

Response surface optimization and modeling in heavy metal removal from wastewater—a critical review

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Abstract The existence of hazardous heavy metals in aquatic settings causes health risks to humans, prompting researchers to devise efective methods for removing these pollutants from drinking water and wastewater. To obtain optimum removal efficiencies and sorption capacities of the contaminants on the sorbent materials, it is normally necessary to optimize the purifcation technology to attain the optimum value of the independent process variables. This review discusses the most current advancements in using various adsorbents for heavy metal remediation, as well as the modeling and optimization of the adsorption process independent factors by response surface methodology. The remarkable efficiency of the response surface methodology for the extraction of the various heavy metal ions from aqueous systems by various types of adsorbents is confrmed in this critical review. For the frst time, this review also identifes several gaps in the optimization of adsorption process factors that need to be addressed. The comprehensive analysis and conclusions in this

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review should also be useful to industry players, engineers, environmentalists, scientists, and other motivated researchers interested in the use of the various adsorbents and optimization methods or tools in environmental pollution cleanup.

Keywords Adsorption · Factors · Modeling · Optimization · Process · Variables

Introduction

Potable water is essential for all human beings around the world to meet their fundamental needs, including drinking and using it to cook. However, due to fast industrialization, globalization, and rising population, the plurality of freshwater bodies around the world are polluted with a range of heavy metals and other harmful substances (Mohd et al., [2021\)](#page-30-0). The globe has evolved enormously in recent years in all sectors, including humankind, society, science, and technology. As a result, various harmful contaminants, including toxic heavy metals ions, are frequently being found in aquatic settings (Anfar et al., [2020](#page-26-0)). The growing use of these heavy metals in scientifc, agricultural, residential, industrial, and clinical applications has increased their environmental transport into waterways. The severe toxicity and negative impacts of heavy metals on human health have prompted awareness about their removal from various water sources (Abedpour et al., [2020\)](#page-26-1).

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Several treatment procedures have been explored to eliminate heavy metals from synthetic and real wastewaters, including fltration, ion exchange, chemical precipitation, electrocoagulation, bioremediation, and adsorption (Mahmoud et al., [2021\)](#page-30-1). Except for the adsorption approach, all of these procedures and technologies have many drawbacks, including high costs, the creation of large volumes of waste, and the inability to meet the international community's standards (Egirani et al., 2021). The efficiency of the adsorption system depends on a number of independent factors, including solution pH, temperature, adsorbent type, dosage, and the presence of organic or inorganic components in the medium (Nazari et al., [2021\)](#page-30-2).

Over the years, one variable at a time is applied to track the infuence of the operating factors on the adsorption of heavy metals onto sorbent materials. This method necessitates a large number of experiments, and it also fails to highlight the interactive behavior of the process factors (Karmaker et al., [2021\)](#page-29-0). Furthermore, when dealing with a high number of variables, this technique is time consuming and costly (Cheng et al., [2021\)](#page-27-0). To overcome these limitations in terms of adsorption system scale-up, statistical analysis has various advantages, for instance, higher reliability and quicker than traditional methods, allowing us to better appreciate the interactions between the adsorbates and adsorbents and decreasing the total number of experiments required. Using the statistical design of experiments in the formulation of adsorption processes could lead to improved cleanup efficiency, lower process variability, and lower overall expenses in wastewater treatment (Çiçek et al., [2012\)](#page-27-1).

The response surface methodology (RSM) is one of the applications of design of experiments (DOE), which is a set of mathematical and statistical tools for designing, enhancing, and optimizing processes. It may be applied in assessing the relative importance of several infuencing variables (Aslani et al., [2018\)](#page-26-2). This method can also be used to analyze studies in which one or more independent factors are infuenced by a wide range of factors, with the goal of optimizing the responses. To attain the highest adsorption capacity and removal efficiency, the process factors must be optimized. Aside from lowering the number of adsorption tests, one of the benefts of this strategy is the ability to provide a mathematical correlation between the independent and dependent factors (Sagharloo et al., [2021](#page-31-0)). In addition, this procedure gives users the opportunity to study the efect of independent variables in relation to the numerical variables (responses), and it allows them to collect vast volumes of data from a small number of tests (Jamileh et al., [2020](#page-29-1)).

Considering the past 10 years, this review has provided a critical evaluation of the modeling and optimization of independent and dependent parameters infuencing heavy metals removal using central composite design (CCD), Box–Behnken design (BBD), factorial experimental design (FED), and artifcial neural network (ANN).

Independent affecting parameters

During the sorption process, many independent factors affect the removal efficiencies and sorption capacities of the prospective adsorbents or biosorbents. Previous studies have affirmed that the physicochemical characteristics of the solutions, such as contact/shaking time, pH, adsorbent dose, particle size, initial metal ion concentration, agitation/shaking speed, temperature, and interfering ions, have a significant impact on the efficiency of any adsorbent. The adsorbent ability, selectivity, adsorption rate, and the number of heavy metals to be removed are all afected by these process variables. To study the interaction between these independent factors/parameters, a signifcant number of adsorption studies have been compiled.

Contact/shaking time

Heavy metal removal is infuenced by the contact/shaking time between the adsorbing material and the aqueous solution. The longer the contact period, the more likely equilibrium has been achieved, and hence adsorption has achieved its optimum (Azadegan et al., [2019\)](#page-27-2). The adsorption process will not be completed successfully if not enough contact time is allowed (Salah, [2015](#page-31-1)). According to studies, the degree of contaminants removal from wastewater utilizing biomaterials is greater at the start of the remediation phase. This is due to the fact that as the interaction process progresses, the reaction sites become saturated, resulting in a reduced contaminant uptake rate (Rahdar et al., [2019\)](#page-31-2). Adsorbent dosage, contaminant concentration, temperature, and adsorbent surface groups all infuence the optimum contact time.

Agitation/shaking speed

The adsorption process is also affected by agitation/ shaking speed, and heavy metals reduction can be found to be best at a specifc speed that must be determined by testing. The adsorption process can be sped up or slowed down by agitation. A faster agitation speed, on the other hand, does not always imply a faster rate of adsorption (Kaakani, [2012](#page-29-2)). However, according to Dhoble et al. (2018) (2018) , the removal efficacy increases with increased agitation speed, which could be attributed to more collisions within the adsorbent particles, allowing more active sites on the adsorbent surface to be accessible for the number of metal ions.

pH of the solution

Adsorption operations are infuenced by the pH of the aqueous phase because H+ and OH− ions are released into the solution. A higher pH emits more H^+ ions, which may interact with the adsorbent or adsorbate, slowing down the adsorption rate. An alkaline pH solution, on the other hand, produces OH–, which might react with the adsorbent or adsorbate. The detoxifcation of heavy metal ions from the aquatic media is mostly influenced by pH. It affects the surface charge of the adsorbent, ionization degree, and the adsorbate speciation in general (Ittrat et al., [2014](#page-29-3)). As a result, the pH of an aqueous solution being evaluated for its sorption ability has an indisputable efect on the metal ions uptake since, within a particular pH range, the majority of adsorption processes increase with an increase in pH until at a point where an increment leads to a reduction in the adsorption rate. The functional groups on the surface of the adsorbents and the adsorbate aqueous solutions can be linked to the pH dependency on heavy metals uptake (Kumar et al., [2016](#page-29-4)).

The infuence of pH on the elimination of toxic heavy metals in the aqueous systems is critical and the pH efect occurs when heavy metals combine with protons to create hydrogen heavy metals at low pH. Furthermore, at high pH, there is an oversupply of hydroxide ions, which prevent heavy metal ions from difusing (Bayuo et al., $2019a$, [b\)](#page-27-5). As a result of the pH of the aqueous solution controlling the adsorbent's surface charge, it shows greater dependence on the decontamination of heavy metals from wastewater.

Adsorbent particle size

The adsorbent particle size is one of the critical characteristics that have a signifcant impact on the adsorbent's adsorptive capacity. The adsorbent removal efficiency and adsorption capacity of the adsorbent vary with particle size; the adsorption capability rate reduces as particle size increases, while it enhances as particle size lowers. The surface area of the adsorbent increases as its size decreases, and a larger surface area indicates a larger active site for adsorption (Memon et al., [2021\)](#page-30-3). At various sizes, the impact of the particle size on the adsorption should be assessed since this type of information aids in the design of a full-scale adsorption system for commercial use.

Apart from adsorption at the adsorbent surface, intraparticle difusion from the surface of the pores of the adsorbent is a possibility. As a result, bigger particle sizes are more resistant to mass transfer (Ahmaruzzaman, [2011\)](#page-26-3). Due to several inhibitory variables including resistance to mass transfer actions, contact time, and blockage of the difusional route, the interior adsorbent particle surface area may not be fully exploited, resulting in a lower adsorption capacity. Regardless, the adsorption efficiency is mostly determined by the availability of the surface area for the adsorbate interactions (Emenike et al., [2016\)](#page-28-1).

Adsorbent dose

The adsorbent weight determines the adsorbent's ability for a particular adsorbate concentration. As more active adsorption sites are provided by increasing the dosage of the adsorbent, more contaminants are removed from the aqueous phases. This could be due to enhanced surface and pore volume accessibility at higher dosages, as well as a larger surface area (Pyrzynska, [2019](#page-31-3)). Increasing the surface area of the adsorbent is one technique to reduce the amount of adsorbent to be used in removing the adsorbate. This can be accomplished by utilizing adsorbents with very small dimensions. As larger surface areas are more effective at adsorption, the same adsorbent may need fewer dosages once processed than its bigger counterpart (Baby et al., [2018](#page-27-6)). However, with the unsaturation of the available sorption sites generated by large adsorbent dosages and the adsorbate interactions, the adsorption density usually dropped (Mbugua et al., [2014](#page-30-4)).

Heavy metal ion concentration

The initial concentration of heavy metal ions can change the removal efficiency and adsorption capacity due to a range of factors such as the availability of specifc surface functional groups and the ability of these surface functional groups to entangle heavy metal ions from aquatic environments. The solution of the initial metal ion concentration can act as an infuential driving force in overcoming the resistivity of mass transfer between the liquid and solid phases (Jjagwe et al., [2021](#page-29-5)). By increasing the initial metal ion concentration in the aqueous environment, the adsorbent capacity is quickly depleted. This is because the number of accessible adsorption sites for a given dose of the adsorbent is restricted, and at high concentrations, they become saturated (Zhao et al., [2020\)](#page-33-0). The better the adsorption rate, the lower the metal ion concentration, as there is less adsorbate for the adsorbent to extract (Afroze & Sen, [2018](#page-26-4)). Some adsorbents, on the other hand, behave exceptionally well at high initial adsorbate concentrations and so have a higher adsorption capacity. Due to their adsorption capacity, these adsorbents are advantageous (Kaakani, [2012](#page-29-2)).

More so, the initial concentration is one of the ways through which heavy metal ion mobility to the adsorbent's surface might be aided (Sahmoune et al., [2011](#page-31-4); Taha et al., [2011](#page-32-0)). To examine the impact of baseline concentration on metals adsorption utilizing agricultural adsorbents, a large number of experiments have been undertaken and reported. The interaction between optimum adsorption capacity and baseline concentration has been established as a general trend. With rising initial concentration, the trends suggested increasing adsorption ability (Essa, [2012](#page-28-2); Kumar et al., [2012](#page-30-5)).

Temperature

Adsorption is temperature dependent, and it can alter the adsorption system making it endothermic or exothermic in nature (Iftekhar et al., [2018\)](#page-29-6). A large number of biosorption studies on the infuence of temperature on heavy metals removal have been conducted. Not only does temperature afect the solubility of metals, but it also afects the rate of difusion (Mohubedu et al., [2019](#page-30-6)). As a result of the diverse functional groups on the surfaces of agricultural adsorbents, temperature is recognized as an essential factor in heavy metals adsorption (Emenike et al., [2016\)](#page-28-1).

Nonetheless, according to Park et al. [\(2014](#page-31-5)), the increase in temperature can cause physical deformation of the adsorbent. As a result, in many adsorption experiments, ambient temperature is commonly used.

Interference

Anions such as carbonates, chlorides, nitrates, phosphates, and bicarbonates, as well as heavy metals, may be present in aqueous systems. The concentration of these ions varies by geographical region and can infuence contaminant sorption on the adsorbent (Foroutan et al., [2019](#page-28-3)). The order of heavy metals removal interference is given as $HCO_3^- > SO_4^{2-} > Cl^-$ for the various biosorbents. In most studies, heavy metals uptake was not reduced by chloride, sulfate, or nitrate ions, but bicarbonate signifcantly reduced heavy metal take-up. This is attributed to the ability of the bicarbonate to compete with heavy metal ions for active sites on the adsorbents (Kanaujia et al., [2015\)](#page-29-7). In general, the adsorption rate is inversely related to the aqueous system ionic potential. As the ionic strength increased, the adsorbents' ability to absorb metal ions declined. This could be due to an upsurge in the concentration of the competitive cations in the aqueous solution, which affects metal ion activity (He et al., [2018\)](#page-28-4). Real wastewater comprises diverse metal ions, not just only heavy metals. For instance, the common metal ions found in real wastewater include Na^+ , K^+ , Mg^{2+} , and Ca^{2+} but chemical compounds such as KCl, CaCl₂ $MgCl₂$, and NaNO₃ are usually introduced into synthetic wastewater to investigate the effect of ionic potential in competitive heavy metals sequestration.

Dependent affecting parameters

Generally, the adsorption process independent variables are normally studied as a function of dependent variables such as removal efficiency $(\%)$ and adsorption capacity (mg/g). The quantity of heavy metal ions adsorbed or removed from the adsorption system is usually calculated by the simple concentration diference method. Specifically, the removal efficiency and adsorption capacities are estimated using Eqs. [1–](#page-4-0)[3,](#page-4-1) respectively (Afolabi et al., [2021;](#page-26-5) Bayuo, [2021](#page-27-7)).

Removal efficiency (
$$
\% = \frac{(C_0 - C_e)}{C_0} \times 100
$$
 (1)

The adsorption capacity at a particular time (*t*) is given by

$$
q_t = \frac{(C_0 - C_t) \times V}{m} \tag{2}
$$

The adsorption capacity at equilibrium (q_e) is also given by

$$
q_{\rm e} = \frac{(C_0 - C_{\rm e}) \times V}{m} \tag{3}
$$

where C_e is the concentration of metal ion at equilibrium (mg/L), C_0 is the initial concentration of the metal ion (mg/L), C_t is the concentration of metal ion at time $t \text{ (mg/L)}, q_t$ is the amount of metal ion adsorbed at time t (mg/g), *V* is the volume of the aqueous solution (mL), and *m* is the mass (g) of the adsorbent.

Heavy metals removal and optimization methods

Several studies have been reviewed on the removal of heavy metals from wastewater and the applicability of the RSM in optimizing the independent factors afecting the adsorption process of these heavy metals. In this review, the authors provided their comprehensive analysis and insight on the current trends, developments, future directions, and signifcance of the following studies for worldwide scientifc applications.

Modeling and optimization of heavy metals removal using central composite design (CCD)

An RSM based on the central composite design (CCD) was applied in the evaluation of the effects of diverse factors and their interactive characteristics on the effectiveness of $Sr(II)$ decontamination by nanoscale zero-valent iron-zeolite (nZVI-Z) and nano-Fe/Cu zeolite (nFe/Cu-Z) as adsorbents (Karmaker et al., [2021\)](#page-29-0). The study shows that the three variables tested, pH of the aqueous solution, contact time, and initial Sr(II) concentration, are all positively associated with Sr(II) removal. The highest Sr(II) removal happened at pH 12.00, initial Sr(II) concentration of 200.00 mg/L, and a contact time of 30.00 min with a maximum uptake capacity of 32.50 mg/g and 34.00 mg/g for nZVI-Z and nFe/Cu-Z adsorbents, respectively. In comparison to the other isotherm models, the Toth model matched well with the experimental Sr(II) uptake data. In this study, aqueous solutions of Sr(II) were used, and no desorption and regeneration tests were conducted as well as no adsorption kinetics and thermodynamics were reported, which could have further explained Sr(II) adsorption mechanism.

The infuence of the experimental variables on the competitive sorption of $Ni(II)$ and $Cu(II)$ ions by activated carbon derived from sewage sludge was evalu-ated using CCD of the RSM (Khelifi et al., [2021](#page-29-8)). The optimum Ni(II) concentration, Cu(II) concentration, adsorbent dose, contact time, and temperature were found to be 40.00 mg/L, 40.00 mg/L, 4.00 g/L, 100.00 min, and 30.00 °C. The maximal adsorption rate of 7.48 and 4.04 mg/g for Ni(II) and Cu(II) ions, respectively, was achieved under this optimized condition. In this study, aqueous solutions of Ni(II) and Cu(II) were used, and no kinetics and thermodynamics adsorption were studied, which could have provided more insight into the adsorption behavior of these metal ions. Although desorption and regeneration studies were not conducted, it was concluded that the prepared activated carbon has the potential for $Ni(II)$ and $Cu(II)$ ions removal from aqueous solutions for environmental cleaning purposes.

The infuence of the initial Pb(II) concentration, solution pH, and temperature on the fraction of Pb(II) adsorbed by phosphogypsum (PG) was studied (Lamzougui et al., [2021\)](#page-30-7). The experimental design approach was used to model the adsorption tests, and the CCD of the RSM is used to optimize the parameters. The fndings reveal that the amount of Pb(II) removed upsurges with the initial Pb(II) concentration and reduces as the pH and temperature of the solution rise. At Pb(II) initial concentration of 109.64 mg/L, 5.25 pH, and 70.00 \degree C temperature, the optimal adsorption capacity of Pb(II) on the PG was attained. The kinetic data shows good representation with the pseudo-second-order model and it was discovered from the thermodynamic studies that the adsorption process was exothermic and spontaneous. In this study, the equilibrium isotherms were not investigated and so how well the adsorbate interacted with the adsorbent in the aqueous systems is not known. In addition, there is no information on the desorption, regeneration, and reusability of the used adsorbent. Yet, it was concluded that PG will be an efective adsorbent for Pb(II) sequestration from aqueous solutions since it is of low cost.

For the extraction and purifcation of As(III) from wastewater, activated red mud–doped calcium-alginate beads were utilized (Naga et al., [2021\)](#page-30-8). The CCD of the RSM-based statistical modeling was applied in the optimization of the operational parameters (pH, sorbent dosage, contact time, and baseline concentration) for maximized As(III) removal. The highest adsorption of 92.00% of As(III) removal was found under the ideal operating parameters of pH 7.00–8.00, initial concentration of 10.00 mg/L, 0.80 g/L dosage, and 120 min time of contact at 303.00 K. The thermodynamics and isothermal studies confrm the sorption process occurs non-spontaneously and endothermically with the Langmuir isotherm being the best ft to the As(III) equilibrium data. Furthermore, the kinetic analysis demonstrates that the experimental data best fts the pseudo-second-order model. In comparison to other adsorbents reported in several studies, the prepared adsorbent is efficient in sequestering $As(III)$ up to an optimum capacity of 1.81 mg/g, and it could be regenerated up to five cycles with 74.00% As(III) retrieval.

An immobilized $ZnO/TiO₂$ activated carbon was developed for As(III) subtraction from wastewater and was modeled by CCD of the RSM to fnd optimized conditions (Sagharloo et al., [2021\)](#page-31-0). According to optimized results, the best dosage, pH, contact time, and concentration to meet environmental regulations are 5.19 g/L, 6.76, 287.57 min, and 9.77 mg/L, respectively. The Langmuir isotherm and pseudo-secondorder models show better agreement with the obtained data. The adsorption thermodynamics, retrieval, and reusability of the used adsorbent were not tested, but the study infers that the new immobilized $ZnO/TiO₂$ activated carbon has a very superior efficiency in As(III) removal while being cost-efective, making it extremely useful for practical applications.

Lead[Pb(II)] was removed by two low-cost industrial products of ground granulated blast-furnace slag (GGBFS) and phosphorus slag (PS) (Shafaghat & Ghaemi, 2021). To compare the efficacy of both adsorbents in removing Pb(II) from an aqueous phase, the RSM based on the CCD approach was used. The initial Pb(II) concentration, rotation rate, and adsorbent weight were all taken into account in the experimental design. The optimum adsorption capacity (6.41 mg/g) was determined using RSM models at the adsorbent weight (0.10 g/L) , initial Pb (II) concentration (100.00 mg/L) , and rotation rate (195.00 rpm) . In this study, the adsorption equilibrium isotherms, kinetics, and thermodynamics were not conducted; hence, the mechanism of Pb(II) decontamination by the adsorbents is not discussed. More so, there is no information on the desorption of the adsorbed ions and regeneration of the depleted adsorbents for reuse, but the study indicates that GGBFS was more efficacious than PS adsorbent in removing Pb(II) under the same operating conditions.

Pb(II) and Cd(II) were removed from aqueous systems using an alumina modifed onion skin (AMOS) compos-ite (Yusuff et al., [2021\)](#page-33-1). The CCD-RSM was utilized to optimize Pb(II) and Cd(II) removal onto AMOS, and the obtained best condition was Pb(II) and Cd(II) initial concentration of 200.00 mg/L, adsorbent dose of 1.20 g/L, and 75.59 min time of contact, with 92.05% and 94.89% removal efficiencies for $Pb(II)$ and $Cd(II)$, respectively. The Langmuir model gave the best ft to the equilibrium data with monolayer adsorption capacities of 9.74 mg/g and 14.17 mg/g for $Pb(II)$ and $Cd(II)$, respectively. The pseudo-second-order model explains the adsorption data better. The desorption and regeneration of the exhausted adsorbent were studied and the reusability results suggest that AMOS is profcient and could be used and recycled in the wastewater treatment system. However, the thermodynamics parameters (Gibbs free energy, enthalpy, and entropy changes), which could have described the sorption characteristics of these heavy metals were not reported.

A study was carried out to fnd out if utilizing activated carbon prepared from Teff husk may improve Cr(VI) removal from aqueous solutions (Adane et al., [2020](#page-26-6)). To determine the efect of the interactions of the process factors and optimize the process, a CCD-RSM approach was adopted. The optimum removal efficiency (95.60%) of the adsorbent was reached at pH of 1.92, initial Cr(VI) concentration of 87.83 mg/L, adsorbent dose of 20.22 g/L, and 2.07 h time of contact. The best match models for the Cr(VI) adsorption data were the Langmuir and pseudo-second-order. The thermodynamics of Cr(VI) detoxifcation was not investigated and so how well the adsorption system proceeded is not known. In addition, there is no information on the desorption, regeneration, and reusability of the used adsorbent. Besides, the study recommends the Tef husk as being efective and an economical adsorbent for Cr(VI) removal from contaminated water.

In batch mode, CCD-RSM was used to remove Cr(VI) from the aqueous phase using an untreated biosorbent derived from *Arachis hypogea* husk (AHH) (Bayuo et al., [2020\)](#page-27-8). The independent factors afecting the biosorption system were optimized, and the optimal condition was attained as a contact time of 120.00 min, solution pH of 8.00, and an initial Cr(VI) concentration of 50.00 mg/L was achieved with a maximum uptake capacity of 2.36 mg/g. The equilibrium and kinetic data show good ftness to the Redlich–Peterson, and pseudo-second-order models. The thermodynamics parameters, which could have explained the sorption characteristics of Cr(VI) was not reported as well as no desorption and reusability of the adsorbent were studied. Besides, the AHH biosorbent was capable of detoxifcation Cr(VI) from the aqueous systems.

A zeolitic imidazolate framework (ZIF)-8 was prepared and treated with dimethylethylenediamine (ZIF-8-mmen) for Cd(II) removal (Binaeian et al., [2020](#page-27-9)). The CCD-RSM was used to optimize the operations, which included three operational factors: pH of the solution, adsorbent dose, and contact time. With a maximum extraction efficiency of 85.38% , the best setting was reached at solution pH (2.00), dose (0.10 g/L), and time of contact (89.00 min). The equilibrium data best suited the Langmuir isotherm, indicating monolayer adsorption, whereas kinetic analyses of Cd(II) show a pseudo-frst-order model. The Cd(II) sorption process was characterized by spontaneous, endothermic, and physisorption according to thermodynamic studies. In the study, there is no information on the retrieval, and reusability of the used adsorbent; so, the economic potential of the adsorbent is not known.

A CCD of the RSM was applied in modeling batch experiments and optimized the impact of solution pH, contact time, adsorbent dosage, and initial concentration on Pb(II) decontamination by peanut hull-g-methyl methacrylate biopolymer (PH-g-MMA) (Chaduka et al., [2020\)](#page-27-10). The best condition for Pb(II) removal was achieved at 5.70 pH, 63.75 min time of contact, adsorbent dose of 4.50 g/L, and 76.25 mg/L Pb(II) initial concentration. Under this ideal condition, 99.30% of the Pb(II) in the aqueous media was extracted. The isothermal and kinetics analysis show that the experimental results matched well with the Langmuir and pseudo-second-order models, which describe the process as being chemisorptive. The thermodynamics parameters (Gibbs free energy, enthalpy, and entropy changes), suggest that the process happens spontaneously, endothermically, and upsurges the randomness on the adsorbent surface. The desorption and reusability tests reveal that PH-g-MMA could be regenerated up to 9 cycles and was capable of extracting over 75.00% of the Pb(II) from aqueous solutions.

Ecer et al. [\(2020\)](#page-28-5) investigated the infuence of initial Hg(II) and As(V) concentrations, solution pH, adsorbent dose, and contact time in the sequestration of Hg(II) and As(V) by the CCD of the RSM onto sulfur functionalized pumice. By applying numerical optimization, the optimal operating condition for Hg(II) removal was solution pH (6.33), initial metal concentration (36.94 mg/L), adsorbent dose (0.15 g/L), and time of contact (120.00 min), and the maximized condition for As(V) removal was solution pH (3.94), initial As(V) concentration (7.17 mg/L), adsorbent dose (0.15 g/L), and contact time (155.40 min). The adsorption yield of $Hg(II)$ and $As(V)$ in the attained optimized conditions were found as 92.14% and 88.02%, respectively. The equilibrium data for both metals were coherent with the Freundlich and Langmuir models, and the kinetic data of both metal ions were compatible with the pseudo-second-order model. In the study, the thermodynamics, retrieval, and reusability of the used adsorbent were not tested, but from the fndings, the adsorbent could be prudent for extracting various heavy metals from polluted aquatic systems.

Nanodiopside was employed as a unique, green, and proficient adsorbent for removing Cd(II) ions from aqueous environments (Ghanavati et al., [2020](#page-28-6)). The infuence of sorption variables on the removal efficiency of the Cd(II) was examined by CCD-RSM and the best condition of the process variables for optimum Cd(II) decontamination was found to be a pH of 5.60, adsorbent dose of 0.13 mg, Cd(II) initial concentration of 23.16 mg/L, 43.83 min time of contact time, and temperature of 34.75 °C. The adsorption data show a good correlation with the Freundlich and pseudo-second-order models. The thermodynamics parameters (Gibbs free energy, enthalpy, and entropy changes) that could have provided more insight into the sorption characteristics of Cd(II) were not reported. However, the desorption and regeneration tests were carried out, which show that the adsorbent was still capable of being recycled and recovered 95.95% of Cd(II) in the sixth cycle.

Javid et al. (2020) (2020) carried out a study to examine Cr(VI) removal ability with green-graphene nanosheets (GGN) synthesized using rice straw by the CCD-RSM approach. The interactive infuence of two independent parameters such as KOH-to-raw rice ash ratio and temperature on the GGN surface area development was studied. The study indicates that the optimal condition was obtained at a KOH-to-raw rice ash ratio of 10.85 and a temperature of 749.61 °C for the GGN preparation. The specifc surface area achieved at the optimized operating condition for GGN was $551.14 \text{ m}^2/\text{g}$ at Cr(VI) concentration (48.35 mg/L), adsorbent dose (1.46 g/L), contact time (44.30 min), and solution pH of 6.87. The Langmuir and pseudo-second-order models were the best ft for the obtained data. The adsorption thermodynamics, recovery, and reusability of the used adsorbent were not explored. However, the study suggests that the adsorbent is favorable and could be utilized in Cr(VI) reduction from wastewater.

The optimization of Cu(II) elimination from wastewaters by fy ash utilizing CDD-RSM was studied by Maiti et al. ([2020](#page-30-9)). The interactive effects of the following process factors, namely initial Cu(II) concentration, solution pH, and fy ash dose were determined and optimized. With an optimized initial Cu(II) concentration of 43.00 mg/L, pH 6.00, and a fy ash dosage of 63.00 g/L, the maximum $Cu(II)$ removal efficiency (93.80%) was reached. In this study, the equilibrium isotherms, kinetics, and thermodynamics as well as the regeneration of exhausted adsorbents were not discussed, and this could have ofered more insight into the sorption behavior of Cr(VI). Besides, according to the results, fy ash can be utilized to remediate acidic wastewater from a variety of sectors, including copper smelting, electroplating, and fertilizer manufacturing industries.

Batch experiments were investigated to assess the ability to utilize activated carbon from the *Manilkara zapota* tree in Pb(II) decontamination from aqueous systems by CCD of the RSM (Sujatha et al., [2020](#page-32-2)). The study examined the impact of individual and combination process variables, such as Pb(II) initial concentration, solution pH, and adsorbent dose, on Pb(II) depollution. At 0.837 desirability, the optimal uptake capacity (22.06 mg/g) of the adsorbent was obtained at the optimal condition of Pb(II) initial concentration (60.00 mg/L), pH (4.00), and adsorbent dose (0.2 g/L) . The obtained data was completely fitted by the Langmuir and the pseudo-second-order models. The physisorption mechanism occurs in the elimination of Pb(II), according to the mean adsorption energy calculated with the use of the Dubinin–Radushkevich isotherm. In this study, the used-up carbon was regenerated through desorption processes and about 91.00% of the adsorbed metal ions were recovered from the used carbon. Hence, the used carbon is nonhazardous and can be used for the landfll, which is a safe disposal strategy. The thermodynamics parameters (Gibbs free energy, enthalpy, and entropy changes), which could also explain the adsorption process of Pb(II) was not discussed.

The CCD of the RSM was being applied to study the reduction of $Cd(II)$ and $Pb(II)$ ions from effluents onto cow bone composite (Abdulrahman et al., [2019\)](#page-26-7). The ideal condition was achieved at pH of 4.00, agitation speed of 50.00 rpm, 24.00 h time of shaking, the particle size of 1.00 mm, and adsorbent dose of 12.50 g/L. The experimental results suited well to the Langmuir and Freundlich models for Cd(II) and Pb(II), respectively. Desorption and regeneration experiments were performed to make the sorption process more costeffective. The eluting ability of $Cd(II)$ and $Pb(II)$ was realized as 88.00% and 84.00%, respectively. The thermodynamics parameters (Gibbs free energy, enthalpy, and entropy changes) were not reported and so how the adsorptive removal of Cd(II) and Pb(II) have proceeded is not known.

The CCD of the RSM was used to optimize the variables for removing Hg(II) from water with a novel nanostructured adsorbent (Azadegan et al., [2019\)](#page-27-2). Three process variables including contact time, pH of the solution, and sorbent dose were considered. The optimal condition was obtained as a pH of 4.50, 25.00 min of contact time, and a sorbent dose of 0.06 g/L. The Langmuir and Freundlich models best agreed with the equilibrium data while both the pseudo-frst and second-order models ftted well with the obtained data. The reusability and real wastewater samples tests were carried out and the results indicate high efficacy and promising capability of the adsorbent for actual environmental applications. However, the thermodynamics parameters such as Gibbs free energy, enthalpy, and entropy changes were not reported.

The biosorption of Pb(II) from aqueous media onto Tamarind fruit shell powder (*Tamarinus indica.* L.) was studied (Bangaraiah & Sarathbabu, [2019](#page-27-11)). The operating factors such as agitation time, biosorbent dose, initial Pb(II) concentration, and solution pH were optimized by the CCD-RSM approach. The maximal removal efficiency of $Pb(II)$ is 83.50% at optimum process condition of 33.11 min of agitation time, 0.99 g/L biosorbent dose, 26.44 mg/L initial ion concentration, and pH of 6.98. The experimental data are best suited to Freundlich and pseudo-second-order models. The thermodynamics, desorption, and recyclability of the used adsorbent were not explored; however, the study reveals the *Tamarindus indica.* L. was an efective biosorbent for Pb(II) remediation.

The infuence of three process factors on the decontamination of Pb(II) by groundnut shell was investigated using the CCD-RSM approach (Bayuo et al., [2019a,](#page-27-4) [b](#page-27-5)). Applying the CCD, the optimized contact time (90.00 min), solution pH (8.00) , and initial Pb(II) concentration (75.00 mg/L) gave a maximum uptake of 90.26% and adsorption capacity of 3.43 mg/g of Pb(II), respectively, with the desirability of 0.966. The experimental results were better explained by the Langmuir and pseudo-second-order models. The adsorption thermodynamics, desorption, and reusability of the impregnated adsorbent were not investigated. Besides, the study reveals that the groundnut shell was efficient in $Pb(II)$ removal from the aqueous phases.

The expulsion of Ni(II) and Cu(II) from wastewater was tested by biochar-biopolymeric hybrid adsorbents by employing the CCD-RSM approach (Biswas et al., [2019](#page-27-12)). At the optimized condition, Cu(II) maximal reduction capacity (47.05 mg/g) was obtained at a baseline concentration of 84.80 mg/L, 1.40 g/L adsorbent dose, and temperature of 308.90 K, whereas Ni(II) uptake capacity (28.06 mg/g) was found at an initial concentration of 84.80 mg/L, adsorbent dose of 1.48 g/L, and temperature of 313.00 K. The Langmuir and pseudo-second-order models well explain the equilibrium and kinetic data, respectively. The process occurred endothermically and spontaneously, by the thermodynamic analyses. The recovery and reusability of the used adsorbent were not examined although the study concludes that the adsorbent is very prudent and demonstrates an excellent Ni(II) and Cu(II) ions removal.

A chitosan/rice husk ash/nano-*γ* alumina adsorbent was produced by Fooladgar et al. ([2019\)](#page-28-7) and used in the detoxifcation of Pb(II) from simulated wastewater. The CCD-RSM approach was used employed to maximize operating factors such as solution pH, time of contact, initial Pb(II) concentration, and adsorbent dose, leading to 90.98% Pb(II) removal under the optimal condition of 5.00 pH, 105.00 min time of contact, 30.00 mg/L Pb(II) initial concentration, and adsorbent dosage of 0.01 g/L. The Langmuir model best represented the adsorption behavior of Pb(II), while the pseudo-second-order model best described the obtained experimental data. The thermodynamic

results show that the Pb(II) adsorption was exothermic and spontaneous. The reusability of the used adsorbent in six subsequent adsorption–desorption cycles was conducted, and the uptake capacity was maintained at over 70.00% after six cycles, indicating that the nano-adsorbent demonstrates a high-level removal of Pb(II).

Fly ash was modifed chemically and used in removing Cr(VI) from aqueous environments (Jahangiri et al., [2019](#page-29-10)). To develop models for response prediction and optimize Cr(VI) process parameters, a CCD-RSM was used and a 3.53 g/L adsorbent dose, 35.40 mg/L Cr(VI) initial concentration, 69.32 min time of contact, and pH of 2.77 were found as the optimal parameters values. The Freundlich and pseudo-second-order models best described the data obtained. According to the fndings of the thermodynamic investigation, the sorption system was spontaneous, exothermic, and chemisorptive. The desorption and regeneration of the adsorbent were not conducted yet the study recommends fy ash as a great promising biosorbent for reducing Cr(VI) from aqueous solutions.

The CCD of the RSM has been used to examine the extraction of Cr(VI) from aquatic systems by Amberlite XAD7 resin-loaded titanium dioxide (Ti-XAD7) (Sharif et al., [2019](#page-32-3)). An initial Cr(VI) concentration of 2.75 mg/L, 51.53 min time of contact, 8.70 pH, and Ti-XAD7 dose of 5.05 g/L were found to be the best operating conditions. The Langmuir and Sips models describe the experimental data well. The kinetic studies show that the Elovich kinetic model adequately explains the Cr(VI) adsorption characteristics. The adsorption thermodynamics, recovery, and reusability of the impregnated adsorbent were not investigated. In contrast to untreated XAD7, the modifed XAD7 had a better Cr(VI) removal efectiveness, approximately 98.00% removal.

A study was conducted to explore Cr(VI) removal using both chitosan modifed with polyhexamethylene biguanide (Ch-PHMB NPs) and magnetic chitosan (M-Ch) from aqueous solutions by applying the CCD of the RSM (Aslani et al., [2018\)](#page-26-2). The impacts of four independent operating factors including solution pH, adsorbent dose, time of contact, and initial Cr(VI) concentration were optimized in Cr(VI) elimination. In contrast, all the four factors investigated were found to have significant influences on $Cr(VI)$ removal by Ch-PHMB NPs. However, for Cr(VI) removal by M-Ch NPs, only the interaction between solution pH and adsorbent dose indicates a signifcant efect. The Temkin and Freundlich models indicate a good representation of the obtained data for M-Ch and Ch-PHMB NPs, respectively. Both absorbents followed pseudo-second-order kinetics in extracting Cr(VI) from the aqueous systems. The thermodynamics investigations show Cr(VI) removal was nonspontaneous, exothermic, and decreased in randomness on the solid–liquid interface. The desorption and reusability of these adsorbents were not carried out. However, Ch-PHMB NPs adsorbent was found more efficient at removing Cr(VI), about 70.00% from the aqueous solutions than M-Ch adsorbent.

The infuence of independent process variables namely the initial Pb(II) concentration, adsorbent dose, and contact time on Pb(II) decontamination by nickel ferritereduced graphene oxide nanocomposite was studied by CCD of the RSM (Lingamdinne et al., [2018\)](#page-30-10). The removal of Pb(II) was observed to vary from 77.93 to 99.9%, which was infuenced by the process variables. According to the numerical optimization, the best parameters for achieving Pb(II) removal with 0.953 desirability were 18.38 mg/L Pb(II) initial concentration, an adsorbent dose of 0.55 g/L, and an 83.00 min contact period. The equilibrium data matches the Langmuir among other models analyzed but the kinetics, thermodynamics, recovery, and regeneration of the adsorbent were not reported in this study.

The batch adsorption technique was applied in removing Cd(II) from synthetic solutions using eggshell powder (Sabah et al., [2018\)](#page-31-6). A CCD based on the RSM was utilized in the process optimization, and the best condition was obtained at 44.00 °C temperature, 2.98 g/L adsorbent dose, 36.74 mg/L Cd(II) initial concentration, and solution pH of 7.00. The cleanup yield of Cd(II) was 98.76% under this condition. The Freundlich model shows a good correlation with the isothermal data. The desorption results reveal 45.60% of Cd(II) could be recovered, and the reusability of the adsorbent was not tested to determine its economic prospects. Also, the adsorption kinetics and thermodynamics of the batch system were not performed in this study.

The elimination of $Hg(II)$ from aqueous systems by 3-mercaptopropyl trimethoxysilane-modifed bentonite (B-SH) was conducted, and the process was optimized (Şahan et al., [2018\)](#page-31-7). The CCD-RSM fndings reveal that the best condition was attained at a solution pH of 6.17, 36.95 mg/L Hg(II) initial concentration, a temperature of 37.28 \degree C, and an adsorbent dose of 0.19 g/L. The maximum uptake capacity and percentage of removal were 19.30 mg/g and 99.23%, respectively, under the optimum condition established through the optimization method. The adsorption data ft the Langmuir model well than the other adsorption models. The adsorption process was discovered to be spontaneous, practical, and endothermic by thermodynamics investigations. The desorption and regeneration of the used adsorbent were not reported in this study. Besides, the study shows that the B-SH has a high Hg(II) adsorptive removal capacity and could also be used to remove other metal ions from aqueous environments.

The CCD of the RSM was applied to explore and maximize the mono-component removal of $Cu(II), Co(II),$ and Ni(II) onto trimellitated sugarcane bagasse adsorbent in a continuous fxed-bed column (Xavier et al., [2018](#page-32-4)). The Cu(II), Co(II), and Ni(II) ions had maximal uptake capacities of 1.06, 0.80, and 1.03 mmol/g, respectively, at the optimum operating conditions of initial metal ion concentration and spatial time. The Thomas and Bohart–Adams models were used to model the breakthrough curves, with the Bohart–Adams model predicting the experimental results more precisely. The enthalpy, Gibbs free energy, and entropy changes of adsorption onto the adsorbent indicate the process was endothermic, spontaneous, and improved with increasing disorderliness at the solid–liquid interface. The recovery and reuse of the used adsorbent were not discussed.

A chemical activation was employed to produce activated carbon from the seed shell of *Leucaena leucocephala* in extracting Cr(VI) from aqueous solutions through a batch system (Yusuff, [2018\)](#page-33-2). Applying the CCD-RSM, the variables afecting the system including initial adsorbate concentration, solution pH, adsorbent dose, and temperature, were optimized. The results show that the optimum adsorption rate was found as 95.62% at initial Cr(VI) concentration (71.49 mg/L), solution pH (4.22), adsorbent dose (0.57 g/L), and temperature (26.20 $^{\circ}$ C). The Freundlich and pseudo-second-order models ft the obtained data well. The adsorption thermodynamics, recyclability, and reuse of the adsorbent were not investigated. However, the Cr(VI) sequestration was proven to be effective using the activated carbon from the seed shell of *Leucaena leucocephala*.

Persian *Eucalyptus* leaves were examined as an adsorbent for removing As(III) and Hg(II) from aqueous solutions (Alimohammadi et al., [2017\)](#page-26-8). The uptake capacity and removal efficiency under various operating conditions (solution pH, time of contact, initial heavy metals concentration, and adsorbent dose) within the CCD of the RSM were studied for the optimization and modeling of the process. At the ideal condition of $pH=6.00$, contact time $= 47.50$ min, initial As(III) and Hg(II) concentrations = 2.75 mg/L , and adsorbent dose = 0.15 g/L , the percentage removal of As(III) and Hg(II) was above 94.00%. The best ft model was the Langmuir, whereas Lagergren's pseudo-frst-order and modifed Freundlich kinetic models were very well represented by the results of the kinetic data of As(III) and Hg(II), correspondingly. The thermodynamics, recovery of the adsorbed metals ions, and reuse of the adsorbent were not reported in this study.

The concurrent depollution of Cd(II), Hg(II), and As(III) from aqueous systems was achieved using graphene oxide (GO) treated with 3-aminopyrazole (GOf) (Alimohammady et al., [2017](#page-26-9)). The CCD of the RSM was used to analyze the infuence of solution pH, the dosage of adsorbent, and beginning metal ions concentration on the batch sorption system. The GO-f adsorbent was found to have maximum uptake capacities of 285.71, 227.27, and 131.58 mg/g for Cd(II), Hg(II), and As(III) ions, respectively. Furthermore, the heavy metal ions' competitive uptake capabilities were observed to be lower than their noncompetitive counterparts. For both noncompetitive and competitive adsorption, the same affinity order was observed: $Cd(II) > Hg(II) > As$ (III). The pseudo-second-order model shows a good representation of time-dependent experimental data, and the Langmuir model is well correlated with the equilibrium data. The decontamination of $Cd(II)$, $Hg(II)$, and $As(III)$ by the adsorbent was endothermic and spontaneous as per thermodynamic studies. The GO-f adsorbent was regenerated and reused, and the results indicate $Cd(II)$, $Hg(II)$, and As(III) percentage removal declined by less than 11.00% during one to three consecutive cycles. Hence, this adsorbent has high potentiality and application for heavy metals elimination from contaminated waters.

The effect of different parameters on As(III) sorption onto organically modifed montmorillonite clay was explored, involving dosages of adsorbent and surfactant, As(III) initial concentration, and time of contact (Bandpei et al., [2017](#page-27-13)). To determine the effect of independent factors and identify the optimal condition, a CCD was used under the RSM. The maximum As(III) removal (95.95%) was accomplished at the optimal operating point: an adsorbent dose of 3.70 g/L, surfactant dosage of 3 g/L, and 37.2 min

time of contact. The experimental results obtained closely matched those predicted by the model. The equilibrium adsorption isotherms, kinetics, thermodynamics, renewability, and reuse of the adsorbent were not considered in this study.

The applicability of modifed bentonite and chitosan (MBC) for As(V) removal was performed as a function of solution pH, sorbent dose, As(V) concentration, and time of contact using CCD of the RSM (Dehghani et al., [2017](#page-27-14)). The ANOVA aspect of CCD suggests that the quadratic model was very signifcant, with the optimal condition being solution pH (3.70) , sorbent dose (1.40 g/L) , beginning As(V) concentration (69.00 mg/L), and time of contact (167.00 min). The Langmuir and pseudo-second-order models were found to demonstrate a good representation of the experimental data. The thermodynamic investigations indicate the spontaneity and endothermic nature of the sorption process. The desorption and regeneration of the used adsorbent were not reported in this study.

The CCD of the RSM was applied to investigate the suitability of *Ficus benghalensis* leaf powder for adsorptive depollution of Co(II) from aqueous solutions (Hymavathi & Prabhakar, [2017\)](#page-29-11). The optimal conditions of 20.00 mg/L Co(II) starting concentration, 25.00 g/L adsorbent dose, pH of 5.00, and temperature of 303.00 K resulted in 98.73% removal of Co(II). The Langmuir isotherm proves to be a more accurate model of the equilibrium data. The optimal condition of 20.00 mg/L Co(II) starting concentration, 25.00 g/L adsorbent dose, pH of 5.00, and temperature of 303.00 K resulted in 98.73% removal of Co(II). The Langmuir isotherm and pseudo-secondorder kinetic prove to be more accurate models of the experimental data. The thermodynamics studies indicate that the process proceeded spontaneously and endothermically. The desorption and renewability of the exhausted adsorbent were not reported even though *Ficus benghalensis* L. was regarded as a good, cheap, and easily accessible adsorbent for Co(II) contaminated water treatment.

Chitosan beads (CS) produced from chitosan fakes, cross-linked by glutaraldehyde, and then grafted by means of ethylenediaminetetraacetic acid were employed to remove Cr(VI) using the CCD under the RSM (Igberase et al., [2017\)](#page-29-12). With a solution pH of 5.00, contact time of 70.00 min, a temperature of 45.00 $^{\circ}$ C, an adsorbent dose of 5.00 g/L, and an initial Cr(VI) concentration of 70 mg/L, a maximum uptake capacity of 154.87 mg/g was reached. In this study, the equilibrium isotherms, kinetics, and thermodynamics were not reported. However, the adsorbent was renewed efectively, and the recovered metal ions were disposed of safely to prevent the creation of secondary contaminants.

Natural clinoptilolite (NC), bentonite (NB), modifed clinoptilolite (MC), and modifed bentonite (MB) were utilized in the elimination of Cd(II) from aqueous environments (Kashi et al., [2017](#page-29-13)). To determine the infuence of pH of the aqueous solution and time of contact on the rate of sorption, the RSM was utilized in conjunction with the CCD for NC and NB adsorbents. The Cd(II) quadratic model was extremely signifcant as shown by very low *p*-values, according to statistical analysis. The pH of 5.35 and 3.89, as well as contact durations of 20.49 and 16.27 h, were determined to be optimal for Cd(II) removal on NC and NB adsorbents, with removal efficiencies of 94.86% and 87.42%, respectively. The models Jossesns, Unilan, Baudu, and Freundlich were chosen as the best for the experimental data. The sorption kinetics, thermodynamics, desorption, and reusability of the exhausted adsorbent were not explored.

A study was carried out to examine the ability of cyanobacterial biomass in the extraction of Cu(II) ions from synthetic wastewater (Kushwaha & Dutta, [2017\)](#page-30-11). The effects of many experimental factors including Cu(II) initial concentration, pH of the solution, and dosage of adsorbent on the sorption process have been investigated. The CCD of the RSM was applied in the optimization of the process factors utilizing both the dried and carbonized biomass. With both adsorbents, maximal removal (95.08%) was realized at the best operating condition of 20.00 mg/L $Cu(II)$ initial concentration, 5.00 g/L dosage of adsorbent, and solution pH of 6.00. The Langmuir and pseudo-second-order models show a good ftness to the attained experimental data. More so, the renewability of the used adsorbents was performed, and over 70.00% and 80.00% of the adsorbed Cu(II) ions were retrieved successfully by the dried and carbonized biomass, respectively, in the third cycle of the desorption-regeneration study. The thermodynamics parameters (Gibbs free energy, enthalpy, and entropy changes) were not reported.

The adsorption efficiency of activated carbon obtained from waste tires for Hg(II) elimination was explored as a function of some infuencing factors namely solution pH, contact time, initial concentration, and solution temperature (Saleh et al., [2017](#page-32-5)). The CCD was used to fnd the best experimental condition under RSM. The results indicate that the optimal setting for maximal removal (90.00%) includes a contact period of 35.00 min, a dosage of 0.06 g/L, a solution pH of 5.00, and an initial Hg(II) concentration of 25.00 mg/L at 100.00 rpm speed of swirling. In analyzing the data obtained, the Langmuir and pseudo-second-order were the best-correlated models. The negative enthalpy and Gibbs free energy changes indicate the exothermic and non-spontaneous nature of the adsorption system. Also, the desorption-recycling of the used adsorbent was done, and 95.00% metal ions recovery was reached even up to the third cycle. Hence, the adsorbent is highly capable, lucrative, and selective for removing Hg(II) from wastewater samples.

The reduction of $Cu(II)$, $Ni(II)$, and $Pb(II)$ using an activated carbon obtained from banana peels was optimized utilizing RSM involving the CCD (Van Thuan et al., [2017\)](#page-32-6). The interactive impact of three factors on the adsorption capacity such as metal ions concentration, solution pH, and dosage of adsorbent were maximized in the CCD investigation. The greatest adsorption capacity appeared to take the following sequence: 14.30, 27.40, and 34.50 mg/g for Cu(II), Ni(II), and Pb(II), respectively, which was comparable with the predicted results by the CCD. Furthermore, the isotherm analyses reveal that the Langmuir model best describes the sorption behavior of Cu(II) and Ni(II) on the banana peel-activated carbon. The kinetics, thermodynamics, desorption, and regeneration of the used adsorbent were not reported. Besides, the study demonstrates that the banana peel has a high ability in eliminating metal ions from aqueous media.

The CCD-RSM technique was applied to optimize Hg(II) detoxifying operating conditions in a batch system using 3-mercaptopropyl trime-thoxysilane-modifed kaolin (MMK) adsorbent. (Yılmaz et al., [2017](#page-33-3)). The best condition for Hg(II) removal, according to the quadratic model developed from the CCD in the RSM was attained at 30.83 mg/L Hg(II) initial concentration, 0.10 g/L dosage of adsorbent, solution pH of 7.44, and a temperature of 31.41 °C. The highest amount of Hg(II) adsorbed and removal rate under the optimal setting was obtained as 30.10 mg/g and 98.01%, respectively. The Langmuir and Dubinin–Radushkevich models show good ftness to the obtained data. The removal of Hg(II) was spontaneous, physical, and exothermic according to thermodynamic studies. The kinetics, desorption, and regeneration of the used adsorbent were not reported, but the results show

MMK has a lot of possibility for removing Hg(II) from aqueous systems.

The impact of operating settings on Mn(II) reduction from wastewater utilizing zero-valent iron nanoparticles was investigated (Agarwal et al., [2016\)](#page-26-10). The parameters that had an efect on Mn(II) removal were optimized using a multi-step CCD of the RSM. The highest elimination of Mn(II) was found as 92.50% for a time period of 6.00 h at pH 9.00, a temperature of 25.00 °C, 5.00 g/L dosage of adsorbent, and $Mn(II)$ baseline concentration of 2.07 mg/L. The equilibrium isotherms, kinetics, thermodynamics, desorption, and regeneration of the used adsorbent were not investigated in this study.

The detoxifcation of Cr(VI) from wastewater was performed by employing sulfate-reducing bacteria as the adsorbent and some independent factors afecting Cr(VI) removal underwent optimization using a CCD of the RSM (Ahmadi et al., [2016](#page-26-11)). The optimum condition was found as solution pH of 7.50, 130.00 mg/L Cr(VI) initial concentration, and 7.75% inoculation, and the optimum detoxifcation of Cr(VI) were given as 82.00%. The pseudo-frst-order model was well correlated with Cr(VI) experimental results. The equilibrium isotherms, thermodynamics, desorption, and renewability of the used adsorbent were not reported.

The best condition for removing $Pb(II)$ and $Cu(II)$ from aqueous phases by defatted papaya seeds (DPS) was investigated (Garba et al., [2016\)](#page-28-8). The influences of three independent factors (adsorbent dose, shaking speed, and initial metal ions concentration) were optimized by CCD under the RSM. The ideal adsorption condition attained was 0.30 g/L dosage of adsorbent, 180.00 rpm speed of shaking, 150.00 mg/L Pb(II), and Cu(II) initial concentration with 0.987 desirability value. The maximal removal rates attained at this ideal condition were 99.96 and 97.55% for Pb(II) and Cu(II), respectively. The Cu(II) and Pb(II) equilibrium data were found to be better suited by the Langmuir isotherm model, yielding 17.29 and 53.02 mg/g, respectively as monolayer uptake capacities. The sorption kinetics, thermodynamics, regeneration, and reusability of the used adsorbent were not discussed in this study.

Natural zeolite was used in the decontamination of Ni(II) from wastewater using the CCD of the RSM (Hosseini et al., 2016). The optimal condition for Ni(II) uptake was obtained as 10.00–15.00 mg/L Ni(II) initial concentration, 0.37–0.43 g/L dosage of adsorbent, 56.00–68.00 min time of contact, and solution pH of 4.80–6.00. The equilibrium isotherms, kinetics, thermodynamics, regeneration, and reusability of the used adsorbent were not investigated. However, based on the experimental fndings and model variables, it could be concluded that this adsorbent with a relatively high uptake capacity may be used to remove Ni(II) from aqueous media.

A study uses date palm fber modifed with acid to find the best possible condition for removing Cr(VI) from aqueous systems through biosorption (Hossini et al., [2016\)](#page-28-10). The CCD in the RSM was used to optimize three infuencing variables namely solution pH, Cr(VI) initial concentration, and dosage of the biosorbent. At the optimal operating condition of solution pH 3.30, Cr(VI) initial concentration of 180.00 mg/L, and 0.80% (w/v) dosage of the biosorbent, the highest Cr(VI) uptake of 95.00% was realized. The Langmuir and pseudo-second-order models both provide a detailed description of the experimental data. The thermodynamics parameters, desorption, regeneration, and reusability of the used adsorbent were not considered in this study.

A CCD of the RSM was employed in the optimization of pH of the solution, Cd(II) initial concentration, adsorbent dose, and sample volume for the efective elimination of Cd(II) ions (Ince et al., [2016\)](#page-29-14). The efect of relevant factors on one another was determined by ANOVA. According to the process optimization, solution pH of 8.50, 166.00 mL volume of sample, 0.57 g/L dosage of adsorbent, and 82.00 min time of contact were best for Cd(II) removal, and the adsorption rate of Cd(II) was 895.00 mg/g under this condition. The Freundlich model represents the attained equilibrium data of Cd(II). The sorption kinetics, thermodynamics parameters, desorption, regeneration, and reusability of the used adsorbent were not examined in this study.

The possibility of employing shoe waste adsorbents (polyurethane ethylene-shoe material type-I and vinyl acetate-shoe material type-II) was examined for Cd(II) reduction from synthetic wastewater (Iqbal et al., [2016\)](#page-29-15). The CCD of the RSM was harnessed to estimate the effect of operational factors for example $Cd(II)$ concentration, the dosage of adsorbent, solution pH, and time of contact. For shoe type-I, the optimum condition for the reduction of Cd(II) was attained as 305.00 mg/L Cd(II) initial concentration, solution pH of 4.90, 932.00 min time of contact, and 1.30 g/L dosage of adsorbent whereas, for shoe type-II, the optimum condition was Cd(II) initial concentration of 402.00 mg/L, solution pH of 5.00, 881.00 min time of contact, and 1.20 g/L dosage of the adsorbent. At these optimized conditions, optimum Cd(II) uptake capacities of 180.22 mg/g (66.66%) and 396.31 mg/g (94.66%) ions were achieved by shoe materials I and II, respectively. The Freundlich and pseudo-second-order models show a good representation of the obtained data. The thermodynamics parameters, retrieval, regeneration, and reusability of the used adsorbent were not reported in this study.

A highly selective 2-hydroxyethyl ammonium sulfonate immobilized on γ -Fe₂O₃ nanoparticles (γ -Fe₂O₃-2-HEAS) as magnetic nano-adsorbent was produced and explored in removing Pb(II) from aqueous solutions (Khani et al., [2016\)](#page-29-16). The CCD based on the RSM was utilized to examine the efects of three diferent process factors on Pb(II) removal, including pH, shaking time, and adsorbent dose. The best condition for maximum Pb(II) sequestration (80.00%) was 11.70-min shaking time, pH 6.50, and 0.02 g/L adsorbent dosage, by the optimization procedure. The Freundlich model best describes the isothermal data. The thermodynamic studies show that Pb(II) decontamination was exothermic, spontaneous with increasing randomness on the adsorbent surface. The sorption kinetics of Pb(II) decontamination by the adsorbent were not examined in this study. From the desorption and regeneration of the Pb(II) laden adsorbent, the removal rate ranged between 68.00 and 77.00% after four cycles. Therefore, the study reveals that the synthesized composite is highly effective in Pb(II) decontamination, signifying that it could be used in the water treatment industry.

A CCD of the RSM was employed to optimize As(V) depollution from aqueous environments onto acid-treated magnetic nanoparticles (Nikraftar & Ghorbani, [2016](#page-30-12)). The infuence of solution pH, the temperature of the solution, initial As(V) concentration, and sorbent dose were studied. The optimal solution pH, As(V) initial concentration, and adsorbent dose were 2.00, 5.00 mg/L, and 0.10 g/L, respectively, producing an optimum uptake capacity of 44.99 mg/g and an optimum removal efficiency of 42.69% with a desirability of 0.862. The Langmuir and pseudo-second-order models provide a good correlation of the attained data. The thermodynamic variables show the process spontaneous, endothermic, and high affinity of As(V) ions for the adsorbent. More so, the successive regeneration cycles indicate that the adsorbent has profcient desorption and reusability possibilities. Hence, the adsorbent is anticipated to have extensive applications in the elimination of various heavy metals from wastewater.

The sorption potential of an adsorbent derived from tamarind wood and chemically treated with zinc chloride was investigated to remove Pb(II) ions from aqua systems (Sahu et al., 2016). The Pb(II) detoxification was optimized using a CCD of the RSM. The ideal condition was obtained as a 1.44 g/L dosage of adsorbent, a temperature of 50.00 \degree C, 49.23 mg/L Pb(II) initial concentration, and a solution pH of 4.07. The Pb(II) elimination was reported to be greater $(>99.00\%)$ at optimized process conditions. The equilibrium isotherms, kinetics, thermodynamics, desorption, and reusability of the used adsorbent were not investigated. The RSM demonstrates to be one of the most efective strategies for optimizing operating conditions and maximizing Pb(II) removal.

The adsorbents derived from custard apple seeds and *Aspergillus niger* were employed in the decontamination of Cr(VI) and Ni(II) from wastewater (Saravanan et al., [2016](#page-32-7)). The CCD of the RSM was applied to study the efects of various operational factors including metal ions' initial concentration, temperature, solution pH, and biomass dose. The determined ideal situation for Cr(VI) removal was obtained as an initial concentration of 100.00 mg/L, solution pH of 3.00, a temperature of 36.00 °C; and 10.00 g/ L loading of the biosorbent, with a maximum Cr(VI) removal of 95.70%. However, the best condition for Ni(II) removal was an initial concentration of 100.00 mg/L, solution pH of 5.60, a temperature of 30.00 °C; and 10.00 g/L loading of the biosorbent. The highest elimination of Ni(II) was found to be 96.41% under the optimal setting. The equilibrium isotherms, kinetics, thermodynamics, desorption, and reusability of the used adsorbent were not discussed. However, when it comes to removing $Cr(VI)$ and $Ni(II)$ ions from aqueous systems, the mixed biosorbent was found to have better adsorption characteristics.

A hydrochar was produced hydrothermally using maize straw, and CCD-RSM was used to identify the best char with the maximum Pb(II) removal capacity (Sun et al., 2016). The impacts of hydrothermal temperature, duration, solid–liquid ratio, and the combined efect of hydrothermal temperature and duration were all found to be signifcant for the Pb(II) removal, per the statistical analysis. The following condition was determined to be best for obtaining maximum Pb(II) adsorption capacity: hydrothermal temperature (205.00 °C), duration (28.00 min), solid–liquid ratio (12.00), and Pb(II) removal capacity of 47.00 mg/g. The Langmuir and pseudo-second-order models present a better ft to the obtained data. The thermodynamic variables show the process spontaneous, endothermic, and high affinity of Pb(II) ions for the adsorbent. The retrieval, regeneration, and reusability of the used adsorbent were not reported in this study.

The RSM based on CCD was employed in the analysis of operating factors (adsorbent dose, Ni(II) initial concentration, and solution pH) infuencing Ni(II) removal by dried *B. cereus* (Zhang et al., [2016](#page-33-4)). At a solution pH of 4.00, a biomass concentration of 2.00 g/L, and an initial Ni(II) concentration of 150.00 mg/L, the maximum Ni(II) uptake rate was established. The greatest amount of Ni(II) adsorbed and percentage removal was 20.79 mg/g and 41.44%, respectively, under this maximized condition. Here, the equilibrium isotherms, kinetics, thermodynamics, desorption, and reusability of the used adsorbent were not discussed. However, the dried *B. cereus*, as an environmentally friendly biosorbent, performed admirably in the removal of Ni(II).

Fraxinus tree leaves as a biosorbent of As(III) were investigated using CCD of the RSM (Zolgharnein et al., [2016](#page-33-5)). Simultaneous optimization of two responses (removal efficiency $(\%)$ and adsorption capacity (mg/g) was performed using the desirability function. The simultaneous optimization yielded 70.00% removal efficiency and 80.60 mg/g uptake capacity with 67.00% desirability with an initial As(III) concentration of 600.00 mg/L, adsorbent dose of 0.10 g/L, and pH 3.90. The sorbent-sorbate attained data were best explained by the Langmuir, Freundlich, and pseudo-second-order models. The thermodynamics, desorption, and reusability of the used adsorbent were not discussed.

Modeling and optimization of heavy metals removal using Box–Behnken design (BBD)

For the extraction of Cr(VI) from synthetic wastewater, a combination of coconut shell and the coir has been used, and the statistical tool Box–Behnken design (BBD) of the RSM was utilized to optimize the process factors such as solution pH, adsorbent quantity, and reaction time (Kumari et al., [2021](#page-30-13)). For 99.00% of Cr(VI) reduction, the optimal pH, adsorbent quantity, and reaction duration were established to be 2.00, 0.10 g/L, and 100.00 min, respectively. The Freundlich isotherm and pseudo-second-order models were found to have the best correlation with the experimental data. The endothermic and spontaneous characteristics of the adsorption system were confrmed by the positive enthalpy and negative Gibbs free energy values. Based on this study, it was concluded that the adsorbent is good at removing Cr(VI) and that it could be employed on a larger scale for human and ecological benefts. This is attributable to the fact the adsorbent has regenerative properties and can be reused three to four times and gave more than 60.00% removal of Cr(VI) at the fourth cycle.

The production of Fe-impregnated biochar from food waste (Fe-FWB) was maximized for As(III) removal using BBD of the RSM, and the pyrolysis time, temperature, and Fe concentration were considered independent factors (Lyonga et al., [2021\)](#page-30-14). The results demonstrate that pyrolysis temperature and Fe concentration had a considerable impact on As(III) elimination, but pyrolysis time had little efect. The maximum condition for the biochar production was 1.00 h at 300.00 °C with 0.42 M Fe concentration. The Langmuir and pseudosecond-order models show good ftness to the As(III) experimental data. The thermodynamic studies suggest As(III) removal was spontaneous, endothermic, and caused augmented randomness at the solid–liquid interface during the process. In this study, the retrieval of the ions on the impregnated adsorbent and its recyclability was not carried out. However, the study indicates that Fe-FWB may be used to remove As(III) from aqueous solutions and that it is a cost-efective and easily available resource.

Taşdemir et al. [\(2021\)](#page-32-9) used the BBD of the RSM to study the optimization of adsorption variables including initial Pb(II) concentration, pH, and temperature. The adsorption process fts the reduced cubic model, according to the optimization results. At pH 8.00, the temperature of 25.00 $^{\circ}$ C, and an initial Pb(II) concentration of 480.70 mg/L, maximum removal of 79.70% was obtained using a cross-linked polycarboxylate-based adsorbent with a maximal Pb(II) adsorption uptake of 255.40 mg/g by the adsorbent. The equilibrium data ftted well to both the Langmuir and Freundlich models; additionally, the adsorption mechanism was more preferential at greater initial Pb(II) concentrations. The negative Gibbs free energy, enthalpy, and entropy changes show spontaneous, exothermic, and decrease in randomness on the adsorbent surface. In this study, the desorption of the metal ions on the impregnated adsorbent and its recyclability were carried out suggesting

the adsorption process was highly chemical, and the adsorbent was found to be efficient.

The BBD of the RSM was employed to determine the infuences of some operating factors (pH, time, agitation, and temperature) on Cr(VI) removal by *Bauhinia rufescens* pod-activated carbon (BRPAC) (Alabi et al., [2020](#page-26-12)). The highest adsorption uptake and removal rate of Cr(VI) were found as 9.762 mg/g and 87.62%, respectively, attained at 6.00 solution pH, 75.00 min time of contact, agitation speed of 80.00 rpm, and temperature of 32.50 °C. The pseudo-second-order model was well represented by the experimental data. The equilibrium isotherms, thermodynamics, recovery, and reusability of the used adsorbent were not tested, but BRPAC was suggested as a new biosorbent for heavy metals remediation from the aquatic systems.

Temperature, contact time, sorbent dose, solution pH, and initial Cd(II) concentration were all examined in the removal of Cd(II) from aquatic systems by orange peels (OP) using RSM-BBD (Akinhanmi et al., [2020](#page-26-13)). At a contact time of 120.00 min, an initial Cd(II) concentration of 240.00 mg/L, an adsorbent dose of 0.04 g/L, a temperature of 45.00 $^{\circ}$ C, and a solution pH of 5.50, the maximum uptake of Cd(II) was achieved. The best ft Langmuir model suggests that the adsorbent had an uptake capacity of 128.23 mg/g. The pseudo-frst-order model shows a good correlation with the kinetic data. The thermodynamic parameters (enthalpy and entropy) indicate the elimination of Cd(II) by the adsorbent was nonspontaneous, endothermic (physisorption), and the level of disorderliness reduced as the temperature increased. The reusability of the OP was investigated, and it was found that as the number of regeneration cycles increased, the uptake capacity of the OP declined from 88.34 to 73.42%, which makes the adsorbent more economical and efficient.

Activated carbon was made from commercial polyurethane and polymer wastes using zinc chloride in removing Cu(II) from aqueous solutions (Arslanoğlu et al., [2020\)](#page-26-14). The experimental factors of initial Cu(II) concentration, solution pH, temperature, and adsorbent dose were optimized by BBD of the RSM. The optimal condition was found as the solution pH of 5.70, a temperature of 53.00 \degree C, Cu(II) initial concentration of 27.00 mg/L, and an adsorbent dosage of 4.20 g/L, which resulted in an 80.00% Cu(II) removal. The Langmuir and pseudo-second-order models were the ideal models for the sorption process. It was established that the adsorption system is spontaneous, endothermic, and physical. The desorption of the metal ions on the impregnated adsorbent, and its recyclability were not conducted. However, the results show that the activated carbon produced is capable of removing Cu(II) ions from aqueous solutions.

The infuence of various independent factors including solution pH, contact time, Cr(VI) concentration, and adsorbent dose on the Cr(VI) elimination by modifed sesame hull was studied using the BBD-RSM (Ghasemi et al., [2020\)](#page-28-11). The best operating condition was attained at pH of 3.00, contact time of 120.00 min, initial Cr(VI) concentration, and adsorbent dose of 4.94 g/L, which gave 100.00% removal of Cr(VI). The experimental results were found to follow the Langmuir and pseudosecond-order models with a very good coefficient of determination. The thermodynamics, desorption, and recyclability of the used adsorbent were not examined, but the study suggests that the treated sesame hull could be applied as an effective adsorbent for Cr(VI) elimination from aqueous phases.

Activated carbon from jute stick (*Corchorus olitorius*) was utilized for eliminating Cd(II) from waste-water (Ghosh et al., [2020](#page-28-12)). The interactive effects of independent adsorption factors such as Cd(II) initial concentration, solution pH, adsorbent dose, and agitation time were investigated by applying RSM. Using the BBD matrix model under RSM indicates that the optimum Cd(II) decontamination was realized at an initial concentration (60.87 mg/L), dosage (0.50 g/L), solution pH (7.00), and agitation time (30.00 min). The Langmuir model provided the best ft to the equilibrium data. The adsorbent had a high Cd(II) uptake ability of 73.53 mg/g. The adsorbent was regenerated and reused for fve consecutive cycles. Nonetheless, the adsorption kinetics and thermodynamics that could have provided further information on the metal removal were not investigated.

The infuence of experimental factors including pH solution, temperature, Cr(VI) initial concentration, and contact time was studied through BBD-RSM using sagwan sawdust biochar (Gupta & Mondal, [2020\)](#page-28-13). The optimal condition for the maximal removal was a pH of 2.07, a temperature of 30.72 °C, and Cr(VI) initial concentration of 160.93 mg/L. The Langmuir and pseudo-second-order dominated the equilibrium data of Cr(VI) with an uptake capacity of 9.62 mg/g. The process was shown to be exothermic and spontaneous by the thermodynamics analysis. The Cr(VI)-laden adsorbent was recycled efectively and then applied for Cr(VI) removal for the next 3 cycles. The study suggests that the sagwan sawdust biochar is capable of decontaminating Cr(VI) from aqueous environments.

A study was carried out to prepare copper(II) oxides (CuO) using leaf, dendrite, and feather morphologies for Zn(II) extraction from aqueous media (Jamileh et al., [2020\)](#page-29-1). The impact of the diferent CuO morphologies was studied and feather morphology was the most efficient of the morphologies reported in the study. To determine the factors (CuO feather dose, pH of the solution, and time of contact) in $Zn(II)$ elimination, the BBD of the RSM was employed. At the best condition of adsorbent dosage (0.14 g), pH of the solution (7.99), and time of contact (44.78 h), the maximum Zn(II) removal reached was 99.98%. The kinetic data were well correlated with the pseudo-secondorder model. The equilibrium isotherms and thermodynamics as well as regeneration of the adsorbent were not discussed, which could have ofered more insight into the sorption behavior of Cr(VI) by the adsorbent.

Okolo et al. [\(2020\)](#page-30-15) studied to optimize the reduction of Pb(II) from simulated wastewater employing mucuna shell, Africa elemi seed, and oyster shell treated with orthophosphorous acid and utilized. The efects of process factors such as dosage, solution pH, and time of contact were examined by the BBD-RSM approach. The optimized operational condition for Pb(II) optimum removal was attained at pH 2.00, adsorbent dose of 1.00 g/L, and time of contact 70.00 min for Africa elemi seed adsorbent. For both mucuna and oyster shell adsorbents, the optimum condition was determined as $pH = 6.00$, adsorbent dose = 1.00 g/L, and contact $time = 40.00$ min. The equilibrium isotherms, kinetics, thermodynamics, and regeneration of the impregnated adsorbents were not investigated in this study, and this could have given more insight into the $Pb(II)$ biosorption mechanism. Nevertheless, the study reveals that these adsorbents can successfully remove Pb(II) ions from industrial wastewaters.

Demir et al. (2019) evaluated the efficacy of using an adsorbent obtained from discarded Turkish coffee grounds in the purifcation of wastewater contaminated by Cd(II) ions. The BBD, which is a subclass of the RSM, was applied to assess the impact of the various independent factors on the Cd(II) rate of removal. When the desirability function approach was used, the optimum Cd(II) removal rate of 96.00% and uptake capacity of 1.32 mg/g was reached at an adsorbent dose (3.63 g/L) , initial Cd(II) concentration (67.97 mg/L) , and solution pH (8.87). The Langmuir and pseudosecond-order models were compatible with the Cd(II) adsorption data. The desorption and regeneration of the used adsorbent were tested and the results indicate that after fve cycles of adsorption/desorption/regeneration, the adsorbent maintained 90.00% of uptake capacity for Cd(II) removal. However, adsorption thermodynamics studies were not reported.

To eliminate Zn(II) ions from aqueous systems, diferent component powders of rape straw were used as adsorbents (Liu et al., [2019\)](#page-30-16). The efects of several parameters on $Zn(II)$ removal efficiency were explored, and the operating settings were optimized by the BBD-RSM. When the optimum conditions were reached, the removal efficiencies of $Zn(II)$ via straw pith core, seedpods, and rape straw shell were 100.00%, 78.02%, and 17.00%, respectively. The Langmuir model best described the equilibrium data, indicating monolayer adsorption, while the pseudo-second-order model described the kinetic data. The thermodynamics, desorption, and regeneration of the used adsorbent were not investigated in this study.

The BBD under the RSM was used to optimize Cr(VI) removal by alkali-treated chicken feathers (Mondal et al., [2019a](#page-30-17)). About 89.00% of Cr(VI) was adsorbed at the optimal operating condition of initial concentration (5.64 mg/L) , adsorbent dose (0.15 g/L) , contact time (29.60 min), and solution pH (1.06). The adsorption data follows the pseudo-second-order model as well as the Langmuir and Freundlich models. The thermodynamics, desorption, and regeneration of the used adsorbent were not investigated in this study. Even though the results indicate that the adsorbent could be considered a cheap option for detoxifying Cr(VI) from aqueous media.

Waste mosambi peel dust was utilized to explore the biosorption of Cr(VI) from aqueous systems, and the BBD-RSM approach was used as an optimization technique (Mondal et al., [2019b](#page-30-18)). The optimized process condition for Cr(VI) reduction was found as pH $= 2.00$; dose $= 10.00$ g/L, Cr(VI) initial concentration $= 5.00$ mg/L, time of contact $= 30$ min, and agitation speed $= 150$ rpm. The D–R isotherm and pseudosecond-order model best suit the obtained data. The thermodynamic results show that Cr(VI) removal was spontaneous and endothermic, indicating chemisorption. The desorption and regeneration of the adsorbent were not reported. Nonetheless, the study found that the adsorbent is efective and that its adaptability makes it an environmentally benign option for agricultural waste treatment.

In a batch investigation, a magnolia leaf was applied to remove Cr(VI) from simulated solutions (Mondal et al., [2019c](#page-30-19)). A multi-step RSM was employed to investigate the efects of the operating factors on the Cr(VI) biosorption. The BBD determined the best biosorption condition as an initial 40.00 mg/L Cr(VI) concentration, solution pH of 2.00, a contact period of 45.00 min, and a dosage of 0.50 g/L with an optimum biosorption rate of 3.96 mg/g and removal rate of 98.80%, respectively. The data obtained correlates well with the pseudo-second-order model and provided a good ft for the Langmuir model. The exothermic and spontaneous behavior of the biosorption process was demonstrated by the thermodynamic variables. The retrieval and reusability of the used adsorbent were tested, and an optimum 83.00% of Cr(VI) can be recovered showing that the adsorbent is both effective and cost-efficient.

The infuences of important process factors including solution pH, initial Pb(II) concentration, and time of contact on the adsorption system were ascertained using a three-level BBD of the RSM, which was then used to optimize Pb(II) reduction by $C_{16-6-16}$ incorporated mesoporous MCM-41 adsorbent (Saini et al., [2019\)](#page-31-9). The quadratic model was recommended by the BBD, and process optimization was carried out, yielding a theoretical maximum uptake capacity (81.92 mg/g) at the optimal condition of 99.02 mg/L Pb(II) initial concentration, pH of 5.85, and 118.49 min time of contact with the desirability of 1.00. The equilibrium, thermodynamics, desorption, and regeneration of the used adsorbent were not investigated in this study.

The reduction of $Pb(II)$ ions in a batch system by date palm seeds (*Phoenix dactylifera* L.) was performed, and the biosorption optimization was done by RSM (Çetintaş & Bingöl, [2018](#page-27-16)). The BBD of the RSM was used to evaluate the impact of the process factors and at the best optimal condition of $pH = 5.00$, initial $Pb(II)$ concentration = 100.00 mg/L, and biosorbent dose $= 0.10 \text{ g/L}$, an optimum uptake capacity (24.07 mg/g) of Pb(II) was achieved. The Langmuir and pseudosecond-order models correlate well to the isothermal and kinetic data of Pb(II), respectively by the date palm seeds. The thermodynamics, regeneration, and reusability of the used adsorbent were not reported.

The sol–gel process was utilized to produce a nanocomposite of manganese oxide and was employed in removing Fe(III) from wastewater (Langeroodi et al., [2018\)](#page-30-20). The impacts of solution pH, initial Fe(III) concentration, the dosage of adsorbent, and time of contact were investigated using the BBD-RSM approach. The maximum removal of Fe(III) was achieved at 95.80% with the following ideal parameters: contact period of 62.50 min, initial Fe(III) concentration of 50.00 mg/L, the adsorbent weight of 0.50 g/L, and pH of 5.00. The Langmuir and pseudo-second-order models best describe Fe(III) sorption data. The thermodynamics, desorption, and regeneration of the used adsorbent were not reported in this study, but the adsorbent was regarded as one of the reliable and inexpensive adsorbents for heavy metals decontamination from various aqueous systems.

In analyzing the efects of solution pH, initial metal ions concentration, and adsorbent dose, the BBD of the RSM was used to examine As(III) and As(V) ions removal from wastewater by $CeO₂/Fe₂O₃/gra$ phene nanocomposite (Sahu et al., [2018](#page-31-10)). The ideal condition for maximum As(III) removal was solution pH of 7.84, 10.52 mg/L As(III) initial concentration and, an adsorbent dose of 0.198 g/L, while optimum As(V) reduction was reached at solution pH of 3.05, 10.78 mg/L As(V) initial concentration, and adsorbent dose of 0.20 g/L. Under these optimal conditions, the removal percentages of As(III) and As(V) were determined as 98.53% and 97.26%, respectively. For both As(III) and As(V) eradication, the Langmuir isotherm was the preferred model. The adsorption kinetics, thermodynamics, recyclability, and reuse of the adsorbent were not studied though the results demonstrated an excellent efficiency of the $CeO₂/Fe₂O₃/graphene$ nanocomposite in removing arsenic from aqueous solutions using the RSM.

An adsorbent was developed by combing poly(ophenylenediamine) with hydrous zirconium oxide for the decontamination of Cd(II) from aqueous media (Rahman & Nasir, [2018\)](#page-31-11). The BBD-RSM was used to examine the infuence of solution pH, contact time, adsorbent dose, and Cd(II) initial concentration. Using the desirability function, the best variables for 99.60% Cd(II) elimination were: contact time $= 45.00$ min, solution pH = 6.00, adsorbent dose=1.25 g/L, and initial concentration of $Cd(II) = 50.00$ mg/L. The Freundlich isotherm was shown to be the fnest model for explaining Cd(II) removal based on the equilibrium data. According to the kinetic data, Cd(II) depollution by the adsorbent follows the pseudo-second-order,

Weber–Morris intraparticle difusion, and Bangham models. It was found that the adsorbent could be recycled and 99.35% of the adsorbed Cd(II) were desorbed from the adsorbent surface. According to the results, the adsorbent can be exploited as a possible adsorbent for removing Cd(II) from polluted water.

The BBD of the RSM has been used to maximize the reduction of Cr(VI) in aqueous systems employing *Litchi chinensis* (Uddin & Salah, [2018\)](#page-32-10). For the optimization procedure, three experimental factors were chosen: dosage, temperature, and pH. The results reveal that the actual and adjusted coefficient of determination values were relatively close, signifying that the model was successful in analyzing the experimental data. With low *p*-values, the linear terms, square values, and their two-way interaction were shown to be signifcant, implying that these factors play an essential role in Cr(VI) elimination. At a pH of 2.00, a dosage of 1.00 g/L, and a temperature of 40.00 °C, the highest Cr(VI) removal (96.00%) was achieved. In this study, the equilibrium isotherms, kinetics, and thermodynamics as well as the regeneration of the exhausted adsorbent were not discussed.

The simultaneous decontamination of Cr(VI) and phenol from bicomponent systems were conducted using tea waste biomass modifed by an iron (Gupta & Balomajumder, [2017](#page-28-14)). The infuence of process variables such as dosage of adsorbent, pH of the solution, Cr(VI) concentration, and phenol concentration were optimized using BBD-RSM. Based on the optimization process, 99.99% of Cr(VI) and phenol were removed successfully at an adsorbent dosage of 15.00 g/L, solution pH of 2.00, and initial Cr(VI) and phenol concentrations of 55.00 and 27.50 mg/L, respectively. In this study, the equilibrium isotherms, kinetics, thermodynamics, desorption, and renewability of the exhausted adsorbent were not carried out.

 $Fe₂O₄$ nanoparticles were utilized in removing As(III) from aqueous environments and BBD-RSM was employed to examine the infuence of several variables such as adsorbent dosage, initial As(III) concentration, and solution pH (Sahu et al., [2017\)](#page-31-12). The ideal condition for As(III) detoxifcation was determined as 0.70 mg/L dosage of adsorbent, solution pH of 7.70, and 33.32 mg/L As(III) initial concentration. Nearly 90.50% of As(III) was extracted from the aqueous solutions at this optimal condition. The Langmuir model shows good agreement with the data attained. The kinetics, thermodynamics, desorption, and the ability to reuse the adsorbent were not tested.

Nonetheless, the study shows that $Fe₃O₄$ nanoparticles could be utilized to decontaminate As(III) from aqueous systems in an environmentally acceptable and efficient manner.

The removal of As(V) by chitosan-coated bentonite (CCB) from groundwater in a fxed-bed column system was explored (Arida et al., [2016\)](#page-26-15). The BBD based on the RSM was used to optimize parameters including adsorbent dose, rate of flow, and initial concentration, as well as to assess the combined interactive impacts of these parameters on As(V) uptake capacity at the point of the breakthrough. The results demonstrate that at 418.12 μ*g*/g As(V) initial concentration, 6.6 g/L dosage of adsorbent, and 0.65 mL/min rate of fow, the maximum As(V) uptake capacity of 10.57 μ*g*/g at the point of the breakthrough was achieved. The equilibrium isotherms, kinetics, thermodynamics, recovery, and reusability of the used adsorbent were not investigated.

The adsorption capacity of *Ageratum conyzoide* leaf powder was determined for the reduction of Cr(VI) from aqueous solutions (Ezechi et al., [2016](#page-28-15)). Three infuencing factors, namely solution pH, initial Cr(VI) concentration, and adsorbent dose were investigated using BBD of the RSM. The optimal removal of Cr(VI) was 92.00%, which was achieved at a solution pH of 2.00, 50.00 mg/L Cr(VI) initial concentration, and 0.30 g/L dosage of adsorbent. The pseudo-second-order model correlates with the kinetic data, whereas the Langmuir model with an optimum uptake capacity of 437.00 mg/g best describes the equilibrium data. The thermodynamic variables point to a physisorption process that is spontaneous and exothermic. The desorption and renewability of the used adsorbent were not reported.

The optimization of Cr(VI) decontamination from synthetic wastewater onto *Enteromorpha* sp. immobilized in sodium alginate (ESA) and *Enteromorpha* sp. immobilized in polysulfone (EPS) were performed by a three-level BBD in the RSM (Rangabhashiyam et al., [2016](#page-31-13)). The BBD-RSM was applied to examine the efects of independent factors, namely solution pH, temperature, and Cr(VI) concentration. The optimized condition for Cr(VI) decontamination was determined as a solution pH of 2.00, a temperature of 318.00 K, and 20.00 mg/L Cr(VI) initial concentration with optimal removal rates of 90.52 and 90.86% for ESA and EPS, respectively. While solution pH of 8.00, a temperature of 318.00 K, and 20.00 mg/L initial Cr(VI) concentration was the best operating

condition for total Cr(VI) reduction, which gave maximal removal percentages of 81.14 and 79.90% for ESA and EPS, respectively. The equilibrium isotherms, kinetics, thermodynamics, recovery, and reusability of the used adsorbent were not investigated. However, the study implies that ESA and EPS could be efective biosorbents for eliminating Cr(VI) from aqueous systems.

Modeling and optimization of heavy metals removal using factorial experimental design (FED) and artificial neural network (ANN)

A two-step technique was employed in the optimization of Cr(VI) depollution in a fxed packed bed column using *Sargassum* sp. (Prabhu et al., [2021](#page-31-14)). According to the ANOVA, the baseline metal ion concentration has a greater infuence on the adsorption rate. The use of the artifcial neural network-genetic algorithm (ANN-GA) to optimize the models yielded good results. With Cr(VI) initial concentration of 25.00 mg/L, bed height of 11.97 cm, and fow rate of 5.29 mL/min, the biosorption efficiency increased to 90.79%. The adsorption behavior was explained using kinetic models, with the Yoon–Nelson model and bed depth service time providing a good fit to the kinetic data. Furthermore, efforts were made to retrieve the metal ions from the saturated column and metal retrieval of 79.00% was accomplished efectively. In this study, the equilibrium isotherms and thermodynamics were not explored, which could have ofered more insight into the sorption behavior of Cr(VI). Nonetheless, the study creates a greater picture of applying this priceless immobilized *Sargassum* sp. biomass for an industrial scale continuous heavy metals removal.

Sivamani et al. ([2021\)](#page-32-11) used feed-forward backpropagation neural network (FFBPNN)-BBD modeling to examine the applicability of orange zest biochar to remediate Cu(II) ions from its aqueous systems. The optimum uptake capacity of 94.76 mg/g was attained at Cu(II) initial concentration (100.00 mg/L), temperature (38.00 \degree C), and adsorbent dose (1.93 g/L), while 99.61% Cu(II) removal was realized at the optimal condition. The Langmuir and pseudofrst-order models represent the obtained data better. In the study, the thermodynamics, recovery, and recyclability of the used adsorbent were not tested, but the orange zest biochar was suggested as one of the possible adsorbents for removing Cu(II) from its aqueous phase.

The biosorption efficiency of deoiled carob seeds was evaluated for Cd(II) and Co(II) ions depollution from aqueous media (Farnane et al., [2018\)](#page-28-16). Using the full-factorial experimental design (FFED) of RSM, some parameters such as solution pH, biosorbent dose, and baseline metal ions concentration were chosen for process optimization. The best condition for high biosorption of Cd(II) and Co(II) was solution pH of 6.00, 1.00 g/L biosorbent dosage, and initial metal ions concentration of 50.00 mg/L, with adsorption capacities of 85.73 and 51.90 mg/g for Cd(II) and Co(II), respectively. The biosorption isotherm and kinetics were well ftted by the Langmuir and pseudo-frstorder model. Besides, the thermodynamics, regeneration, and reusability of impregnated biosorbent were not investigated.

The depollution of La(III) from wastewater was evaluated by cellulose chemically treated with sodiumglycerophosphate (Gabor et al., [2017](#page-28-17)). The best experimental condition obtained following the factorial design indicates that solution pH of 6.00, contact time of 60.00 min, and temperature of 298.00 K, with an equilibrium concentration of La(III) of 250.00 mg/L and a maximum uptake capacity of 31.58 mg/g. Further analysis by RSM during the optimization process reveals that the optimum adsorption density ranged between 30.87 and 36.73 mg/g at a solution pH of 6.00 and 256.00 mg/L La(III) initial concentration. Again, in this study, the equilibrium isotherms, kinetics, thermodynamics, desorption, and reuse of the adsorbent were not considered.

The capability of bentonite to eliminate Ni(II) from aqueous systems was investigated to determine the best condition, equilibrium, and kinetic models as well as thermodynamic factors (Sadeghalvad et al., [2017\)](#page-31-15). The adsorbent demonstrates its ability to adsorb Ni(II), with the optimum removal (87.70%) of Ni(II) attained at the optimum condition of pH of 6.00, 7.00 g/L dosage of adsorbent, Ni(II) concentration of 100.00 mg/L, a temperature of 298.00 K, 150.00-mm particle size, 500.00-r/min stirring speed, and 30.00 min time of contact, by the factorial design results in the RSM. The Freundlich and Dubinin–Radushkevich isotherm models ftted best to the data attained. The pseudosecond-order model best follows the time-dependent data, while the spontaneous, associative, and exothermic processes are dominated in Ni(II) removal,

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A=2.00 mg/mL, and *T*=32.00 °C

For column bed

For column bed $T = 32.00 °C$

 $Cr(V1)=91.70%$ $Cu(II)=94.00%$

 $\begin{array}{l} \rm Cr(VI)=91.70\% \\ \rm Cu(II)=94.00\% \end{array}$

studies
BH = 20.00 cm and
FR = 300.00 mL/h $BH = 20.00$ cm and FR=300.00 mL/h

35 13×molecular sieves Co(II)

 $13 \times$ molecular sieves

35

 $CCD-RSM$ $H=20.00$ mg/L,

CCD-RSM

 $Co(II)=90.21%$ $Ni(II)=96.83%$

 $Co(II) = 90.21\%$
Ni(II) = 96.83%

-

 Yes (Wen $& Wu, 2012$)

 Yes

(Wen & Wu, 2012)

T=40.00 °C for both the metals; and pH=6.29 and 8.00 for Co(II) and Ni(II)

 $H = 20.00 \text{ mg/L},$
 $T = 40.00 \text{ °C}$ for

both the metals; and
pH = 6.29 and 8.00
for Co(II) and Ni(II)

and
Ni(II)

(Korrapati & Y, 2015)

(Khandanlou et al., 2015)

according to the values of Gibbs free energy, entropy, and enthalpy changes. The desorption and renewability of the exhausted adsorbent for further use were not reported.

The possibility of using neem bark powder in As(III) decontamination from wastewater has been examined (Roy et al., [2017\)](#page-31-21). Several biosorption tests were used to investigate the elimination of As(III) using batch and column operations. The operating conditions were optimized by an artifcial neural network (ANN) and CCD in RSM, respectively. The study shows that the adsorbent was efficient in removing about 89.96% As(III) ions from the wastewater under optimal batch conditions. When the infuencing parameters were kept at an adsorbent dose of 2.00 g/L, initial As(III) concentration of 2.00 mg/L, and 3.0 mL/min rate of fow, with 0.969 desirability coefficient, an optimum 653.90 min breakthrough time was accomplished. The obtained experimental data correlates well with the Langmuir isotherm and pseudo-second-order models. The enthalpy, Gibbs free energy, and entropy changes of adsorption onto the adsorbent indicate the process was endothermic, spontaneous, and improved with increasing disorderliness at the solid–liquid interface. However, the desorption and renewability of the exhausted adsorbent were not reported yet the study indicates that the adsorbent is inexpensive and economical for wastewater purifcation.

The modeling and optimization of independent parameters (contact time (CT), stirring speed (*S*), temperature (*T*), adsorbent dose (*A*), heavy metal initial dose (*H*), activation time (AT), impregnation ratio (IR), bulk density (*P*), carbon to sulfur ratio (CR), bed height (BH), and flow rate (FR) of some other heavy metals removal using diferent adsorbents) are summarized in Table [1](#page-20-0).

Conclusions

Pollutants from water systems must be eliminated to provide an appropriate supply of potable water for living organisms and domestic use. In terms of determining interactive behavior between the independent components during the adsorption process, traditional experimental techniques for heavy metal adsorption from wastewater have several limitations. It is still limited when it comes to optimizations that take into account the change of one or more parameters at the same time without using statistically designed experiments. As a result, response surface statistical modeling can be considered a useful and strong technique for optimizing heavy metal adsorption on diverse adsorbents. Furthermore, this review identifes some limitations in the literature that must be addressed.

- 1. There has been no study on the application of RSM to optimize desorption parameters for optimum metal ion retrieval on depleted biosorbents from mono-ion and multi-ion adsorption systems. The regeneration and reusability of the used adsorbent is a vital concern for the selection of a more costefective and applicable adsorbent for real water and wastewater treatment systems.
- 2. More importantly, little is known about employing RSM to optimize adsorption variables from binary and multi-ion adsorption systems.
- 3. The batch adsorption mode is used for almost all the optimization operations. Only a few studies used a fxed-bed column to study the continuous adsorption process.
- 4. Two experimental designs (CCD and BBD) are typically utilized in RSM among the several available. It is essential to look at the possibility of applying additional experimental designs from the design of experiments to efficiently optimize adsorption process variables.
- 5. Almost all of the investigations are being carried out in batch mode utilizing only metal-based synthetic wastewater. Real industrial effluents and sewages containing heavy metals must be evaluated, such as textile, dyeing, electroplating, and tanning effluents and sewages. As real wastewater consists of a complex combination of ions and chemicals rather than a single one. The presence of various ions may increase adsorption, act independently, or interact with one another and could change the adsorption mechanism.

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Declarations

Conflict of interest The authors declare no competing interests.

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