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A REVIEW OF TOXICOLOGICAL, ENVIRONMENTAL AND HEALTH EFFECTS OF CHROMIUM FROM AQUEOUS MEDIUM; AVAILABLE REMOVAL TECHNIQUES

REVIEW

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ABSTRACT

Heavy metals are unwanted pollutants introduced directly and indirectly into the environment and ecological currents through the discharge of industrial wastewaters. Many of them, like hexavalent chromium, easily enter biological organs resulting in acute toxicity and damage to kidney, liver, and lung due to their maximum oxidation state in comparison with their other compounds. On the other hand, they incur irrecoverable effects on the environment and ecosystems. Accordingly, human beings need processes and technologies to lessen the danger of these pollutants; in order to remove chromium from aquatic environments, various methods including physical, chemical, and biological methods have been important. Among them, reverse osmosis, ion exchange, electro dialysis, chemical deposition, and adsorption are popular. In practice, in order to apply each of the mentioned treatment methods, preliminary studies for applicability, the required expertise, and the costs of construction and operation are necessary.

***Key words**: Chromium, Toxicological effects, Environmental effects, Health effects, Removal techniques.*

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Introduction

First-degree toxic contaminants are materials that have environmental hazards and are damaging to human health(1). Heavy metals are stable contaminants that, unlike organic compounds, cannot be degraded to chemical or biological processes in the nature, bringing about considerable harmful effects on vegetation and animal environments health(2-3). Chromium is one of the widely utilized metals in today's world and industrial processes. Hexavalent chromium, has been regarded as carcinogenic materials by the international agency of cancer research, European Union, and American toxicology program. The allowable limit for them to be discharged into surface waters is 0.1 mg/L and for potable water is determined to be 0.05 mg/L(3-4).

Chromium

Chromium has a Greek root, adapted from the word Chroma, meaning color. Similar to many other metals, chromium can exist in various forms including oxidation, neutral, and zero (metal) up to six (hexavalent). The most common oxidation states are found in the form of trivalent and hexavalent(6) .The hexavalent chromium in groundwater is usually found in the form of the anions of chromate (CrO_4^{2-}), bichromate ($HCrO_4^{-4}$), and dichromate ($Cr_2O_7^{2-}$), depending on pH(5-7).

Applications and sources of chromium

In the 20th century, chromium was recognized as a strategic metal to be used in tens of special industries to produce numerous consumer goods as a raw material. It is widely used in electroplating, tannery and tanning, cannery, dye making, and wood industries as stabilizer(5). Chromium is also used in manufacturing green glasses, light colors in automobile manufacturing, steel industries, tanning and dyeing processes and even washing laboratory dishes(5). The chromium available in the ecosystems and nature reflects its two origins. The first group is those that are found naturally in water and soil. On the other hand, the second groups are the resources with an anthropogenic or man-made origin.

In general, the major sources of chromium exist as follows(5); The natural sources for chromium (Chromium with natural origin and available in soil, stone, surface, groundwater and atmosphere), Man-made sources of chromium (Chromium with human origin available in the soil, agricultural materials, Sewage, Coal residues and ash, the mines and smelting wastes, water and atmosphere).

Disease and adverse Health effect

The chromium ions can be introduced to human and animal body from follows ways: (i) inhalation of aerosols, fumes, or mists, (ii) Skin, and (iii) Eating, drinking, or smoking (in regions in which chromium is present).Chromium compounds are toxic and cause damage to nasal mucosa, asthma, skin inflammation, dizziness, stomach ulcer, convulsion, and kidney damages. Its accumulation in animal and plant tissues can lead to serious dangers. The hexavalent form is the most toxic form of chromium, while the trivalent for has the least toxicity (500-1000 times less), bearing in mind that it is an essential nutrient for the body (an essential agent for the metabolism of lipids, proteins, and fats), commonly seen in the form of deposit, capable of leaving Mai phase(3).

Toxicological mechanism of chromium

According to several regulatory and non-regulatory agencies, hexavalent chromium [Cr(VI)] is a toxic industrial pollutant that is classified as human carcinogen. Cr(VI) compounds, which are powerful oxidizing agents, appear to be much more toxic systemically than Cr(III) compounds, given similar amount and solubility(9). Cr(VI) in the form of the tetrahedral chromate anions, pass through cell membranes and accumulate in cells(10). Within cells, Cr(VI) is reduced by hydrogen peroxide (H2O2), glutathione (GSH) reductase, ascorbic acid, and GSH to produce reactive intermediates, including Cr(V), Cr(IV) ions and oxidative radicals, and ultimately convert to the Cr(III). When reduction of Cr(VI) occurs at a distance from the target site, it considered as being a detoxification process while reduction of Cr(VI) may serve to activate chromium toxicity if it takes place in or near the cell nucleus of target organs(9).

However, the precise mechanism(s) of Cr (VI) carcinogenicity is not well understood, three mechanisms have been proposed to attributes chromium(VI) to the genotoxicity (Fig. 1).

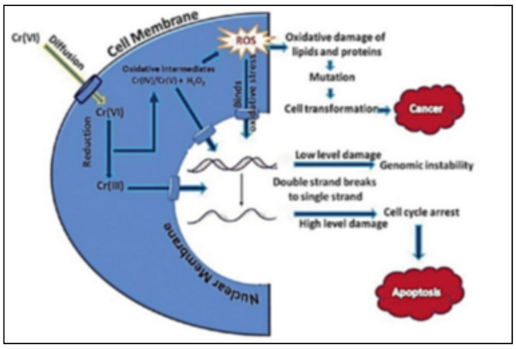

Fig. 1: Possible mechanism of hexavalent chromium intracellular toxicity, carcinogenicity leading to cell cycle arrest(16).

- Produce the highly reactive radicals,
- Direct binding of chromium(V) to the DNA,
- Binding to the DNA of the end product of the chromium(III) reduction(11).

It has been suggested that Cr (VI) mediates a majority of its genotoxic and cytotoxic effects by inducing oxidative stress, protein-DNA crosslinks, DNA single and double-strand breaks, and forming stable-Cr-DNA adducts(12). These Cr DNA adducts are mutagenic and may altering the DNA code during the cell transformation process(10). Cr (VI) induced DNA-damage can alterred gene expression by affect DNA replication, transcription, and translation(13).

Chromium also can be interferes with DNA damage response and repair, induce apoptotic celldeath(10) through inhibition ofantiapoptotic mediators (PI3K and AKT)(14).

Chromium causes changes in DNA methylation, histone modifications and miRNA expression(15). Endocytosis or pinocytosis of compound containing Cr(III), especially those that are less water soluble, by cells have been interpreted to be the mechanisms responsible for the widely observed genotoxic effects of Cr(III) compounds. Cr(III) reacts in vitro with DNA and proteins, that are likely involved with its cytotoxic effects(10).

The adverse effect on environment

Due to the toxic nature of chromium, it brings about many problems in the environment through producing wastes, mine wastes, and the final ash. In response to improper discharge of chromium, it enters soil and aquifers, causing many environmental problems including the mortality of creatures and aquatics. This toxic metal can involve a wider range of the environment through the food cycle. Chromium is toxic to the majority of plants and is considered as unnecessary substance for the growth. This element is introduced to plant structures in conjunction with nutrients essential for the plant(such as iron, sulfate, and nitrate).

Many researchers have attributed low growth of stalks and leaves, low stature of the plant, disorders in the growth and function of roots and the germination process to introduction of certain amounts of chromium. In addition to physiological damages, chromium also disrupts photosynthesis and metabolism in plants. Matvieieva and Duplij have been demonstrated that the duckweed plants growth in presence of the hexavalent chromium can be undertaken(17). Chromium involves enzymatic and metabolic activities in the systems inside plants, producing active oxygen. This oxidative oxygen can then oxide trees and plants(18,19).

The processes for removing chromium

Since the majority of heavy metals interfere with biological reactions of living cells, they result in disruptions of metabolic activities, thus these methods are less applicable(20). Removing chromium from water and wastewater is carried out through various methods including physical, chemical, and biological methods. In this regard, reverse osmosis, ion exchange, electro dialysis, chemical deposition, froth flotation, absorption of a solution, biological absorption, and adsorption can be mentioned.

Chemical oxidation-reduction

Typically, for removing chromium from wastewaters, chemical oxidation reduction processes are used. In this method, by reducing pH to around 2 and developing acid conditions, the chromium is reduced, and by adding an alkaline agent and increasing pH to about 9-10, chromium is discharged in the form of deposit in the next stage from the aqueous matrix.

Reduction of hexavalent chromium

Hexavalent chromium is a strong oxidant and accordingly can be reduced in the presence of electron donor materials. The most common form of chromium has been reported as soluble in natural waters at the normal pH in the forms of chromate(CrO_4^{2-}), bichromate($HCrO_4^{-4}$), and dichromate($Cr_2O_7^{2-}$). These compounds are the most frequent forms of hexavalent chromium and are easily reduced in the presence of electron donors such as organic compounds and minerals.

Important factors in this regard are soluble bivalent iron, the compounds containing divalent-iron, sulfides, and organic compounds. Also, chromium is naturally reduced in the atmosphere and top surfaces of waters, known as Photoreduction. Other ways to reduce chromium and change it into the trivalent form are biological and chemical reactions. The biological reduction requires a longer time in comparison with the chemical method so that it can reduce chromium. For example, the chemical reduction of chromium using bivalent iron is about 100 times faster than biological reactions. However, use of iron and sulfide for reduction of hexavalent chromium is the preferred method. Although use of aerobic and anaerobic biological processes is suggested for reduction of chromium, anaerobic conditions have shown better(5)

Reduction and oxidation by inorganic and organic mater

Reduction of hexavalent chromium in natural waters is performed by divalentiron ion. Divalentiron is supplied by erosion of iron containing materials and discharge of industrial wastewaters. Monovalent copper(reduced) [Cu(I)] can also be effective in reducing chromium. These conditions are suitable for surface waters and atmosphere with low pH and acid power(21). Pyrite(FeS_2) contains both reducer compounds of iron and sulfur. From among the compounds containing iron, hematite and biotite have been suggested by Palmer(22).

Application of iron under alkaline and neutral conditions has been reported to be far better than using acid pHs. When FeO is used, the reaction of chromium reduction is done as follows(5);

$$[3FeO] + 6H^+ + Cr^{6+}_{(aq)} \rightarrow Cr^{3+}_{(aq)} + 3Fe^{3+}_{(aq)} + 3H_2O \quad (1)$$

In acidic waters, the final product is in the form of chromium and iron(III), while at alkaline and neutral pHs, owing to insolubility of Fe(OH)3, chromium deposits in the form of Cr(OH)3. Although most sulfides(S^{2-}) have low insolubility, reduced sulfur(encompassing; S, S^{2-}, H_2S and $S_2O_3^{2-}$) can be regarded as a major factor in reducing chromium(VI) to chromium(III). Studies have shown that reduction is carried out by sulfide factors at the outset of the reaction with a high rate, which then quickly begins to decline. Reduction reactions are typically completed after around one day. The main reaction for this cease is as follows(5),

$$Cr_2O_7^{2-} + 8H^+ + 3H_2S \rightarrow 2Cr^{3+} + 3S + 7H_2O \quad (2)$$

Organic compounds are among important reductive compounds in soil capable of reducing chromium. Organic compounds such as humic and folic acids as well as amino acids in soil are notable reducing agents. Furthermore, pH, the concentration of chromium and the level of reductive organic compounds such as humic acid do not affect the level of reduction. The half-life of reduction of chromium with humic acid is around 3 days, where within the pH range of 4-7, several days is required for reduction to the trivalent form(5).

Adsorption techniques

Adsorption is a process in which molecules are concentrated onto the surface of an adsorbent. In this process, the adsorbed molecules move away from the bulk phase towards the spaces available in the surface. The driving force required for adsorbing contaminants is the ratio of its concentration to the solubility of the present compound. Adsorption is widely used in the removal of chromium from industrial effluents. Investigations have shown that factors such as the initial concentration, pH, available surface area, temperature, and the type of adsorbent are among the important factors in the adsorption process(23). The usefulness of adsorption manifests itself based on advantages including low cost, availability, profitability, ease of action and operation in comparison with other methods such as ion exchange and filtration with membranes, both environmentally and economically(24).

In this regard, a wide range of natural and artificial compounds have been used for removing hexavalent chromium, including active carbon, biological adsorbents, zeolite, chitosan, and commercial adsorbents(25). Active carbon has been evaluated out adsorbents with high-capacity and extraordinary capability in adsorbing chromium. High efficiency in the active carbon has been reported thanks to huge spaces in the surface of the adsorbent, high specific area(500-1500 m^2/g), and presence of hydroxyl genera(26).

Bioadsorption

With regard to technical and economic aspects of chromium removal methods, development of a simple, robust, and cost-efficient technology for chromium removal is critical. Recently the use of biomaterials(such as: waste sludge, algae, yeast and fungal biomass) for chromium ions removal from industrial wastewater has become as a non-conventional alternative technique to conventional techniques. Beside the cost-efficient aspects, this method is provided notable removal efficiency. For example, Hossini et al. (2015) have been reported that the removal efficiency more than 95% was obtained when date palm fiber was used for initial concentration ofCr(VI) about 200(mg/L)(27).

Filtration

Today, application of membrane techniques in treating wastewater has shown a remarkable increase. Furthermore, their high potential for removing heavy metals such as hexavalent chromium has also been of great interest. From among the various types of membrane filtration one can mention organic, polymer, and liquid membranes used for removing chromium. Organic membranes are one of the most important porous materials with high thermal and chemical stability(29). Ceramic and organic membranes belong to this group, nowadays mainly developed in nanostructures. Polymer membrane separation systems are now regarded as an important technology in treating wastewater, facilitating removal and recovery of contaminants from solutions such as water(30). High selectivity is one of the important characteristics of liquid membranes. Two of the most important types of liquid membranes are emulsion liquid membrane (ELM) and immobilized liquid membrane(ILM), where the latter is also called supported liquid membrane (SLM)(31).

Ion exchange

In recent years, among the physiochemical methods developed for removing chromium from wastewater, ion exchange has changed into a popular method to this end. Lin et al. used Resin Ambersep 132 to recover chromium through a four-stage ion exchange process(32). The type of resins utilized in the study by Sapari was Dowex2-X4, managing to treat real wastewater with an efficiency of 100%(33).

Biological processes

In general, the aim of biological methods in treating wastewaters is to remove biodegradable organic compounds, where the available evidence indicates low efficiency for removing heavy metals by this method. During studies by Stasinakis et al on active sludge process, it was found that the efficiency of removing trivalent chromium can be 90% due to adsorption on suspended solids of the wastewater. However, the efficiency of the removal of hexavalent chromium across all experimented samples did not exceed 15%(34). This is because the majority of heavy metals interfere with the biological reactions of living cells, resulting in disorders of metabolic activities. Therefore, these methods are not used very frequently. There are few valuable microbes that are able to reduce hexavalent chromium in the presence of acetone or H2 as electron donor. In this group, one can mention *Pseudomonas aeruginosa, Enterobacter cloacae, P. fluorescens, and actinomycetes*(35). In the biological reduction process of chromium, the efficiency of anaerobic bacteria has been reported to be far greater than that of aerobic bacteria. In biological reactions, chromium acts as electron acceptor(5).

Sulfate reducing bacteria

Various strains of Sulfate Reducing Bacteria (SRB) are able to convert hexavalent chromium (with concentrations > 500 μM/L) into trivalent chromium with less toxicity. When the redox potential reaches around -400 mV, the best performance in the reduction of chromium has been reported. The ability for reducing the metal by hydrogenase enzyme has been reported by Chardin et al.(36).

The main mechanism of hexavalent chromium reduction is taken during the ATP synthesis, and sulfate reduced to sulfite, then hexavalent from reduced to trivalent (green precipitate) by transferring electron from cytochrome.

Sulfate reducing bacteria convert the sulfate/thiosulfate/sulfite to sulfide ions, and consequently, the sulfide ions reduce the Cr^{6+} to Cr^{+3} and theirs oxide to S^0(37).Following reactions can be showing this conversion;

$$SO_4^{2-} + 10H^+ + 8e^- \rightarrow H_2S + 4H_2O \tag{3}$$

$$S_2O_3^{2-} + 10H_2O \rightarrow SO_4^{2-} + HS^- + H^+ \tag{4}$$

$$4SO_3^{2-} + H^+ \rightarrow 3SO_3^{2-} + HS^- \tag{5}$$

$$3HS^- + Cr_2O7_3^{2-} + 11H^+ \rightarrow 3S^0 + 2Cr^{3+} + 7H_2O \tag{6}$$

And finally, the overall reaction;

Electrocoagulation (EC)

Today, electrochemical techniques, owing to presence of toxic compounds and resistant to biological decomposition in industrial wastewaters, have attracted a great deal of attention as an option for treating wastewaters. EC process has been introduced as simple design, low startup and operational cost, no need to special chemicals, and production of little side sewage(38). The results of conducted research have revealed the success of iron and aluminum electrodes in reducing chromium. Aluminum, as an anode in the electrode-coagulation process, easily develops monomeric, polymeric, and amorphic complexes, resulting in the deposition and reduction of dichromate. Due to low internal resistance, the aluminum electrode conducts currents easily and reduces the potential of oxidation-reduction(39).

Conclusion

Chromium is common and widely used metal, which enters the ecosystem in high amounts through discharge of industrial wastewaters. The high potential of destructive health and environmental effects of chromium has resulted in the fact that special strategies are consider for treating and disinfecting effluents and waters containing it. Among the common methods for treating chromium and heavy metals one can mention processes such as physical and chemical adsorption, ion exchange resins, membrane techniques, chemical deposition, and electro chemistry, each of which is helpful in some way in removing these toxic pollutants. Furthermore, usage of each of these techniques is done under economic conditions and based on availability of the process.

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