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Heavy Metal Removal Techniques Using Response Surface Methodology: Water/Wastewater Treatment

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Abstract

Advanced water/wastewater treatment techniques including ion exchange separation, filtration separation, and adsorption are essential in the removal of non-biodegradable toxic wastes from water. In the current study, removal of heavy metal ions from water/wastewater and the use of response surface methodology (RSM) for experimental optimization were examined thoroughly. The objective of this work was to summarize the removal of heavy metal ions from water/wastewater using various chemical techniques and to emphasize the superiority of RSM in these studies.

Keywords: response surface methodology, water, chemical techniques, toxic wastes, optimization

1. Introduction

Due to the vital role of water for humanity, it is necessary to improve and maintain its quality. Environmental and global changes especially industrial wastes and domestic and agricultural activities are the main water pollution source. Worldwide, several water resources even underground water resources are contaminated, and they are not a suitable quality for drinking. Because of the rising living standards, growing world population, unconscious water consumption, and urbanization lead to increasing water supply costs. In most cases, as it contains different and large number of pollutants, wastewater lead to ecosystem hazards for being released around without being processed. So a few decades later, the world could face a major problem with freshwater supply [1]. In the past, very little financial resources have been allocated for wastewater because water supply received more priority than wastewater treatment (WWT). But, because of the increasing rapid population growth and trends in urbanization, WWT plays an important role in human life. Recently, because of the impact of sewage contamination of groundwater, rivers, and lakes, the growing awareness of wastewater treatment is now receiving greater attention from researchers and environmentalists. Research study results revealed that WWT, which is managed appropriately, has a large share in the growing economy when water resources treatment and supply are done in an appropriate manner [2, 3]. Safe, reliable, and sustainable treated WWT strategies have a vital role because of several

challenges including adoption of low-cost WWT technologies. To prevent the spread of diseases, WWT systems are crucial, and they should have high levels of hygienic standards for reuse in agricultural and other areas. Lack of WWT can lead to environmental pollution, and it may cause a hazardous effect for the health of humans. To improve global health and to prevent spread of disease, reliable collection and treatment of wastewater are very important. Wastewater treatment and their reuse need innovative and appropriate technologies. Recently, WWT technologies including electrochemical technologies have regained their importance worldwide. In some cases, the electrochemical mechanism for metal recovery is very simple. These technologies have reached comparably with other technologies in terms of cost and efficiency [4]. Economic issues besides environmental and social aspects must be considered when choosing the most appropriate WWT method [5, 6]. All scientists and environmentalists desire widespread recognition of the need to implement more sustainable WWT techniques. Wastewater treatment technologies follow two main approaches: first is the development of a single indicator integrating different criteria and second is the development of a set of multidisciplinary indicators [7, 8]. When large volumes of treated wastewater contain low concentrations of chemical constituent discharge-receiving water body, it may still lead to water quality problem. Discharges from industrial activities have been identified as one of the major sources of aquatic pollution in industrialized countries. After 1990, to remove toxic pollutants in wastewater, scientists focused on persistent organic pollutants including PCBs, PAHs, and especially heavy metals due to destructive effects [9, 10]. People's anxieties also increase because of pollutions caused by heavy metals. Pollutions caused by heavy metals spread into the aqueous systems from many industries such as metal plating and smelters, eluents from plastics, mining, and textile industries [11]. Toxic heavy metals including mercury and chromium are discharged to the environment, and unfortunately they cannot biodegrade in nature [12, 13]. Heavy metals can be traveled through the food chain via bioaccumulation, the increase of heavy metals in human body causes some major diseases like brain, pancreas, and heart diseases, and they can lead to wide spread capillary damage and gastrointestinal irritation besides possibly necrotic changes in some tissue [14]. Even at low concentrations, heavy metals can cause serious toxic and harmful effects on the organism and the environment. The World Health Organization (WHO) limited heavy metal concentrations. Such as in drinking water, maximum acceptable limit of copper concentration is offered as 1.5 mg L^{-1} , when the limit concentrations of metals containing hazardous waste are different [15, 16]. Ion exchange, extraction, membrane filtration, and chemical precipitation especially adsorption techniques have been applied to remove heavy metals; on the other hand, generally adsorption technique is one of the most chosen method because of its simplicity, nontoxicity, cost-effectiveness, and local availability to remove toxic heavy metals from aqueous medium [12, 17–19]. In addition, heavy metal removal from different samples by natural adsorbents using adsorption is in the most appropriate technique, and the use of natural adsorbents has been the preferred choice for many researchers [20, 21]. In large number of studies, activated carbon, carbon nanotubes, clays, nanosized metal oxides, zeolites, and various biosorbents were used. However, statistical and optimization research using RSM with CCD or Box-Behnken design about heavy metal removal under various physicochemical parameters is restricted and very rare. Although numerous studies are in literature about heavy metal removal sorption using different materials, there are very little studies with the application of WWT using methodological approach. Classical and conventional methods cannot depict all factor combinations, which affect the experiment. At the same time, these methods take a lot of time to experiment for

the determination of the optimum levels. Limitations can be eliminated using a statistical experimental design, which is optimizing all the effecting parameters collectively. In order for modeling of process parameters, RSM that contains a small number of experiments is widely used in various processes especially in adsorption [22]. Experimental design technique is a suitable tool for developing, improving, and optimizing process and multifactor experiments. It researches the common relationship between various factors for the most favorable conditions of the process, which helps to determine the interactions among optimized parameters [22, 23]. The primary target of RSM is to detect the optimum operational conditions for the system or to detect a region that compensates the operating specifications. The aim of this study was to present heavy metal removal from wastewater using RSM as a statistical technique. After discussion of wastewater treatment techniques as detail, several heavy metal removal methods from industrial wastewater will be presented.

2. The aim of wastewater treatment

There are two aims of wastewater treatment: firstly to purify wastewater without harming the public health and/or causing other nuisance and secondly to gain energy, nutrients, water, and other valuable resources from wastewater during purification steps.

3. Wastewater composition

Contaminated waters contain (**Figure 1**) various pollutants such as nutrients, various chemical compounds, and numerous pathogenic microorganisms besides toxic compounds. Inorganic solids, organic solids, and pathogenic microorganisms along with metals constitute a significant part of wastewater. While inorganic solids include salt, sediment, soil, and especially metals, organic solids contain food wastes, paper, and another household waste material. During WWT step, the removal of primarily organic particles especially suspended solids is vital prior to discharge to the environment. The proteins, lipids and carbohydrates are biodegradable components of wastewater. Biodegradable components contain carbon, and they can be converted to carbon dioxide. If these biodegradable organics are not removed from the wastewater, oxygen demand will exert

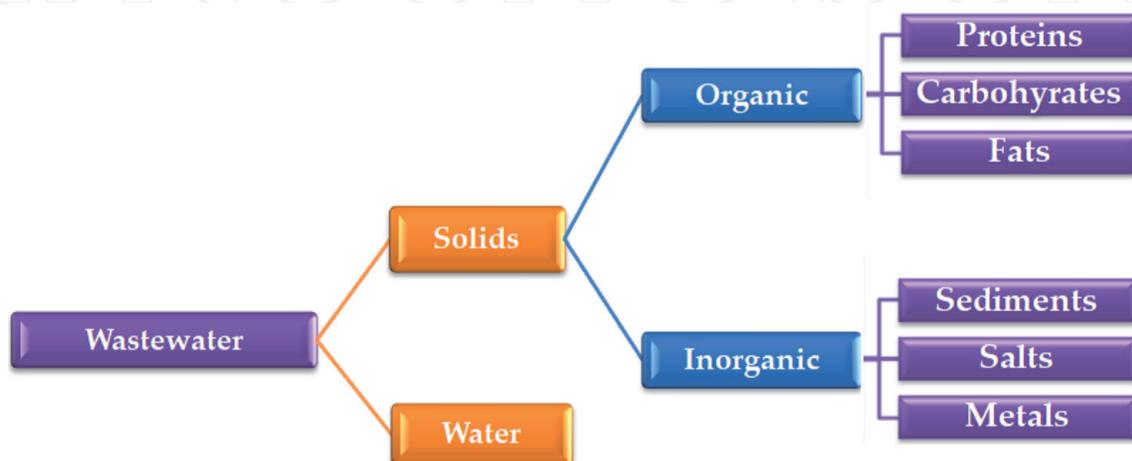


Figure 1.
Typical wastewater composition.

in the receiving watercourse. Biochemical oxygen demand (BOD) or chemical oxygen demand (COD) is typical measures of organic matter. BOD is the most widely used parameter to quantify organic pollution of water. BOD is the measurement of the dissolved oxygen that is used by microbes in the chemical oxidation of organic matter.

4. Water pollutants

It is important to understand the nature of water pollutants because wastewaters contain a large number of pollutants; however, toxicity is observed when the acceptable limits are exceeded. Wastewater contents depend on industrial, agricultural, and municipal wastewater. There are various water pollutants in nature, and they can be categorized as microbiological, radioactive, particulate, organic, and inorganic chemical contaminants. Harmful microbes such as viruses, fungi, bacteria, algae, plankton, and other microorganisms are basic components of bio-pollution in the water. These microorganisms may be responsible for various diseases. Organic toxic pollutants include many insecticides such as dichlorodiphenyltrichloroethane, herbicides, and other pollutants were manufactured for use in various industries. However, heavy metals are the most common inorganic water pollutants. Microbiological, radioactive, particulate, organic, and inorganic chemical water contaminants remain either in suspended, colloidal, or in solvated form.

5. Wastewater treatment methods

Because of the increasing population and rapid pollution of water resources, WWT and reuse are an important issue. The efficient use of existing water resources and treatment of polluted water resources with affordable and cheap technologies have been the focus of scientists. WWTs are needed for three reasons; these are water source reduction, WWT, and recycling. Recently, during purification step, while primary treatment includes preliminary physical and chemical purification processes, secondary treatment depends on biochemical decomposition of organic solids to inorganic or stable organic solids. Finally, after the third step called tertiary treatment processes, wastewater is converted into good-quality water, and it can be used for drinking or medicinal supplies. At the end of this step, almost all of the pollutants (up to 99%) can be removed from water. To producing good-quality and safe water, all these three processes should be combined together. Otherwise, it will not be possible to obtain safe water from the wastewater. Many advanced methods and techniques have been used for the recycle of safe water from wastewater, but economic and effective water treatment is still a serious problem. Treatment of wastewater and recycling technologies have been classified (**Figure 2**), and it is carried out in three stages. They are:

- Primary treatment methods
- Secondary treatment methods
- Tertiary treatment methods

These methods are briefly described below.

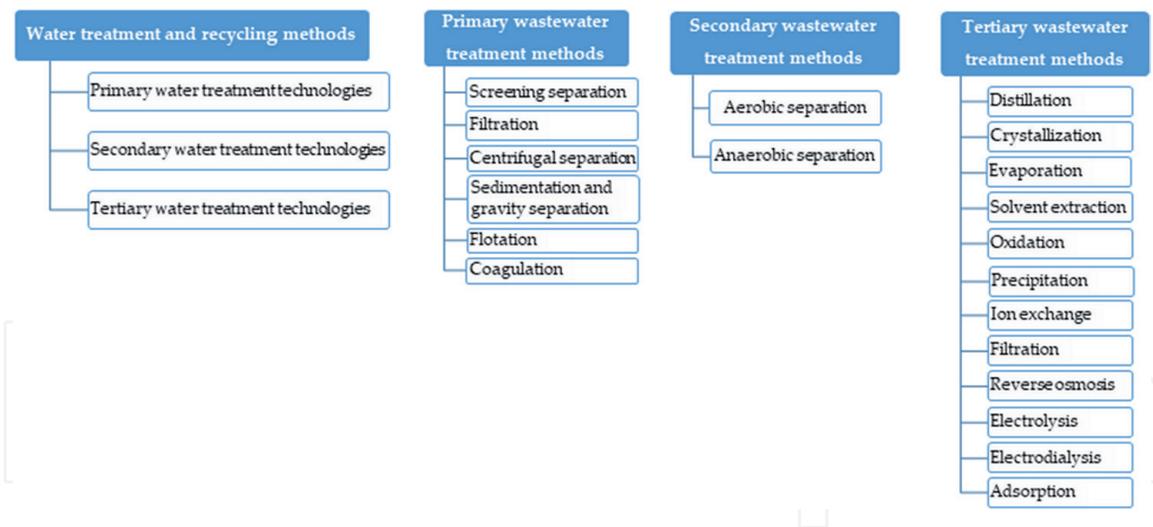


Figure 2.
 Wastewater treatment and recycling methods.

5.1 Primary treatment methods

In order to remove organic matter and suspended solids from wastewater by means of physical operations, for example, sedimentation and gravity separation, they are done in primary treatment stage. Preliminary treatment, which is described as preparation for secondary treatment, is in fact intended to produce a liquid waste suitable for biological treatment.

5.1.1 Screening separation method

Screening separation method is used to remove solid wastes from wastewater. It is the process where suspended and floating materials including wood, paper, kitchen refuse, pieces of cloth, cork, hair, fibers, and fecal solids are removed from wastewater. In a WWT, screening is generally used as the first operation step. For this purpose, various size screens are used, and their size is selected as per the requirement. Finer particles such as sand and small pebbles can be eliminated by using screening separation method.

5.1.2 Filtration method

About 0.1–0.5 mm pore size is used in filtration separation method, water is passed through a medium having fine pores, and the filtration process is completed. Various membranes and filters, for example, cartridges, can frequently be used to remove suspended solids, greases, oils, and bacteria from the wastewater. The main purpose of filtration separation method is to separate the small solids and remove oil (they can be reduced up to 99%). Filtered water is used for many purposes such as ion exchange, adsorption, or membrane separation processes. In pharmaceutical and biotechnological industries, to the production of pure water, filtration separation method has become the main focus as promising separation tool for WWT. The used membrane has a key role due to selectivity, low fouling, and performance stability for long-term operation in the filtration separation method. Because of these advantages, this method and its performance are becoming more and more important. In addition, it is one of the important enrichment techniques for trace heavy metal ions along with simplicity and rapidity of the procedure. For all these reasons, many scientists have focused on this subject to develop and use alternative and effective membranes [24, 25].

5.1.3 Centrifugal separation method

This method is provided for separating components of a fluid or solid particles, but it is used especially for suspend solid from wastewater. Various types of centrifugal machines have been used to remove suspended noncolloidal solids in the centrifugal separation method. To separate solids from wastewater, centrifugal devices with various sizes are used. Density of suspended solids is the most important parameter when separating solid materials by centrifugation. In addition, oils and greases can be reduced and separated during application of centrifugal separation method.

5.1.4 Sedimentation and gravity separation method

Sedimentation and gravity separation method are based on the removal of suspended solids, grits, and silts from aqueous media. Suspended solid materials settle down to the bottom of the tank under the influence of gravity; this event may vary depending on solid size and density. Some chemicals can sometimes be added to accelerate sedimentation process. Although this method can reduce suspended solids only up to 60%, purification of wastes is a very useful separation application. Water treatment in this technique can be used in many areas such as water for membrane filtration processes and ion exchange method. It is generally applied out prior to conventional treatment.

5.1.5 Coagulation method

Coagulation processes are a particularly effective cleaning method for containing oil-in-water emulsions such as sea, lakes, and rivers besides most industrial wastes contain especially oil or petroleum. After sedimentation and gravity separation method, if there are non-settleable solids in wastewater, this is called processing coagulation with the addition of certain chemicals to precipitate these non-settleable solids and non-precipitating deposits. There are some natural coagulants such as aluminum salts, iron materials, alum, starch, and activated silica and also some polymers that can be used as coagulants. In this process, the most important controlling factors are contact time, temperature, and pH. In addition, during biological treatment processes, to remove microbes and any organics in the water, some certain coagulants can be added. Coagulation processes play an important role in recycling and removing pollutants from wastewater.

5.1.6 Flotation method

In order to remove suspended solid including oils, greases, biological solids, and other solids from wastewater, flotation separation method is used. In these processes, suspended solids are removed by adhering them with either air or gas. Various chemicals like alum and activated silica are used to successfully apply the flotation process to wastewater because they help flotation separation method. For paper and refinery industries, flotation separation method is an effective method for WWT because suspended solids that oil and grease is can easily be removed (up to 75–99%) by these processes. Recently, to separate mixed plastic is too difficult using gravity separation; therefore, for WWT and recycling purposes, plastic flotation method has been used as effectively [26].

5.2 Secondary treatment methods

Secondary treatment techniques have been used to remove soluble and insoluble pollutants from wastewater as biological. The main objective of this process is to convert the organic and inorganic solids into fluorinated residues that are finely divided and dissolved in the wastewater and to remove of soluble and colloidal organics and suspended solids besides reducing BOD and COD through biological process. When water has a high microbe concentration like bacterial and fungal strains, secondary treatment techniques should be selected for treatment because organic matter is converted into other products via these microbes; besides, they detoxify toxic inorganic matter. After this process is applied to wastewater, toxic organic and inorganic substances can be removed [27].

5.2.1 Aerobic separation method

In biological treatment processes, organic matter can be biodegradable by aerobic and facultative bacteria. Aerobic processes depend on temperature, the oxygen amount and availability of oxygen, and the biological activities of the bacteria. If bacterial growth is accelerated by adding some chemicals to the medium, the organic pollutant oxidation rate as biological will also be increased. Aerobic treatment techniques are the most effective method for removing suspended, volatile, and dissolved organics, nitrates, and phosphates besides BOD and COD. Because of the production of a huge amount of biosolids, aerobic treatment techniques have a big disadvantage; however, the biodegradable organic amount can be reduced substantially (up to 90%) using this method.

5.2.2 Anaerobic separation method

Anaerobic decomposition, called putrefaction, occurs when free dissolved oxygen is not present in wastewater, and this process is called as anaerobic treatment technique. In this treatment technique, organic matters convert into other organics including sulfur and carbon by anaerobic and facultative bacteria. There are two metabolic phases named acidogenic phase and methanogenic phase in the anaerobic separation technique. Some gases such as methane, hydrogen sulfide, ammonia, and nitrogen can be released. To reduce the biological load of wastewater, this method is very vital [1].

5.3 Tertiary treatment methods

For the production of safe water that people can consume, tertiary water treatment techniques are very important, and they should be applied to wastewater. In this last step, wastewater is subjected to final treatment using some vital techniques, and they are briefly summarized below.

5.3.1 Distillation method

The distillation method is based on the principle that the water is evaporated to the boiling point and the steam is distilled by cooling. After this process, purified water can be obtained free from impurities up to 99% in addition to wastewater is also freed from the volatile pollution. The obtained water by the distillation method is usable in levels of laboratory applications and medicinal preparations. In addition, to prepare potable water from the sea, distillation separation method is an effective tool.

5.3.2 Crystallization method

The crystallization method, which is based on the increasing principle of the concentrations of pollutants up to the crystallization point, is an effective method for obtaining quality water. Crystallization technique is useful to remove high concentrations of total dissolved solids including soluble organics and inorganics from wastewater, and it can be created either by mixing some solvents or by evaporation. This process is generally used for wastewater released to the environment from paper and dyeing industries. In addition, crystallization can be used for pH control because of other constituents including sulfite bicarbonate [1].

5.3.3 Evaporation method

When compared to other techniques, evaporation separation method is a natural process and suitable method but only for small wastewater volumes due to its high-energy consumption. However, this technique has some problems such as pollution, calcification, and foaming that have occurred in the presence of suspended solids and carbonates in the wastewater. Thus, to increase the evaporation rate and to reduce energy consumption, vacuum evaporation step can be used. Under natural conditions, water surface molecules escape from the surface, and they generally collect pure water. Recently, to recycle water process, mechanical evaporators and sometimes vacuum evaporation have also been used. Using evaporation separation technique is effective for the removal of pollutants including organic and inorganic compounds, but some volatile organic compounds may recirculate into the water during the evaporation phase. Evaporation treatment technique is applied to various industry wastewaters like pharmaceutical, petroleum, and fertilizer industries. The obtained water from evaporation treatment technique has been used for different purposes including cooling in towers and boilers [28].

5.3.4 Solvent extraction method

Solvent extraction separation method is an important tool to dissolve pollutants from wastewater using various organic solvents like phosphoric acid. Acetone, methanol, hexane, ethanol, and acetonitrile are the most commonly used organic solvents. In this technique, some organic solvents are added to the wastewater to facilitate contaminant removal. The technique is very effective to remove oils, greases, and various organics. However, the process is often used for extraction and separation of heavy metals like lead, cobalt, and chromium using extraction and separation techniques from various industrial wastewater and effluents [29].

5.3.5 Oxidation and advanced oxidation method

To remove various toxic and hazardous chemicals especially endocrine-disrupting chemicals from wastewater, chemical oxidation techniques are preferred, and it is a promising technology for the treatment of wastewaters containing pharmaceutical products. Organic compounds that are oxidized by oxidation of readily degradable species such as alcohols and carboxylic acids are the main components of this process [30]. Ozone, hydrogen peroxide, and Fenton's reagent are commonly used as chemical oxidation reagent. The chemical oxidation rate depends on some variables such as the presence of catalyst, temperature, and pH. Also, pollutants and nature of oxidants identify the rate of chemical oxidation. Various organic pollutants including hydrocarbons, dyes, and phenols can be removed from wastewater using chemical oxidation treatment technique. Recently, there has been

a continuously increasing worldwide concern for the development of alternative wastewater reuse and recycling methods. Single oxidation separation method can sometimes be inadequate for the total decomposition of organic contaminants in wastewater. This requires advanced oxidation processes, which involve the use of more than one oxidation process at the same time [31]. Summarize, advanced oxidation process has big advantage because in this process all organic contaminants can be commonly oxidized to carbon dioxide form.

5.3.6 Precipitation method

The precipitation method based on the principle that the solubility of the contaminants is reduced and the precipitates which are converted into the solid form are easily separated from the water surface is an effective method for removing metal ions and various organic contaminants from wastewater. Chemical precipitation is a physicochemical process and a very flexible approach to various pollutant removals and can be applied at several stages during wastewater treatment. In industrial applications, precipitation has been the most common technology for metals [32]. In this process, to reduce solubility of the dissolved pollutants, it can be carried out either by lowering the temperature of the water or by adding some chemicals like sodium bicarbonates and ferric chloride, but chemical addition is not preferred because it increases the cost. Common applications of precipitation separation method are wastewater treatment from chromium and nickel plating industries and water recycling besides water softening and removal phosphate from water.

5.3.7 Ion exchange method

Ion exchange technique provides advantages due to it being technologically simple and enables efficient removal of even traces of impurities from solutions, high treatment capacity, high-removal efficiency, and fast kinetics when compared other usual methods. It can be applicable to various industrial wastewaters to remove hazardous materials. Ion exchange treatment technique depends on toxic or undesirable ions, which are replaced with others ions. There are two types of ion exchangers, which can be classified as cation and anion exchangers. Ion exchangers are natural or synthetic resins with active sites on their surface. Synthetic resins are widely preferred because of their effectiveness in removing heavy metals from wastewater [33]. In order to remove hazardous ions from wastewater, some resins including zeolites, sodium silicates, and acrylic and metha-acrylic resins are used as the most common. Reversible process and low-energy requirements are the most important advantages of this method. Using this method, organic and inorganic pollutants can be reduced about by 95%, but pretreatment may be needed if the wastewater contains oil or grease.

5.3.8 Filtration method

Recently, from the industrial sources, a large amount of oily wastewaters has been generated. The most serious pollutants are oil-in-water emulsions because of treatment cost and ineffective of using treatment methods [34]. Using micro-filtration, a suspended solid pollutant that is a particle size from 0.04 to 1 mm can be removed. Microfiltration separation technique has been widely used to remove macromolecules, emulsion droplets, suspended particles, and microorganisms from various industrial fields including food, pharmaceutical, biotechnological, and petrochemical. In the last decade, membrane separations have been developed

using various organic/inorganic membranes like ceramic membranes. It is becoming a promising technology for industrial processes and is utilized currently for oil field-produced WWT. When compared to traditional treatment methods, they have some advantages including high oil removal efficiency, low-energy cost, and compact design. Perhaps the most important advantage is that it does not require any chemicals. Some materials such as cellulose, fiberglass, and cotton can be used as filters in filtration method. Recently, several researchers focused on the new inorganic membrane development, for example, natural mineral-based ceramic membranes, carbon membrane, and zeolite membrane [35].

5.3.9 Reverse osmosis method

As membrane technology has been developed, membrane filtration mechanism became a feasible option for wastewaters. Reverse osmosis treatment technique that is called as hyperfiltration is the wastewater purification system that relies on the membranes' development technology. Using membrane filtration mechanism has shown results of very high efficiency in the filtration of wastewater. According to various studies from literature, when it is used, removal percentage has been achieved as at least 99.9% for COD, total organic carbon, suspended solids, coliforms, and pathogens. To achieve the required filtration, various membranes including cellulose, polyether, and polyamide are used in this process. In this process, the most important parameter is free energy, and other considerable parameters can be identified as pressure, pH, and operation time. To remove the soluble pollutants which contain macro- and microlevel nonpolar, ionic and toxic materials from the wastewater reverse osmosis is a very suitable separation technique. Reverse osmosis treatment technique is the most economic process because the water obtained from this process is of ultrapure water. It can be used in pharmacy and medicines because it can remove various microbes, bacteria, and viruses at high percentages (up to 99.99%) when compared other techniques [36].

5.3.10 Electrolysis method

Electrolysis method based on the redox reaction principle can be expressed as the separation and deposition of the dissolving materials on an electrode surface. During electrolysis separation method, metal ions are deposited on the electrode and separated from the wastewater. In the last decade, electrochemical oxidation methods have been an increasing interest because they can be applicable to WWT. In this process, various electrodes and anodes such as iron electrode, boron-doped diamond electrode, PbO_2 electrode, and graphite electrode [37, 38] have been used to remove different pollutants from wastewater.

5.3.11 Electrodialysis method

To remove various ions and other pollutants which have serious impact on the environment from wastewater, several methods have been used. Electrodialysis technique may be one of the most effective methods among these techniques because of recent progress in membrane technology. Electrodialysis, which is a membrane separation technology, depends on an electric potential difference, which is used to drive ion migration toward oppositely charged electrodes. In this process, under the influence of electric current, water-soluble ions pass through the membranes that are made of ion exchange material [39]. Certain factors, for example, nature of pollutants, applied current amount, temperature, and pH, must

be kept in mind to remove dissolved solids. This method has been used to produce potable water from brackish water and for water source reduction [40].

5.3.12 Adsorption method

Adsorption separation method is an attractive process because it can be easily applied to WWT, which includes efficiency and flexibility. When it is compared with other treatment methods, it appears superior than others. Some factors that affect adsorption efficiency including the type of adsorbents, pollutant concentration, adsorbent particle size, pH, contact time, and temperature are very important for this process. A pretreatment may be needed to successfully apply the adsorption technique to wastewater because of the presence of suspended particles and oils. To remove pollutants especially heavy metals from wastewater, various adsorbents such as activated carbons from different materials [41, 42], Astragalus [19], carbon nanotubes [43], and a large number of biosorbents [44] have been used by different studies in the literature. However, novel and effective adsorbents with local availability besides economic suitability are still needed. Adsorption technique has two main problems: the first is the regeneration of columns and column life used as an adsorbent and the second is the management of the exhausted adsorbent.

6. Heavy metal removal from industrial wastewater using response surface methodological approach

Nowadays, because of rapid technological development especially in developing countries, environmental pollution is a serious problem for the ecosystem because wastewaters contaminated with toxic heavy metals are discharged directly or indirectly into the environment. Unlike most organic contaminants, heavy metals including As, Hg, and Cr are hazardous due to its nonbiodegradable nature [33, 45]. Thus, to protect the people and the environment, these hazardous ions should be removed from wastewater [46]. For example, while industrial wastewaters which contain Cr ions range from 0.5 to 270 mg L⁻¹, inland surface water tolerance limits 0.1 mg L⁻¹, and potable water Cr level should not exceed 0.05 mg L⁻¹ according to various health organization such as the WHO and EPA [47, 48]. To remove heavy metal ions from wastewater, many conventional techniques such as membrane filtration, reverse osmosis, ion exchange, chemical precipitation, electrodialysis, electrochemical treatment, and adsorption have been employed. While most of these methods suffer from operational costs for the treatment process and high capital, the adsorption method is better than the other methods due to its flexibility in design, simplicity of operation, and facile handling, and it is considered more efficient and economical [45, 49]. Since the dynamic characteristics of the adsorption process are complex, it is essential to have optimum working conditions in order to achieve optimum pollution removal efficiency. Process optimization is crucial to determine design parameters value, which is achieving the optimal obtained response level. The RSM is one of the most used methods because of its developing, improving, and optimizing of the processes especially in the presence of complex interactions. It is also used to determine the ideal points of independent variables that are effective under optimum conditions and to evaluate the interactions of these variables [50]. Its greatest advantage is the decreased experimental trial number required to interpret multiple parameters. Therefore, RSM optimization process contains three main steps: (a) appropriate experimental design selection, (b) model coefficient estimation using analysis of variance (ANOVA), and (c) model validation based on prediction and experimental runs of the process

response validation of the final model [51]. This experimental design method for an adsorption process is more practical than other approaches because it allows for the opportunity to monitor and interpret interactions between variables and to describe the overall effect of the parameters on the process. The RSM has been successfully used; in addition, its greatest applications have been in industrial research [52].

There are numerous studies, and different results were obtained using various adsorbents reported such as by Anupama et al. [53]. They used a CCD with RSM for removing Cr(VI) from aqueous medium [53]. They investigated the effect of some parameters including pH and temperature on adsorption, and the optimum pH, time, and adsorbent dose were found to be 2.32, 25.76 min, and 1.79 g L^{-1} . Also various adsorption kinetic models and isotherms were compared to find fit model. Jain et al. [54] studied Cr(VI) removal from aqueous solution using Box-Behnken model with combined RSM approach by chemically treated *Helianthus annuus* flowers. They investigated three effective factors for Cr(VI) removal. It was reported that the optimum pH, adsorbent dose, and initial concentration of Cr(VI) were found to be 2.0, 5.0 g L^{-1} , and 40 mg L^{-1} , respectively [54]. In another study [55], Box-Behnken design has been applied to evaluate operating variables interaction for Cr (VI), Ni (II), and Zn (II) ions adsorption on *Bacillus brevis*. They carried out a total of 17 experiments and used a quadratic model. Based on this model, it was reported that the regression equation coefficients were calculated, and the data fitted to a second-order polynomial equation for these metal ions removal with immobilized on *B. brevis*. According to another study, to evaluate and optimize Cr ions, adsorption on activated carbon experimental conditions using RSM as an efficient approach for predictive model building was performed by Sahu et al. [56]. A full factorial CCD was employed, and based on ANOVA, a high coefficient ($R^2 = 0.928$) was obtained. In addition, satisfactory prediction of second-order regression model was derived. According to optimized process parameters, Cr(VI) removal percentage was obtained higher than 89% [56]. Kaplan Ince et al. [57] studied a batch experimental system for removal Pb(II) using clay, and optimized experimental approach was applied to some alcoholic beverages including beer and wine samples. Various effective parameters were investigated using a Box-Behnken experimental design methodology and RSM. They reported that the optimal conditions used for Pb(II) removal were pH of 5, contact time of 31 minutes, 75 mg for adsorbent dosage, and 100 rpm for agitation speed. Based on these results, maximum Pb(II) ion removal was calculated as 120 mg g^{-1} from aqueous medium using an ETAAS [57]. Balan et al. (2009) examined the efficiency of Cd(II) removal from aqueous solutions using sphagnum moss peat as biosorbent. They carried out a CCD for experimental design to evaluate an analysis of results and to optimize process parameters including the pH of solution, biosorbent dosage, and Cd(II) initial concentration. The optimum values of experimental parameters were obtained as 4.72 for pH, 14.7 g L^{-1} for biosorbent amount, and $13.64 \text{ mg Cd L}^{-1}$ for initial concentration of Cd(II) [58]. In another study, removal of Cr(VI) from simulated wastewater using RSM was examined by Bhatti et al. [59]. They investigated the performance of a laboratory scale electrocoagulation system for the removal of Cr(VI) using Al-Al electrodes. They obtained an interaction between voltage \times time and amperage \times time coefficient of determination as 0.8873 and 0.9270, respectively. For the optimization of process variables including pH, voltage, and treatment time, the RSM was used. Prediction model results were validated through laboratory scale batch experiments [59]. In another similar study, to remove arsenic from contaminated water by arsenite, an electrocoagulation method with stainless steel electrode was used. A response surface methodology approach was performed to optimize significant process variables such as treatment time and solution pH. They obtained

pH as 5.2, treatment time $\frac{1}{4}$ 20 min for 10, and 55–100 mg L⁻¹ of initial arsenic concentration. It was stated that the waste elimination with electrocoagulation is a sustainable treatment technology with quick start-up, shorter treatment time, and minimum sludge generation [60]. An alginate-coated chitosan nanoparticle was carried out for heavy metal removal from industrial effluents by Esmaeili and Khoshnevisan [61]. To optimize the process of biomass for heavy metal removal from synthetic and industrial effluents containing nickel, an RSM approach was performed. Under optimum experimental conditions, which they obtained as a dose of 0.3 g biomass, pH of 3, 70 mg L⁻¹ of initial concentration nickel, and 30 min contact time, maximum removal efficiency of biomass was found as 94.48% [61]. The Cd removal from wastewater and simulated aqueous solution was examined by Iqbal et al. [62] using a polyurethane material as adsorbent. The effect of operating parameters including adsorbent dosage, pH of solution, and metal ion concentration was modeled by RSM combined with CCD. Experimental runs and independent variables optimum values for Cd adsorption were obtained as 305 mg L⁻¹ Cd ion initial concentration, pH 4.9, contact time 932 min, and adsorbent dose 1.3 g for polyurethane material. Based on the experimental results, to predict the response with good accuracy and reliability, it was mentioned that the RSM proved to be the best statistical model [62]. Ince and Kaplan Ince [63] examined the removal of Cr from industrial wastewater using RSM combined with CCD besides investigated as an efficient approach for examining predictive model building and optimization. To predictive regression models and optimize experimental variables, statistical design was modeled. The experimental parameters such as pH and agitation speed were selected for optimization. They obtained ideal Cr ion removal conditions as pH of 5.0, contact time 23.0 minutes, adsorbent dosage of 69.4 mg, and agitation speed of 135 rpm. The Cr removal efficiency was found at 23.16 mg g⁻¹. Also, significant independent parameters and their interactions were verified by means of the ANOVA. The proposed adsorption process was applied to various industrial wastewaters. It was stated that a CCD method was identified to yield a maximum Cr ion removal of 99% [63].

7. Conclusions

The choice of method to be used in the treatment of water/wastewater depends on the wastewater type and its composition besides the economic aspect. For example, high-grade contaminated water containing solid waste and poor color must be subjected to tertiary water treatment after primary and secondary water treatment processes. If the water does not contain any solids and is contaminated by other contaminants including inorganic, organic, and biological pollutants, the application of the tertiary treatment technique is sufficient. While surface waters are often polluted by organic, inorganic, and biologic pollutants, secondary and tertiary methods of treatment are needed in the treatment of these waters, and only tertiary methods of treatment should be used since groundwater is exposed to hazardous metal ions and anion pollution. The present study summarized removing heavy metal ions in various industrial wastewaters exposed to heavy metal pollution and was focused on optimizing the removal method and determining optimum experimental conditions.

Conflict of interest

The authors declare that they have no conflicts of interest in the research.

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