturation of binding sites of sludge with heavy metals [91,97,115].

The removal efficiency of sludge-based adsorbents reduces with the increase in metal ion concentration due to the lack of binding sites compared to metal ions present in aqueous solutions [91,93]. For example, the removal efficiency of Cd^{2+} and Ni^{2+} by fired drinking water treatment sludge has been reduced rapidly with an increase in the initial concentration of Cd^{2+} and Ni^{2+} from 100 to 650 mg L^{-1} for both metals. Almost 100% removal has been reported by Cd^{2+} and Ni^{2+} at the optimum metal concentration of 100 mg L^{-1} at pH 5.5. Nevertheless, Pb^{2+} was completely removed from aqueous solutions regardless of the initial concentration increase (100–650 mg L^{-1}) in the same study [92]. Siswoyo et al. [93] studied the removal efficiency of Cd^{2+} into H_3PO_4-treated drinking water treatment sludge and found that the removal efficiency decreased from 74% to 30% with the increase in initial Cd^{2+} concentration from 25 to 250 mg L^{-1} due to the saturation of binding sites. Since the removal efficiency of sludge-based adsorbents reduces with the increase in metal ion concentration, it is important to add an adequate amount of sludge depending on the available heavy metal concentration in wastewater to increase the effectiveness of the adsorption process.

4.3.1. Adsorption Isotherm Models

Isotherm models describe the relationship between the equilibrium concentration of adsorbate in the liquid phase and the equilibrium amount of adsorbate adsorbed into the solid adsorbent. These models could be used for the investigation of adsorption mechanisms, maximum adsorption capacity, and the properties of the adsorbent [124]. Adsorption isotherm models such as Langmuir and Freundlich isotherm models are used to describe the adsorption process of adsorbents [112,125].

Langmuir Isotherm Model

The Langmuir isotherm model describes monolayer adsorption into a homogeneous surface with identical active sites [60,89,105,120]. The nonlinear form of the Langmuir isotherm equation is shown in Equation (5).

$$q_e = \frac{q_m b C_e}{1 + b C_e} \tag{5}$$

where q_e (mg g^{-1}) is the adsorption capacity at equilibrium; q_m is the maximum adsorption capacity (mg g^{-1}); b is the Langmuir isotherm constant (L mg^{-1}); and C_e is the equilibrium concentration (mg L^{-1}) [63,88,126].

Dimensionless constant (R_L) is calculated from Equation (6) using the Langmuir isotherm constant (b) and initial metal ion concentration (C_0). R_L indicates the favorability of adsorption. If R_L is > 1, adsorption is unfavorable. If R_L = 1, adsorption is linear. If 1 > R_L > 0, adsorption is favorable. If R_L = 0, adsorption is irreversible [88,109,123].

$$R_L = \frac{1}{1 + b C_0} \tag{6}$$

Many heavy metal removal processes of sludge-based adsorbents have been fitted well with the Langmuir isotherm model. A study by Duan and Fedler [88] found that the adsorption of both Pb^{2+} and Cu^{2+} into drinking water treatment sludge is monolayer adsorption by homogeneously distributed active sites with maximum adsorption capacities of 89 mg g^{-1} for Cu^{2+} and 224.4 mg g^{-1} for Pb^{2+} at pH 4 and pH 5, respectively. The

dimensionless constant (R_L) of both ions indicated that the adsorption process is favorable. Zhu et al. [68] found the adsorption of Cu^{2+} into magnetized groundwater treatment sludge signified monolayer adsorption with a maximum adsorption capacity of 73.1 mg g^{-1} within two hours at pH 5. Co^{2+} adsorption into drinking water treatment sludge suggested a homogeneous distribution of binding sites on the sludge surface with a maximum adsorption capacity of 17.3 mg g^{-1} [89]. Another study investigated the adsorption of metal ions Cu^{2+}, Zn^{2+}, and Cd^{2+} into clay-added drinking water treatment sludge granules via monolayer adsorption with maximum adsorption capacities of 1.23 mg g^{-1} for Zn^{2+}, 1.53 mg g^{-1} for Cd^{2+}, and 2.76 mg g^{-1} for Cu^{2+}, respectively. The maximum adsorption capacity has been highest in Cu^{2+} compared to Cd^{2+} and Zn^{2+} [62]. Adsorption of Ni^{2+} into recovered MnO_2 from groundwater treatment sludge was favorable according to R_L values under acidic conditions and reached a maximum adsorption capacity of 145.56 mg g^{-1} at 25 °C [123]. Siswoyo et al. [93] used $H_3PO_4^-$-activated drinking water treatment sludge and raw drinking water treatment sludge for the adsorption of Cd^{2+} from water and found adsorption capacities of 40.2 mg g^{-1} and 24.9 mg g^{-1}, respectively, suggesting that H_3PO_4 activation enhanced the adsorption capacity of drinking water treatment sludge. Furthermore, encapsulation studies found that both alginate gel encapsulated and non-encapsulated forms for adsorption of Cd^{2+} fit well with the Langmuir model, with a decrease in maximum adsorption capacity due to encapsulation. A study by Ho et al. [105] found that the Pb^{2+} adsorption into anaerobically digested pyrolyzed sewage sludge is monolayer adsorption where the sludge surface has identical adsorption capacity and homogeneous distribution of active sites with a maximum adsorption capacity of 51.2 mg g^{-1}. In addition, with the elevation in temperature from 293 K to 313 K, the maximum adsorption capacity was increased from 49.93 mg g^{-1} to 53.96 mg g^{-1}, suggesting an improvement in adsorption with a temperature rise. Cai et al. [63] reported that Cd^{2+} adsorption into pyrolyzed sludge inferred a favorable, monolayer, and homogeneous process with a maximum adsorption capacity of 139.28 mg g^{-1}, suggesting the suitability of applying sludge for Cd^{2+} removal from wastewater. Pb^{2+} adsorption into thiourea-modified pyrolyzed sludge was monolayer adsorption with a maximum adsorption capacity of 143.13 mg g^{-1}, which could occur due to the presence of sulfur-containing functional groups enhancing chemical adsorption [101]. A study conducted by Goh et al. [99] found that Cu^{2+} and Cd^{2+} adsorption into pyrolyzed palm oil mill sludge achieved maximum adsorption capacities of 48.8 mg g^{-1} and 46.2 mg g^{-1}, respectively, indicating monolayer adsorption where the adsorbent surface has similar adsorption sites. Sludge of the petrochemical industry has effectively removed Cd^{2+}, Cu^{2+}, and Pb^{2+} in aqueous solutions with the maximum adsorption capacities of 106.16, 128.04, and 140.65 mg g^{-1}, respectively, indicating monolayer adsorption in the ascending order of $Cd^{2+} < Cu^{2+} < Pb^{2+}$ [66].

Freundlich Isotherm Model

The Freundlich isotherm model describes the adsorption into a heterogeneous adsorbent surface with active sites with different adsorption energies [69,88,117,120]. The nonlinear equation of the Freundlich isotherm model is given in Equation (7).

$$q_e = K_F\, C_e^{1/n} \tag{7}$$

where q_e (mg g^{-1}) is the adsorption capacity at equilibrium; K_F (L mg^{-1}) is the Freundlich isotherm constant; C_e is the equilibrium concentration (mg L^{-1}), and the dimensionless empirical parameter (n) [69,88,109,126].

Some heavy metal adsorption processes of sludge-based adsorbents have been fitted well with the Freundlich isotherm model. Lee et al. [69] found that the adsorption of Cd^{2+} into palm oil sludge has shown a high adsorption favorability and a heterogeneous surface of palm oil sludge with a maximum adsorption capacity of 18.49 mg g^{-1} at pH 5.8 and initial concentration of 200 mg L^{-1}. A study conducted by Wang et al. [104] on incinerated sewage sludge ash suggested favorable multilayer adsorption into heterogeneous adsorption sites with maximum adsorption capacities of 0.13, 0.11, and 0.06 mmol g^{-1} for Cu^{2+}, Cd^{2+},

and Zn^{2+}, respectively. Yang et al. [106] found that the surface of ferric-activated sewage sludge into which Pb^{2+} was adsorbed by multilayer heterogeneous adsorption obtained a maximum adsorption capacity of 42.96 mg g^{-1}. Table 1 shows different types of sludge-based adsorbents that have been used for heavy metal adsorption from water. Further, it shows the potential of sludge-based adsorbents with different treatments to remove heavy metals from wastewater and the nature of the adsorption process. A study by Kan et al. [97] investigated the adsorption of Cr^{6+} into groundwater treatment sludge coated on silica sand indicating the presence of heterogeneous and active sites that are exponentially distributed on the adsorbent surface. Lin et al. [96] studied the adsorption of Cd^{2+} ions in deionized water and found that the multilayers of Cd^{2+} ions precipitated on the surface of pyrolyzed groundwater treatment sludge. The adsorption fit well with the Freundlich model, showing the possibility of developing it as a potential biosorbent for removing heavy metals from water. A study by Song et al. [117] found that the adsorption of Cr^{6+}, Cu^{2+}, Se^{2+}, and Pb^{2+} into modified anaerobic sewage sludge was multi-layered adsorption, which was obtained a Freundlich constant (n) greater than 1, suggesting the process is favorable.

Table 1. Sludge-based adsorbents with different treatments to remove heavy metals and the nature of adsorption.

Raw Material	Treatment for Adsorbent	Best-Fitted Isotherm Model	Nature of Adsorption	Maximum Adsorption Capacity (mg g^{-1})	Reference
Surface water treatment sludge	Drying at 105 °C	Langmuir	monolayer adsorption	Pb^{2+}: 224.4 Cu^{2+}: 89	[88]
Surface water treatment sludge	Air drying	Langmuir	monolayer adsorption	Co^{2+}: 17.3	[89]
Groundwater treatment sludge	Recovery of MnO_2 from sludge	Langmuir	monolayer adsorption	Ni^{2+}: 145.56	[123]
Surface water treatment sludge	Adding clay and making granules	Langmuir	monolayer adsorption	Cd^{2+}: 1.53 Cu^{2+}: 2.76 Zn^{2+}: 1.23	[62]
Groundwater treatment sludge	Magnetization by hydrothermal method	Langmuir	monolayer adsorption	Cu^{2+}: 73.1	[68]
Oily sludge from the Petrochemical industry	Pyrolysis at 750 °C	Langmuir	monolayer adsorption	Cd^{2+}: 106.16 Pb^{2+}: 140.65 Cu^{2+}: 128.04	[66]
Electroplating sludge	Calcination at 500 °C	Langmuir	monolayer adsorption	Cu^{2+}: 91	[60]
Iron Ore Sludge	Drying at 80–105 °C	Langmuir	monolayer adsorption	Pb^{2+}: 1.305 As^{2+}: 1.113 Cd^{2+}: 0.771 Zn^{2+}: 0.745 Mn^{2+}: 0.710	[67]
Tannery sludge	Pyrolysis at 800 °C	Langmuir	monolayer adsorption	Cr^{6+}: 352	[127]

Table 1. *Cont.*

Raw Material	Treatment for Adsorbent	Best-Fitted Isotherm Model	Nature of Adsorption	Maximum Adsorption Capacity (mg g^{-1})	Reference
Sewage sludge	Activation by $ZnCl_2$ and pyrolysis at 600 °C	Langmuir	monolayer adsorption	Ni^{2+}: 74.06 Pb^{2+}: 88.76	[109]
Sewage sludge	Anaerobic digestion and Pyrolysis at 600 °C	Langmuir	monolayer adsorption	Pb^{2+}: 51.2	[105]
Sewage sludge	Pyrolysis at 400 °C	Langmuir	monolayer adsorption	Pb^{2+}: 116.2 Cd^{2+}: 97.3	[103]
Sewage sludge	Activation by KOH, and pyrolysis at 700 °C	Langmuir	monolayer adsorption	Pb^{2+}: 57.48	[111]
Sewage sludge	Urea and NaOH modification and carbonized at 850 °C	Langmuir	monolayer adsorption	Cr^{6+}: 15.3	[113]
Sewage sludge	hydrothermal treatment at 120 °C	Langmuir	monolayer adsorption	Pb^{2+}: 62.441	[118]
Sewage sludge	H_2SO_4 treatment	Langmuir	monolayer adsorption	Cd^{2+}: 56.2	[115]
Sewage sludge and sugarcane bagasse	KOH treatment	Langmuir	monolayer adsorption	Pb^{2+}: 137.12	[128]
Palm oil mill sludge	Pyrolysis at 400 °C	Langmuir	monolayer adsorption	Cu^{2+}: 48.8 Cd^{2+}: 46.2	[99]
Palm oil mill sludge	Drying at 80 °C	Freundlich	multi-layered adsorption	Cd^{2+}: 18.49	[69]
Sewage sludge	Activation by $FeSO_4$ and pyrolysis at 750 °C	Freundlich	multi-layered adsorption	Pb^{2+}: 42.96	[106]

4.4. Effect of Contact Time on Adsorption

Contact time is an important parameter that affects the adsorption process. The adsorption capacity and removal efficiency of sludge-based adsorbents vary with the time that the adsorbent is in contact with metals in water. Duan and Fedler [88] found that the adsorption of Pb^{2+} and Cu^{2+} into drinking water treatment sludge showed a sharp increase in the initial two hours and stabilized afterward at pH 5 and initial concentration of 160 mg L^{-1} for Pb^{2+} and 100 mg L^{-1} for Cu^{2+} due to the abundance of adsorption sites. A study on Cu^{2+} adsorption into diethylenetriaminepentaacetic acid–chitosan-modified sewage sludge showed a rapid increase in adsorption capacity during the first 60 min due to the abundance of free binding sites and then reached equilibrium adsorption capacity of 19.22 mg g^{-1} for 10 mg L^{-1} and 25.77 mg g^{-1} for 20 mg L^{-1} initial Cu^{2+} concentration, indicating the saturation of binding sites with Cu^{2+} [120]. Adsorption rates of Cd^{2+}, Cu^{2+}, and Zn^{2+} by incinerated sewage sludge ash elevated with contact time till 15 min and reached an equilibrium state after 720 min, indicating the saturation of the adsorption sites and repulsive forces between the metal ions [104]. In the same study, the adsorption capacity of Cd^{2+} was 0.060 mmol g^{-1} after 60 min with an initial concentration of 0.45 mmol L^{-1}.

Goh et al. [99] found that the removal efficiency of heavy metals, Cd^{2+} and Cu^{2+}, by pyrolyzed palm oil mill sludge could be raised by increasing the contact time, which enhances the contact between sludge and Cd^{2+} and Cu^{2+}. Nevertheless, after reaching the equilibrium after 240 min, the increase in removal was reduced due to the saturation

of adsorption sites and repulsion between the adsorbed and free metal ions in water. The maximum adsorption capacities obtained by Cu^{2+} and Cd^{2+} at the equilibrium are 48.8 mg g^{-1} and 46.7 mg g^{-1}, respectively. A study on fired drinking water treatment sludge by Abo-El-Enein et al. [92] investigated the rapid removal of heavy metals that was induced by the abundance of heavy metal adsorption sites and found that the equilibrium was obtained after 4 h with the removal efficiencies of 100% for Pb^{2+}, 91% for Cd^{2+}, and 78% for Ni^{2+} at pH 5.5 and an initial concentration of 100 mg L^{-1} of each metal. A study conducted by Zhang et al. [113] found that 99% removal efficiency of Cr^{6+} could be obtained by urea- and NaOH-modified carbonized sewage sludge within 30 min, which could not be achieved by carbonized sewage sludge and urea-added carbonized sewage sludge that reached only 45% and 80% removal efficiency, respectively, after two hours, suggesting that equilibrium contact time depends on the properties of sludge-based adsorbent. Therefore, when applying sludge for wastewater treatment, the efficiency of the adsorption process could be improved by evaluating the minimum time it takes to reach the maximum adsorption of desired heavy metals to be removed.

4.4.1. Adsorption Kinetic Models

Adsorption kinetic models are used to investigate the performance of the adsorbent, the adsorption rate, and the mechanism of mass transfer processes in adsorption [129]. Adsorption kinetic models such as the pseudo-first-order model and pseudo-second-order model are used to describe the adsorption process of sludge-based adsorbents [68,88,110].

Pseudo-First-Order Model

The pseudo-first-order model indicates that the rate-limiting mechanism of the adsorption process is physical adsorption, which occurs by diffusion through the adsorbent interface [91,123,130]. The non-linear equation of the pseudo-first-order model is given in Equation (8).

$$q_t = q_e \left(1 - e^{-k_1 t}\right) \quad (8)$$

where q_e (mg g^{-1}) is the adsorption capacity at equilibrium; q_t (mg g^{-1}) is the adsorption at the time (t) per unit mass of adsorbent; k_1 (h^{-1}), is the rate constant [88,111].

Zhang et al. [111] found that the adsorption of Pb^{2+} into KOH-activated pyrolyzed sewage sludge followed the pseudo-first-order model and reached an equilibrium adsorption capacity of 10.27 mg g^{-1} at 35 min with an initial concentration of 200 mg L^{-1}. It indicates the suitability of chemical activation by KOH for the preparation of sludge-based adsorbents. Cd^{2+} and Pb^{2+} adsorption into KOH-treated pyrolyzed sewage sludge best fits the pseudo-first-order model with equilibrium adsorption capacities of 32.3 mg g^{-1} and 41.2 mg g^{-1} for each metal, respectively, at an initial concentration of 100 mg L^{-1} [103]. A study by Shahin et al. [91] found that Cu^{2+} adsorption into calcinated drinking water treatment sludge also followed the pseudo-first-order model, indicating the dominant adsorption process is physical adsorption in which the rate-controlling step depends on collisions between heavy metal ions with unoccupied binding sites on the surface of sludge. Duan and Fedler [88] found Pb^{2+} adsorption into drinking water treatment sludge at pH 5, 20 °C with an initial concentration of 160 mg L^{-1} achieved an equilibrium adsorption capacity of 136.9 mg g^{-1} and a rate constant of 1.511 h^{-1}, suggesting that drinking water treatment sludge could rapidly absorb Pb^{2+} from stormwater under acidic conditions.

Pseudo-Second-Order Model

The pseudo-second-order model describes the chemisorption of heavy metal ions into adsorbents related to the exchange or sharing of electrons between ions and the adsorbent [119]. The pseudo-second-order model provides the best correlation of the

experimental data when a chemical reaction is the rate-controlling step [131]. The non-linear form of the pseudo-second-order model is given in Equation (9).

$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t} \quad (9)$$

where q_e (mg g^{-1}) is the adsorption capacity at equilibrium; q_t (mg g^{-1}) is the adsorption at the time (t) per unit mass of adsorbent; and k_2 (g mg^{-1} h^{-1}) is the rate constant [68,88,106].

Many heavy metal removal processes of sludge-based adsorbents were described well by the pseudo-second-order model. Jiao et al. [89] found Co^{2+} adsorption into drinking water treatment sludge and found that the rate-controlling step mechanism is chemical adsorption with an equilibrium adsorption capacity of 16.72 mg g^{-1} and a rate constant of 0.037 g mg^{-1} h^{-1} in 30 h at an initial concentration of 500 mg L^{-1} and pH 6. Adsorption of Cu^{2+} into magnetized groundwater treatment sludge particles also followed the pseudo-second-order model at pH 5 and Cu^{2+} initial concentration of 50 mg L^{-1}, indicating chemisorption by valence forces through the exchange of electrons between Cu^{2+} and adsorbent [68]. A study conducted by Du et al. [62] has investigated that clay-added drinking water treatment sludge granules best fitted with the pseudo-second-order model for the adsorption of metal ions Cu^{2+}, Zn^{2+}, and Cd^{2+} at pH 5 and initial concentration of 10 mg L^{-1} which suggested that the rate-limiting step was chemical adsorption with electron sharing between the metal ions and the adsorbent surface. Furthermore, the initial adsorption rate increased in the order of Cd^{2+} < Zn^{2+} < Cu^{2+} with equilibrium adsorption capacities of 0.28, 0.33, and 0.39 mg g^{-1} and rate constants of 0.07, 0.12, and 0.13 g mg^{-1} min^{-1} for each metal, respectively. Adsorption of Ni^{2+} into recovered MnO_2 from groundwater treatment sludge was best described by the pseudo-second-order model, indicating it is chemical adsorption by electron exchange or sharing between Ni^{2+} and active sites [123]. Accordingly, different initial concentrations of Ni^{2+} have best followed pseudo-second-order model with increasing equilibrium adsorption capacities of 24.93, 71.68, 107.64, and 137.74 mg g^{-1} and decreasing rate constants of 5.66, 0.0018, 0.0009, and 0.0004 g mg^{-1} min^{-1} for initial concentrations of 10, 50 100, and 200 mg L^{-1}, respectively. The decrease in rate constants indicates the increase in competition between metal ions in binding to active sites on sludge surfaces. Yang et al. [106] have conducted kinetic studies on $FeSO_4^-$ activated pyrolyzed sewage sludge for Pb^{2+} adsorption at pH 5 and an initial concentration of 50 mg L^{-1}, which followed the pseudo-second-order model indicating the rate-limiting step of the Pb^{2+} adsorption process is chemical adsorption. The study achieved an equilibrium adsorption capacity of 42.96 mg g^{-1} and a rate constant of 0.655 g mg^{-1} min^{-1}, suggesting the suitability of $FeSO_4$ for the activation of sewage sludge as a heavy metal adsorbent. Kinetic studies of Ni^{2+} and Cu^{2+} adsorption into hydrolyzed sewage sludge-coated nanofibers were conducted by Khosravi et al. [110]. In that study, with an initial concentration of 50 mg L^{-1} for each metal at 25 °C, Ni^{2+}, and Cu^{2+} could obtain equilibrium adsorption capacities of 80.2 and 98.1 mg g^{-1} and rate constants of 0.0028 and 0.00365 g mg^{-1} min^{-1} for each metal, respectively. The rate-limiting step of Pb^{2+} and Ni^{2+} adsorption into $ZnCl_2^-$ activated pyrolyzed sewage sludge was chemical adsorption [109], where the equilibrium adsorption capacities of both metals increased with the increase in initial concentration of heavy metal as 50, 100, and 200 mg L^{-1}. The respective equilibrium adsorption capacities at each initial concentration were 25.68, 49.89, and 93.37 mg g^{-1} for Pb^{2+} and 14.07, 27.4, and 54.47 mg g^{-1} for Ni^{2+} adsorption. Goh et al. [99] have conducted a study on the adsorption of Cu^{2+} and Cd^{2+} into pyrolyzed palm oil mill sludge which has achieved equilibrium adsorption capacities of 51.81 and 58.48 mg g^{-1} and rate constants of 0.0006 and 0.0003 g mg^{-1} min^{-1} for each metal, respectively, by following pseudo-second-order model, indicating the dominance of chemisorption in the adsorption process, which takes a longer time to complete than physical adsorption. Chemisorption causes the formation and destruction of chemical bonds, which requires larger activation energy [132]. Table 2 shows examples of some

kinetic studies on different sludge-based adsorbents for heavy metal adsorption from wastewater, showing the mechanism of adsorption observed in each study.

Table 2. Sludge-based adsorbents to remove heavy metals by different adsorption mechanisms.

Adsorbent	Heavy Metal/s	Adsorption Capacity at Equilibrium (mg g^{-1})	Best-Fitted Kinetic Model	Mechanism of Adsorption	Reference
Pyrolyzed sewage sludge	Cd^{2+}	32.3	Pseudo-first-order model	Physical adsorption	[103]
	Pb^{2+}	41.2			
KOH-activated pyrolyzed sewage sludge	Pb^{2+}	-			[111]
Surface water treatment sludge	Pb^{2+}	136.9			[88]
	Cu^{2+}	54.5			
Surface water treatment sludge	Co^{2+}	16.72	Pseudo-second-order model	Chemical adsorption	[89]
Pyrolyzed sludge of Petrochemical industry	Cd^{2+}	39.67			[66]
	Pb^{2+}	44.35			
	Cu^{2+}	40.27			
Pyrolyzed palm oil mill sludge	Cu^{2+}	51.81			[99]
	Cd^{2+}	58.48			
Calcinated electroplating sludge	Cu^{2+}	38.46			[60]
Clay-added drinking water treatment sludge	Cu^{2+}	0.39			[62]
	Zn^{2+}	0.33			
	Cd^{2+}	0.28			
Urea and NaOH-modified sewage sludge	Cr^{6+}	13.351			[113]

4.5. Effect of Adsorbent Dose on Adsorption

The removal efficiency of heavy metals by sludge-based adsorbents increases with the increase in adsorbent dose due to the increase in adsorption sites [91,99,119,121]. A study conducted by Goh et al. [99] found that pyrolyzed palm oil mill sludge could increasingly remove Cd^{2+} and Cu^{2+} from water with the rise in the adsorbent dose from 0.1 g L^{-1} to 0.6 g L^{-1} at 25 °C, which conveyed the provision of more binding sites for heavy metals. Further, the optimum adsorbent dose was 0.05 g, which could obtain a removal efficiency of 99% for Cu^{2+} and 98% for Cd^{2+} that remained unchanged with further increases in adsorbent dosage. Abo-El-Enein et al. [92] studied the adsorption of Cd^{2+} and Ni^{2+} into burnt drinking water treatment sludge and found that the adsorption reached the maximum removal efficiency of 99% for each heavy metal at an optimum adsorbent dosage of 40 g L^{-1} in a range of 10 to 40 g L^{-1} at pH 5.5. Another study on drinking water treatment sludge by Ghorpade and Ahammed [94] found that the increase in adsorbent dose from 0.5 to 10 g L^{-1} could result in 100% removal efficiency of Pb^{2+} and 89–94% removal efficiency of Cu^{2+}. Further, the removal efficiency for Hg^{2+} and Zn^{2+} was 97% at 10 g L^{-1} adsorbent dose. The increase in percentage removal could be due to the abundance of adsorption sites [92,94]. The adsorption capacity of amino-functionalized magnetic sludge-biochar for Pb^{2+} has been reduced from 163.48 to 14.97 mg g^{-1} with the increase in adsorbent dose from 0.08 to 0.8 g L^{-1} at 25 °C and 15 mg L^{-1} of Pb^{2+} due to the reduction of Pb^{2+} in water by adsorption of increasing binding sites in a study conducted by Huang et al. [119]. In addition, the

removal efficiency of Pb^{2+} increased rapidly at the beginning and reached a maximum removal efficiency of 98.15% at an adsorbent dose of 0.32 g L^{-1} after reaching the maximum removal efficiency, the process decreased due to the aggregation of adsorbent, which leads to interference between the binding sites and metal ions by excess adsorbents. An increase in the dose of pyrolyzed oily sludge of the petrochemical industry elevated the removal efficiency of Cd^{2+}, Pb^{2+}, and Cu^{2+} and increased the pH, suggesting the formation and precipitation of metal hydroxides on the adsorbent surface [66]. Moreover, the optimum adsorbent dosage for Pb^{2+} and Cu^{2+} was 0.8 g L^{-1}, which led to removal efficiencies of 98.9 and 99.4% for the two metals, respectively. Nevertheless, the optimum adsorbent dosage for Cd^{2+} was 1 g L^{-1} with 99.3% removal efficiency. A study on pyrolyzed sewage sludge found an increase in the removal of Cd^{2+}, Pb^{2+}, Cu^{2+}, and Mn^{2+} with an increase in sludge dose with a removal efficiency of 95.5%, 98%, 97.6%, and 91.5%, respectively, at an optimum adsorbent dosage of 1 mg L^{-1}. This implies the availability of more functional groups and adsorbent surface area, enhancing adsorption [121]. When applying sludge-based adsorbents for wastewater treatment, solid waste generation could be minimized by applying the optimum dose of sludge without using excess quantities.

5. Regeneration of Sludge-Based Adsorbents

The discharge of spent adsorbents without proper treatment causes environmental pollution by both the adsorbed pollutant and the adsorbent [133,134]. Regeneration of adsorbents, which is an essential but less attention-paid aspect of adsorption studies, that can reduce the harmful environmental and health impacts caused by the toxic effects of heavy metals in exhausted adsorbents [135,136]. The recycling of adsorbents and recovery of the heavy metals also provide economic benefits by extracting valuable metals from sludge and increase the reusability of sludge-based adsorbents, enhancing their applicability in industrial scale [75,76,136,137].

Regeneration studies have shown the potential of different desorption methods to remove heavy metals from spent sludge-based adsorbents [69,107,119,138]. Table 3 shows regeneration methods used for sludge-based adsorbents along with the number of regeneration cycles tested with the efficiency of the method.

Table 3. Various methods used for the regeneration of sludge-based adsorbents.

Adsorbent	Heavy Metal/s	Regeneration Method	Number of Regeneration Cycles	Removal Efficiency	Reference
Diethylenetriaminepentaacetic acid–chitosan sewage sludge composite	Cu^{2+}	Stirring with HCl (0.1 M) for 24 h	6	56.79%	[120]
$ZnCl_2$-treated Sewage sludge-derived activated carbon	Cu^{2+}	Treating with HNO_3 (1.5 M)	1	98.9%	[112]
Sulfur-impregnated paper sludge	Ni^{2+}	Stirring with H_2SO_4 (0.05 M) /HCl (0.5 M) for 2 h	1	H_2SO_4—85.9% HCl—99.9%	[77]
TiO_2/ZnO electrospun nanofibers coated-sewage sludge carbon	Ni^{2+}, Cu^{2+}	Treating with HCl (0.1 M)	5	>90%	[110]
KOH-activated sewage sludge and sugarcane bagasse	Pb^{2+}	Treating with HCl (0.1 M) & NaOH (0.1 M)	5	70.82%	[128]
Urea and NaOH-treated carbonized sludge	Cr^{6+}	Impregnating in NaOH (2 M) for 12 h	5	>95%	[113]

Table 3. *Cont.*

Adsorbent	Heavy Metal/s	Regeneration Method	Number of Regeneration Cycles	Removal Efficiency	Reference
Amino-functionalized magnetic aerobic granular sludge biochar	Pb^{2+}	Stirring with Na_2EDTA (0.1 M) for 4 h	5	88.14%	[119]
Zero-valent iron-treated sewage sludge-derived biochar	Cr^{6+}, Pb^{2+}	Centrifugation and filtration	3	Cr^{6+}—78.1% Pb^{2+}—67.4%	[138]

5.1. Regeneration by Acid Treatment

Hydrochloric acid (HCl) has been used to regenerate sludge-based adsorbents. The desorption ratio, the ratio between the amount of metal ions desorbed and the amount of metal ions adsorbed, can be used to determine the desorption efficiency. A study by Lee et al. [69] regenerated Cd^{2+} adsorbed palm oil sludge by HCl with a concentration ranging from 0.05 to 1 mol L^{-1}, where the desorption ratio increased from 0.85 to 0.95. Nevertheless, above 0.4 mol L^{-1}, the increase in desorption ratio was insignificant due to irreversible damage of binding sites inducing incomplete removal of Cd^{2+}. Ni^{2+} has been desorbed by HCl from sulfur-impregnated paper sludge with 99.9% removal efficiency after the first regeneration cycle. The release of sulfide groups from sludge has caused a drastic decrease in sulfur/carbon ratio of the sludge surface from 0.03 to 0.002, reducing both the adsorption and desorption efficiency of sludge after the first regeneration cycle [77]. Over 90% of removal efficiency for both Ni^{2+} and Cu^{2+} with an initial metal concentration of 100 mg L^{-1} was obtained after five regeneration cycles in TiO_2/ZnO electrospun nanofibers coated-sewage sludge carbon regenerated by 0.1 M HCl indicating the high stability of the adsorbent for removal of Cu^{2+} [110].

H_2SO_4 can be used for the regeneration of sludge-based adsorbents. Ni^{2+} has been desorbed by 0.05 M H_2SO_4 from sulfur-impregnated paper sludge with 85.9% removal efficiency after 1st regeneration cycle that was reduced acutely after the second regeneration cycle due to damage of binding sites [77].

Sewage sludge-derived activated carbon has been successfully regenerated by 1.5 mol L^{-1} HNO_3. The removal efficiencies of Cu^{2+} in $ZnCl_2$-treated sewage sludge and KOH-treated sewage sludge were 98.9% and 95.5% after the first regeneration cycle [112]. The desorption efficiency of acid treatment (HCl, HNO_3, and H_3PO_4) in sulfur-impregnated paper sludge increases with the increase in acid concentration in the first regeneration cycle [77].

Nevertheless, a reduction in the removal rate of Cu^{2+} from 42.38% to 24.23% after six regeneration cycles has been observed in diethylenetriaminepentaacetic acid–chitosan sewage sludge composite regenerated by HCl due to a decrease in binding sites of the adsorbent [120]. It indicates the decline of the potential reusability of adsorbents regenerated by HCl treatment with the increase in several regeneration cycles. Therefore, it can be concluded that the acid treatment can effectively be used for the first regeneration cycle of sludge-based adsorbents and repeated afterward depending on the stability of the adsorbent [77,110].

5.2. Regeneration by Bases

Zhang et al. [113] regenerated Cr^{6+} adsorbed carbonized sludge modified by urea and NaOH by NaOH, H_2SO_4, and HNO_3. The results of the study revealed a higher removal rate of 95% by NaOH while the removal rates of acids (H_2SO_4 and HNO_3) were below 60% until five regeneration cycles. This indicates the high reusability of the adsorbent regenerated with NaOH. The adsorption capacity of Cr^{6+} into polyethyleneimine-modified activated sludge showed only a slight decrease from 86.90 mg g^{-1} to 81.78 mg g^{-1} after

the first regeneration with 0.01 mol L^{-1} NaOH at 25 °C, indicating the reusability of the adsorbent [107].

5.3. Other Regeneration Methods

In addition to regeneration by acids and bases, sludge-based adsorbents have been regenerated by centrifugation and filtration. Zero-valent iron-treated sewage sludge-derived biochar was regenerated by centrifugation and filtration, which obtained removal efficiencies of 98.4% and 78.1% for Cr^{6+}, and 91.8% and 67.4% for Pb^{2+} after first and third regeneration cycles [138]. The decrease in removal efficiency occurred due to the release of Fe from the adsorbent with regeneration.

The strength and type of method used to regenerate adsorbent depend on the adsorbate binding [133]. Therefore, it is important to select the appropriate regeneration method to obtain both environmental and economic benefits from water treatment by adsorption.

6. Way Forward

This review clearly shows that sludge-based adsorbents can potentially remove heavy metals from wastewater. Therefore, further developments of this technology for wastewater treatment in industrial scale are important. Both pros and cons can be seen in the application of sludge-based materials in wastewater treatment. Since sludge is a by-product of wastewater treatment that requires disposal in landfills, the provision of large quantities of sludge to prepare adsorbents could be easy [32,51–53]. Nevertheless, the studies on applying sludge-based materials for heavy metal removal have been limited to lab-scale studies [60–63]. Only a few studies have utilized sludge-based adsorbents to remove heavy metals from wastewater, while the majority have used aqueous solutions of heavy metals for adsorption studies [88,139]. Sludge also has the potential to be used in the same industry it was produced. For instance, Ghorpade and Ahammed [94] found that electroplating sludge could be utilized for the wastewater treatment of the same industry. Therefore, conducting future studies on heavy metal removal in multi-ion systems under actual environmental conditions is essential. The challenges in the real-world application of sludge-based materials for heavy metal removal could be minimized by various methods. Difficulties in utilizing, storing, transporting, and replacing spent adsorbents could be reduced by granulation, pelleting, and encapsulation of sludge-based materials [93,140]. Proper modification methods of adsorbents, such as chemical treatment, enhance the stability, reusability, and adsorption efficiency of the sludge-based adsorbents [110,113]. Both chemical modification of sludge and granulation techniques can be used to reduce the possible leaching of heavy metals from sludge-based adsorbents [113,140]. Studies on the regeneration of sludge-based materials also require further attention in improving metal recovery from spent adsorbents and recycling of sludge to enhance their applicability in wastewater treatment facilities.

7. Conclusions

This review shows the importance of using sludge-based materials as a promising and sustainable adsorbent for the removal of heavy metals from wastewater. The main types of sludge used are industrial sludge, drinking water treatment plant sludge, agricultural sludge, and sewage sludge. Physical and chemical activation methods have been utilized for the preparation of adsorbents. pH, temperature, initial heavy metal concentration, contact time, and adsorbent dose have been found as main factors that influence the adsorption of heavy metals into sludge. Functional groups including hydroxyl groups and carboxyl groups in sludge, play an important role by enhancing the adsorption of heavy metals into sludge. Most adsorption isotherm studies of heavy metal removal from wastewater by sludge are monolayer adsorption and multilayer adsorption. Most of the kinetic studies on sludge-based adsorbents suggested the contribution of physisorption and chemisorption in the heavy metal adsorption process. Thermodynamic studies have shown that most heavy metal removal processes by sludge-based adsorbents were thermodynamically feasible.

Some studies have been conducted on the regeneration of sludge-based adsorbents, including treatment with acids, bases, and other regeneration methods, such as centrifugation followed by filtration. These are cost-effective and environmentally friendly solutions for solid waste generation by adsorption. Nevertheless, less attention has been paid to the metal recovery and recycling of sludge-based adsorbents, indicating the need for future studies to enhance sludge reusability in wastewater treatment. In addition, it is important to conduct more studies on eco-friendly and effective adsorbent preparation methods that are not harmful to the environment in the disposal of sludge-based adsorbents. According to this analysis, some studies found that heavy metals such as Pb^{2+}, Cu^{2+}, Cd^{2+}, Ni^{2+}, Cr^{6+}, Zn^{2+}, Co^{2+}, and Hg^{2+} could be best removed by sludge-based adsorbents from actual wastewater such as desalination concentrate, electroplating wastewater, and stormwater. Nevertheless, other studies have mainly focused on the lab-scale removal of heavy metals from aqueous solutions using different sludge-based adsorbents. Therefore, future research needs to be focused on the removal of heavy metals from actual wastewater at field scale.

Author Contributions: Conceptualization, M.B.B., B.G.N.S., M.M.M.N. and B.A.A.; data curation, R.M.A.S.D.R. and B.G.N.S.; funding acquisition, B.A.A.; formal analysis, B.G.N.S., R.M.A.S.D.R. and M.M.M.N.; investigation, B.G.N.S., R.M.A.S.D.R. and M.M.M.N.; methodology, M.B.B., B.G.N.S., R.M.A.S.D.R. and B.A.A.; project administration, B.A.A. and B.G.N.S.; resources, B.A.A., A.T. and B.G.N.S.; supervision, B.G.N.S. and B.A.A.; validation, M.B.B. and A.T.; visualization, M.B.B., A.T., B.G.N.S., M.M.M.N. and B.A.A., writing—original draft, R.M.A.S.D.R., B.G.N.S. and M.M.M.N.; writing—review and editing, M.B.B., B.A.A., B.G.N.S., R.M.A.S.D.R., M.M.M.N. and A.T. All authors have read and agreed to the published version of the manuscript.

Funding: The authors extend their appreciation to the Deputyship for Research and Innovation, Ministry of Education in Saudi Arabia for funding this research through project IFKSUOR3-141-3.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors extend their appreciation to the Deputyship for Research and Innovation, Ministry of Education in Saudi Arabia for funding this research (IFKSUOR3-141-3).

Conflicts of Interest: The authors declare no conflict of interest.

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