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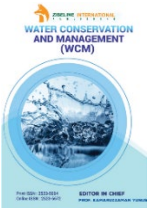

RESEARCH ARTICLE

BUCKTHORN LEAVES AS A GREEN ADSORBENT FOR REMEDIATION OF WASTEWATER POLLUTED BY HEAVY METALS

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ABSTRACT

The current investigation aimed to study the suitability of buckthorn leaves – without any further treatment – as a readily available and inexpensive adsorbent for recovering cobalt cations from contaminated aqueous solutions using a batch-type adsorption unit under various operating agents. The source of buckthorn leaves used was from perennial trees in the city of Baghdad, and the fresh leaves were carefully collected before being washed, dried, ground, and prepared to be an adsorption medium. The results obtained indicated that buckthorn leaves were remarkably efficient in removing cobalt cations from contaminated aqueous media, reaching 72.45% of 45 ppm at 6, 400 rpm, 150 minutes, and 7 g for each of pH, agitation speed, contact time, temperature, and adsorbent dosage, respectively. Thus, agricultural waste has proven its ability to be an important source of useful materials instead of being disposed of as waste that requires effort and cost to remove.

KEYWORDS

Adsorption, remediation, divalent cobalt cations, buckthorn leaves, ZRL

1. INTRODUCTION

Environmental pollution is one of the most prominent contemporary global challenges. In its broadest sense, it is defined as any harmful and undesirable change to the physical, chemical, or biological properties of natural environmental elements, air, water, soil, and space, caused by the introduction of foreign substances or energies resulting from natural changes or direct or indirect human activities (Khaleel et al., 2022). These substances or energies are introduced in quantities or concentrations that exceed the capacity of ecosystems to mitigate, decompose, or absorb them without consequences, leading to damage to living organisms, ecosystems, public health, and natural resources (Hashem et al., 2021). Environmental pollution becomes more dangerous when it is cumulative or toxic in nature, as its pollutants are difficult to eliminate, and its effects persist for long periods of time, posing a multi-dimensional existential threat. Given the vital importance of water to all forms of life, water pollution is one of the most serious forms of environmental pollution affecting one of the most essential elements of life (Al-Ali et al., 2023). It refers to a state of qualitative deterioration in the physical, chemical, or biological properties of water bodies (rivers, lakes, oceans, seas, and groundwater) as a result of disposing of pollutants that impair their ability to be used in a natural or safe manner (Khaleel et al., 2022). This type of pollution comprises from multiple sources, including the discharge of untreated industrial wastewater, household waste, domestic sewage, agricultural waste laden with fertilizers and pesticides, leakage from landfills, as well as oil spills, mining activities, and others (Rajaa et al., 2023). Water pollution occurs when harmful substances, whether chemical, biological, or even physical (such as heat or solid waste), are introduced in concentrations that exceed the natural ability of the body to purify itself through natural processes, thus transforming water from a source of life into a carrier of diseases and toxins (Hashem et al., 2021). Water pollution has severe and multi-layered effects. On an environmental level, it leads to eutrophication phenomenon, which causes algal blooms, oxygen depletion, and the creation of dead zones devoid of marine life. In addition, it causes fish poisoning and death, disrupts food chains, and has a devastating impact on aquatic organisms and aquatic biodiversity. Water pollution also negatively impacts economic activities such as agriculture and fishing, harms fish stocks and tourism, reduces the productivity of agricultural lands irrigated with polluted water, and simultaneously reduces the availability of potable water at a time of increasing global demand, while also increasing the costs of water treatment for drinking and industrial purposes (Rajaa et al., 2023). As for human health, it poses a direct threat, causing the spread of waterborne diseases such as cholera and typhoid, as well as poisoning from chemicals, carcinogens, and other toxins, due to their accumulation in biological tissues. The danger of water pollution lies in the difficulty of treating it once it occurs, especially when it comes to non-biodegradable pollutants such as heavy metals. Water pollutants vary greatly depending on their sources and nature, and can be classified into three main categories: physical, biological, and chemical pollutants. Physical pollutants include turbidity, suspended solids, micro and macro plastic waste, thermal pollution, and radioactivity. Biological contaminants include disease-causing microorganisms (pathogens) such as bacteria (e.g., *Escherichia coli*, *Salmonella*, *Vibrio cholerae*), viruses (e.g., Hepatitis A Virus (HAV), Norovirus), and protozoan parasites (e.g., *Giardia*,

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Cryptosporidium). The last and broadest category is chemical pollutants, which include organic chemical pollutants such as petroleum hydrocarbons from oil spills, industrial solvents (such as methylene chloride), pesticides and herbicides (such as DDT and atrazine), halogenated organic compounds (such as polychlorinated biphenyls), detergents, personal care pharmaceuticals, and hormones. Inorganic chemical pollutants include dissolved salts, acids, alkalis, toxic industrial compounds, nitrate and phosphate ions, cyanide, and fluoride (Abdullah et al., 2023). The most important pollutants of this type are heavy metals such as mercury, lead, cadmium, arsenic, chromium, cobalt, iron, nickel, vanadium, and copper. This type is considered the most dangerous due to its high toxicity and the difficulty of its biological decomposition in the environment. The type and extent of pollution depend on the source, geographical location, and prevailing human activities. The severity of the impact of these pollutants varies according to their concentrations and the duration of exposure. However, even low concentrations can lead to serious long-term environmental and health effects. Cobalt is a transition element of high industrial and technological importance due to its superior magnetic properties, high resistance to heat and corrosion, and ability to form strong and stable superalloys. It is also used as a catalyst in various chemical and petrochemical industries, in the manufacture of plastics and paints (as a desiccant), and in the production of organic acids. Cobalt is also used in medical industries, such as the manufacture of some surgical instruments and radiation therapies, in addition to its vital role as an essential trace element in human and animal nutrition in small quantities. Its biological role as a key component of vitamin B12 (cobalamin), essential for red blood cell formation and nervous system function in humans and animals, cannot be overlooked, making it an important ingredient in nutritional supplements and animal feed. Currently, rechargeable batteries, especially lithium-ion batteries, are among the most popular uses of this element, particularly in the era of modern technologies and the shift towards clean energy. It is a key component in cathode electrodes such as $LiCoO_2$, providing high energy density and cyclic stability, which is vital for powering portable electronic devices such as smartphones, laptops, and electric vehicles that represent the future of sustainable transportation. This diversity of uses makes cobalt a strategic element of high economic value, but it also increases the likelihood of its release into the environment during its exploration, extraction, processing, and disposal. While essential for living organisms, it can become harmful when levels exceed safe limits; the difference between its vital role and its toxic effects lies in its concentration. Water pollution by cobalt occurs as a result of industrial waste discharge, mining activities, battery manufacturing, dye and alloy production, waste from the electronics and metal industries, improper disposal of used batteries, and surface runoff from lands where cobalt-containing fertilizers are used, where its compounds can reach and dissolve in water bodies. Cobalt is mostly detected in contaminated water as divalent cations. Although its toxicity is moderate compared to other heavy metals such as lead, arsenic, and mercury, chronic exposure or exposure to high concentrations can cause serious health effects, including respiratory disorders, heart problems, muscle damage, blood disorders, skin and lung irritations, and toxic effects on the liver and thyroid gland. The International Agency for Research on Cancer has classified some soluble cobalt compounds on a category Group 2B, as potentially carcinogenic to humans (iarc.who, 2025). Therefore, global health and environmental organizations have established maximum permissible concentrations to protect public health. The World Health Organization (WHO) and the Food and Agriculture Organization (FAO) recommend that the cobalt concentration in drinking water not exceed 50 μg/L, and 100 μg/L concentration recommended by the U.S. Environmental Protection Agency (EPA), which classifies cobalt as a minor pollutant, to prevent discoloration or unacceptable taste in the water (Toxicological Profile for Cobalt, 2024). Generally, the permissible concentration of cobalt in drinking water is usually between 0.05 and 0.1 mg/L, and exceeding these values is a serious indicator of water contamination. Currently, several conventional methods are used to treat water contaminated with heavy metals, including cobalt, depending on the nature and concentration of the pollutant. These methods involve applying a range of physical, chemical, and biological techniques aimed at removing heavy metals, converting them into less harmful forms, or reducing their concentration to a level close to safe. Methods include chemical precipitation and coagulation, ion exchange, membrane technology, electrochemical treatment, biological treatment, and adsorption. Adsorption is considered one of the most effective techniques for treating water contaminated with heavy metals. It relies on the transfer of pollutants from the liquid phase to the surface of an adsorbent material with a high surface area (Alsarayreh et al., 2025a). This method is characterized by its ease of operation and allows for the regeneration of highly efficient materials such as activated carbon, zeolite, alumina, and others (Khudair et al., 2024; Shadhan et al., 2024). The importance of adsorption technology lies in its ability to treat multiple types of pollutants with remarkable efficiency even at very low concentrations such as dyes, organic pollutants, acids, inorganic toxins, pharmaceutical waste and pesticides (Ibrahim et al., 2020a). However, the high cost of preparing adsorbents, their continuous decline in viability, and the loss of some of their mass during the regeneration process were important factors in the search for suitable and low-cost alternatives (Al-Hermizy et al., 2025). One of the most promising and successful alternatives with high adsorption efficiency, due to its unique properties, was various agricultural and industrial wastes. Agricultural and industrial waste dumped into the environment is diverse, including pomegranate peels, banana peels, lemon peels, orange peels, tangerine peels, almond shells, eggshells, pistachio peels, mandarin peels, watermelon rinds, used tea leaves, rice husks, mango peels, pineapple peels, sunflower seed husks, algae, tree leaves such as *Eucalyptus*, buckthorn, tangerine, water hyacinth, aluminum foil, plastic waste and others (Ali et al., 2024; Abdullah et al., 2023; Al-Hermizy et al., 2022; Jadooa et al., 2025; Hammed et al., 2025; Ali et al., 2020a; Ali et al., 2025; Alsarayreh et al., 2025b; Abed et al., 2025; Ibrahim et al., 2025a; Abdulkareem et al., 2023; Alhamadani et al., 2025; Mahmood et al., 2025; Abed et al., 2025). It is well known that these wastes accumulate in the environment in large quantities and continuously, leading to multiple environmental problems if not managed properly, as their uncontrolled decomposition contributes to greenhouse gas emissions, soil degradation, and water pollution (Hasan et al., 2025). These wastes can also become a breeding ground for pests and the spread of diseases, threatening agricultural productivity (Ibrahim et al., 2020b). Therefore, converting these agricultural and industrial wastes into absorbent materials for treating water pollution with heavy metals is an important environmental practice that can lead to a sustainable environment in the long term. Although these wastes accumulate and are laden with toxic compounds, the application of the zero-residue level concept has opened new horizons and changed the prevailing view of these wastes from harmful materials to important resources. For example, organic fertilizer, promoted bioethanol, nanomaterials, concrete reinforcers, and acetone were prepared from adsorption residues (Hamdi et al., 2024; Abbas et al., 2022a). The current investigation aims to utilize buckthorn leaves as a readily available, non-valuable, and inexpensive adsorbent for recovering divalent cobalt cations from contaminated solutions using a batch-type adsorption unit under various operating conditions to determine the maximum adsorbent efficiency.

2. EXPERIMENTAL PROCEDURE

2.1 Chemicals used

several chemical substances are used to determine the ability of buckthorn leaves to adsorb divalent cobalt cations. The details of all chemicals used in this investigation are listed in the Table 1.

Table 1: Details of chemical compounds used in the current study

Substance	Chemical formula	Purity, (%)	Molecular weight, (g/mol)	Form	Company	Country
Cobalt(II) nitrate hexahydrate	$Co(NO_3)_2.6H_2O$	≥ 97	291.03	red-brown crystalline solid	Hi-MEDIA	India
Hydrochloric acid	HCl	36	36.5	Colorless and odorless solutions	Bharad Fine Chemicals Industries Pvt. Ltd.	India
Sodium hydroxide	NaOH	≥ 95	40	White pellets	Loba Chemie and SLC Chemicals	India
Double-distilled water	H_2O	2.5 μs/cm at 25 °C	18	Liquid	Laboratory distillation unit	Iraq
Medical Alcohol	C_2H_6O	99%	46.07	Liquid	SLC Chemicals	India

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2.2 Equipment used

several equipment is used to determine the ability of buckthorn leaves to adsorb divalent cobalt cations. The equipment used in this investigation are listed in the Table 2.

Table 2: Details of equipment used in the current study

Device	Model	Company	Origin
Digital Water bath shaker	SHKA7000	Thermo-Fischer	USA
Atomic Absorption Spectrophotometer	AA-7000	Shimadzu	Japan
Benchtop pH meter	inno-PH18	Inno Tech Technologies	USA
Digital Electronic Weight Scale	JA103P	FAITHFUL	China
Laboratory grinder	RRH-100	RIRIHONG	China
Oven	B 30	Memmert	Germany
Vacuum Filtration Kit2	EIS-CH200501	CP Lab safety	China
Glass Water Still	GLF-2208	Lauda	Germany
Glassware	-	BOROSIL® Scientific	India

2.3 Stock solution

Five grams of cobalt(II) nitrate hexahydrate is dissolving using enough amount of double-distilled water using a clean, sterile beaker of appropriate capacity. After the cobalt salt had completely dissolved, the prepared solution is transported to 1-liter volumetric flask. Then the volume is filling up to the indicated mark to produce 1000 ml solution. The volumetric flask was covered with a layer of aluminum foil and kept in a dry, dark place to avoid any exposure to sunlight or light.

2.4 Calibration curve

The AAS calibration curve of divalent cobalt cations is shown in Figure 1.

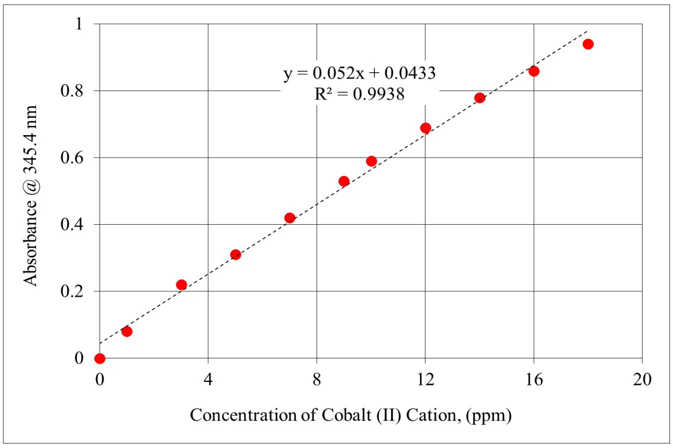

Figure 1: Divalent cobalt cation calibration curve of using AAS equipment

2.5 Adsorption unit

The batch adsorption unit consists primarily of a water bath shaker device. This device provides controlled mixing of samples while ensuring proper contact between the adsorbent and adsorbate, in order to achieve conditions that simulate real environmental conditions. After adjusting the pH and initial concentration of divalent cobalt cations, a specific mass of virgin watermelon rind powder is added to the experimental flask and charged into a water bath shaker. Before starting the experiment, the temperature and shaking speed of the adsorption unit are determined. The operating conditions investigated in this study were pH (1-8), initial concentration of divalent cobalt cations (1-5 ppm), contact time (5-180 minutes), virgin watermelon rinds dosage of (0.5-8 g), and agitation speed (100-500 rpm). Through equations (1) and (2), the percentage removal efficiency (%R) of the adsorbent and the adsorption capacity (q) of the adsorbent are determined, respectively (Alsarayreh et al., 2024):

$$\%R = 1 - \frac{C_f}{C_i} \qquad (1) \qquad\qquad q = \frac{V}{m_w}\left(C_i - C_f\right) \qquad (2)$$

Where: $\%R$: Removal efficiency (-); C_i and C_f: Initial and final concentration of divalent cobalt cations, respectively (ppm); q: Adsorption capacity of virgin buckthorn leaves (mg/g); V: Volume of solution in the experiment (0.1 L); m_w: Mass of virgin buckthorn leaves used (g).

3. RESULTS AND DISCUSSION

3.1 Acidity (pH)

The results obtained from changing this factor, as shown in Figure 2, indicate that the relationship between the two variables is a direct, non-linear relationship, representing ideal behavior, as the removal efficiency increases from 3.62% to 18.53% when the acidity is increasing between 1-6, then stabilizes at an almost constant value despite the increasing alkalinity of the contaminated solution. It is notable that at rising pH numbers, there is direct relation, i.e., the remediation efficiency is decreasing. This is attributed to the fact that this behavior creates strong competition with divalent cobalt cations for active sites on the adsorbent surface, leading to a decrease in the number of target ions bound to the virgin buckthorn leaves, and consequently, low removal efficiency. Increasing the pH value leads to increased removal efficiency, and the protonation of the absorbent surface and the number of positive hydrogen ions in the contaminated solution begin to decrease (Mahmood et al.,2025).

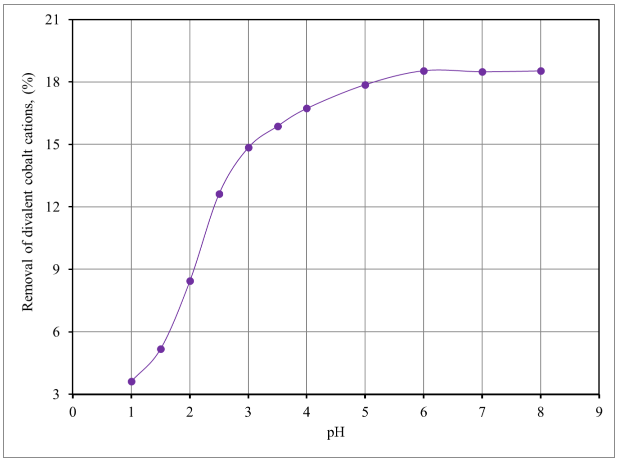

Figure 2: Influence of pH on the removal of divalent cobalt cations from contaminated aqueous solution.

3.2 Agitation speed

The results obtained from changing this factor, as illustrated in Figure 3, indicate that the relationship between the two variables is a non-linear direct relationship, where the removal efficiency increases from 6.471% to 28.65% related with increasing agitation speed from 100-400 rpm, respectively. This enhancement in the performance can be attributed to that the raising the agitation speed results in enhanced mixing and corresponding efficiency (Ali et al., 2024). Upon override the agitation speed the value of 400 rpm, it is evident from the Figure above that the remediation ability stays stable and unaltered. This result may be ascribed to the system reaching a state of dynamic equilibrium between the virgin buckthorn leaves and divalent cobalt cations, wherein the quantity of ions adsorbed is equivalent to the quantity of ions desorbed from the surface of the medium (Ali et al., 2025).

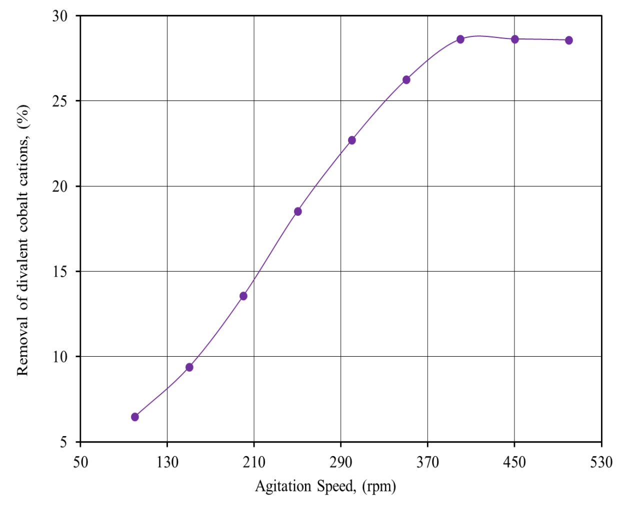

Figure 3: Influence of agitation speed on the removal of divalent cobalt cations from contaminated aqueous solution.

3.3 Initial concentration

As shown in Figure 4, there is an inverse relation between the percentage removal and initial concentration of target cations. It is observed that the removal efficiency decreases from 33.39% at initial concentration of 0.5 ppm to 7.424% at initial concentration of 4.75 ppm. This decline is ascribed to the unchanging quantity of active binding sites on the surface of buckthorn leaves, which is capable of holding a particular amount of the divalent of cobalt cations. At low initial concentration, the adsorbent can be able to effectively store a more significant number of divalent cobalt cations (Hameed and Abbas, 2024).

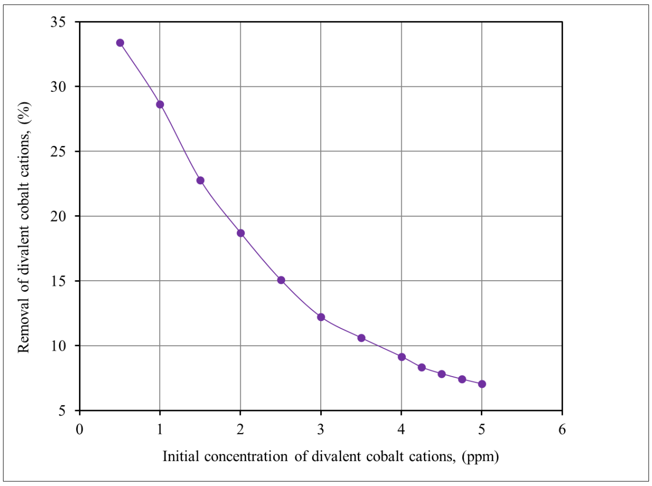

Figure 4: Influence of initial concentration on the removal of divalent cobalt cations from contaminated aqueous solution.

3.4 Adsorbent dosage

As shown in Figure 5, the relationship between the remediation efficiency and the adsorbent dosage is a direct. The increase in remediation efficiency from 7.424% to 72.45% with an increase in the dosage of buckthorn leaves from 1 to 7 g is clearly attributable to several factors. Firstly, the surface area will be increases due to increasing in the amount of the adsorbent, which leads to increasing the active sites responsible for capturing the target polluted cations, as the initial concentration is constant. Upon reaching a dose of 7 g of virgin buckthorn leaves, the adsorption efficiency reaches its maximum value due to saturation. This is a logical behavior that reflects the distribution of a relatively constant amount of adsorbed pollutant over larger masses of adsorbed material, thus reducing the amount of pollutant adsorbed per unit mass (Mahmood et al., 2025).

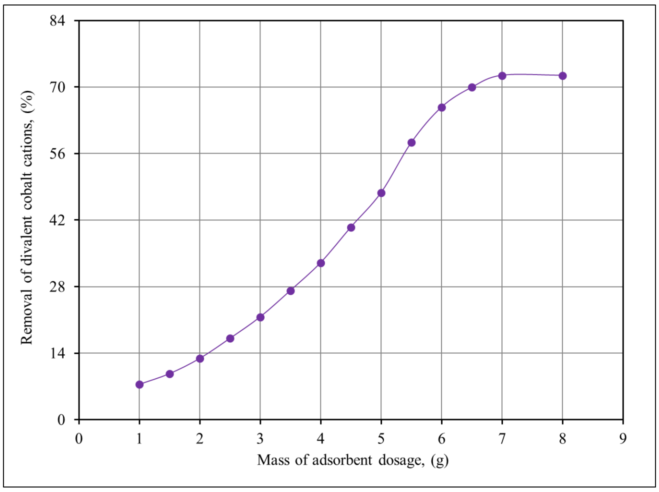

Figure 5: Influence of adsorbent dosage on the removal of divalent cobalt cations from contaminated aqueous solution.

3.5 Contact time

As shown in Figure 6, the relationship between the remediation efficiency and contact time is a non-linear direct relation, as the remediation efficiency rising from 6.92% at 5 min. to 72.45% at 150 min. This known result may be demonstrated by ad the increasing time leads to increase the opportunity of accessing the target cations to adsorbent's surface and captured by the active sites speared (Alhamadani et al., 2025). Once the contact time exceeding of 150 minutes, the maximum value of remediation efficiency is conducted, and no considerable increasing in the adsorption of divalent cobalt cations takes place due to the saturation state, such that the virgin buckthorn leaves cannot receive any further number of divalent cobalt cations.

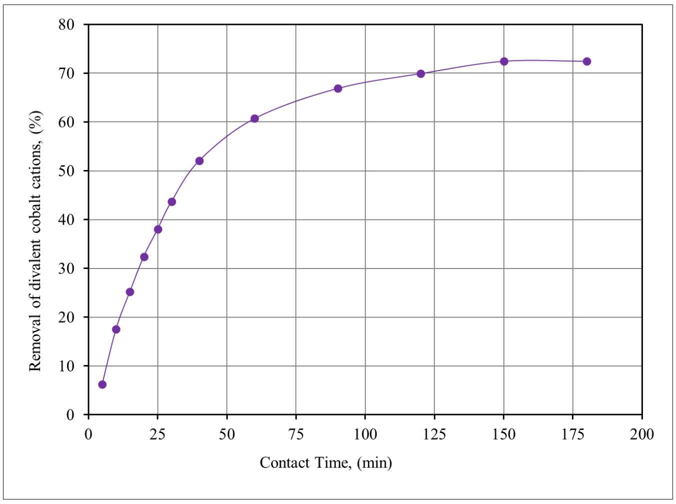

Figure 6: Influence of contact time on the removal of divalent cobalt cations from contaminated aqueous solution.

4. Characterization of Adsorbent

The adsorbent material, (buckthorn leaves powder), was characterized before and after adsorption of cobalt cation using two types of non-destructive analysis: X-ray diffraction (XRD) and X-ray fluorescence (XRF) analysis, to verify the adsorption capacity of adsorbent material. The results indicated highly efficient adsorption, as shown in Figures 7 and Figure 8 which represent XRD and XRF analysis, respectively.

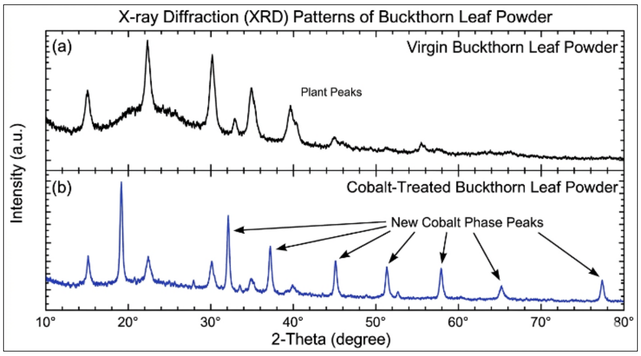

Figure 7: X-ray diffraction (XRD) analysis of buckthorn leaves powder, before and after adsorption of cobalt cation.

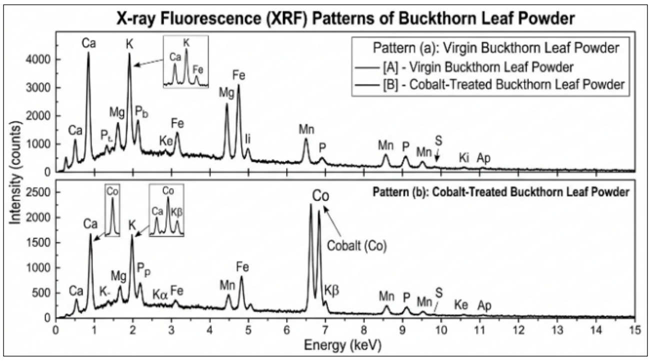

Figure 8 X-ray fluorescence (XRF) analysis of buckthorn leaves powder, before and after adsorption of cobalt cation.

5. CONCLUSIONS

The current study reached several conclusions, the most important of which is that agricultural waste no longer represents an environmental problem, but has become an important resource for addressing environmental problems resulting from human activities. The results of this study proved that a dose of 70 g of virgin buckthorn leaves, is capable of treating one liter of aqueous solution contaminated with divalent cobalt cations with an efficiency exceeding 72.45%, at a pH of 6, 4.75 ppm initial concentration of cobalt in water, at 400 rpm agitation speed, and 150 minutes.

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