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Methods for Fluoride Removal from Industrial Wastewater: A Review Study

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Abstract

Today, due to the increase in industries such as steel manufacturing, a lot of wastewaters is released into the environment, which, due to the presence of harmful ions and salts, causes environmental hazards and human health problems. One of the harmful compounds present in water is fluoride compounds. According to the World Health Organization guidelines, the maximum concentration of fluoride should be limited to 1.5 milligrams per liter in drinking water. Excessive fluoride is harmful to human health, and one of the most important and well-known effects of receiving high levels of fluoride is fluorosis. Various methods such as sedimentation and coagulation, membrane filtration, ion exchange, and adsorption are proposed and used to treat wastewater containing fluoride. This research aims to investigate the methods of fluoride removal from industrial wastewater and to introduce the best economical method for the steel industry. Among all the methods, the coagulation method can be a suitable option for fluoride removal due to its easy implementation and lower economic cost compared to other methods. Among the coagulation methods, coagulation using Fe(VI) salts is recommended for the steel industry due to its lower environmental hazards, better efficiency, and more suitable price.

Keywords: Fluoride, Industrial Wastewater, Coagulation Method, Steelmaking, Fluorosis

Introduction

Water pollution, in addition to causing the spread of various diseases, has also affected the health and limited quality of clean water, and in the long run, has caused significant damage to the economic and social development. In contrast, by using appropriate wastewater treatment methods, not only can the health of the community be guaranteed, but by recovering the treated wastewater and reusing it, part of the water shortage can also be compensated. From this perspective, the recovery of wastewater and industrial effluents, especially in countries suffering from water scarcity or lack of water, has become particularly important [1]. Currently, in Iran, the recycling of wastewater and its return to industry has also received attention, and many industries in the country are taking steps to recover industrial effluents for the purpose of economic savings and creating conditions for development [2]. Wastewater contains a high percentage of heavy metals and harmful compounds, one of which is fluoride, which has adverse effects on nature and human health. Fluoride compounds are widely found in natural water bodies, and fluoride is also one of the toxicological indicators of water quality and essential trace elements for the human body [3]. In general, the fluoride concentration in drinking water should be controlled between 0.4 to 0.6 milligrams per liter. According to the World Health Organization guidelines, the maximum concentration of fluoride should be limited to 1.5 milligrams per liter in drinking water. The fluoride content of water in many parts of the world, including Iran, has been higher than the accepted standards [4]. The fluoride ion, as the most electronegative element in the periodic table and one of the halogen group members, has a strong tendency to form and compound with other cations. One of the most important and well-known effects of receiving high levels of fluoride is fluorosis, which can be chronic or acute [5]. Chronic fluoride poisoning can cause the following damages:1. Corrosion or penetration of the skin 2. Damage to the eyes, trachea, and nose of workers due to the volatility of the fluoride released from the factories 3. Severe damage to hard tissues and bone cells, osteoclasts, tooth cells, prevention of growth and development, and mineralization with hard tissues 4. Reduction of blood calcium content, which can disrupt thyroid function, increase the number of osteoclasts, and intensify bone absorption, and cause disruption in the activity of several enzymes in the kidneys, liver, and other sensitive organs of the body [6]. Given the diversity of materials used in industries, the chemical composition of wastewater can vary under different conditions, and various physical, chemical, and biological methods can be used to separate different substances. Oxidation and disinfection, as well as coagulation and flocculation, are important processes in water and wastewater treatment. In many industrial effluents, suspended particles do not easily settle or float because they repel each other or have a small diameter and settle slowly. By using specific chemicals, under chemical and physical interactions, these particles are neutralized, absorbed by each other, and form larger particles that settle due to their own weight [7].

Work method

In the present article, after searching for relevant articles published in databases such as Springer, Science Direct, John Wiley, Scopus, and Freepaper, the references in the text were selected and examined to obtain the latest findings on methods for removing fluoride from water. In this research, keywords such as fluoride, methods for fluoride removal, and the advantages and disadvantages of fluoride removal methods were used to search the databases.

Findings

Methods for removing fluoride from aqueous solutions include physical, chemical, and biological processes. Many studies have been conducted on the treatment of fluoride from industrial effluents. In this section, the most common methods for removing fluoride from industrial wastewater are briefly reviewed.

Membrane treatment method

Membrane processes such as reverse osmosis, nanofiltration, and electrodialysis have been studied for fluoride removal. In this method, the water contaminated with fluoride is exposed to a membrane, and separation occurs. Membrane processes are classified based on the size of their pores as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis. Membrane processes work based on differences in pressure, temperature, concentration, and electrical potential. Membranes that work based on pressure difference include microfiltration, ultrafiltration, nanofiltration, reverse osmosis, gas transfer, and pervaporation. Usually, this group of membrane processes is among the more common membrane separation processes. In reverse osmosis, a semi-permeable membrane is used. In this method, there is a need to apply pressure to the contaminated water. The selection of this method depends on the available facilities, price, water type, membrane type, etc. [8]. The efficiency of the reverse osmosis method depends on the water characteristics, pressure, temperature, routine conditions, and maintenance. Reverse osmosis also produces a waste stream that needs to be further treated. This method requires a large amount of clean water, which is not suitable for water-scarce regions. The most important factor is the high cost of the reverse osmosis method for treating large volumes of industrial wastewater [9]. The electrodialysis method is a membrane-based process that uses an electric current. Separating fluoride ions from water using this method requires a large amount of energy [10]. It is worth noting that after membrane fouling, the filter needs to be regenerated to be reused. Reverse membrane washing for reuse depends on the amount of heavy metals present in the contaminated water. Usually, industrial wastewater must first undergo preliminary treatment, and then membrane processes can be used to remove heavy metals and fluoride; therefore, membrane processes are costly [11].

Ion exchange technology

Ion exchange resins are a very wide range of organic polymeric compounds that act as a very suitable medium for ion exchange. These resins contain negatively or positively charged sites that can easily absorb oppositely charged ions from the surrounding solution. These resins are porous, lightweight solids that are usually available in the form of sheets, beads, or granules. When these resins are immersed in a solution, they absorb the solution and will eventually swell. These resins are made from styrene-divinylbenzene copolymers [12]. It should be noted that other compounds such as phenol-formaldehyde polymers and methacrylic-divinylbenzene are also used. Interestingly, the charged groups include quaternary ammonium salts, fulvic or sulfuric acid salts. Ion exchange resin is a type of synthetic compound or a natural compound (called zeolite) that has a high adhesion property. These resins are easily soluble in alcohols.
One of the most important and common uses of resins is as a filter in water treatment to remove ions and water hardness. Ion exchange resins are used to remove fluoride from drinking water. Studies have shown that resins with strong anionic functional groups are useful for this purpose [12]. Fluoride can replace the chloride ion on the resin until the chloride ion sites are exchanged with fluoride. This process continues until the ionic sites containing chloride are present. Then the resin is washed with a sodium chloride solution so that the chloride ion replaces the fluoride, and the resin becomes reusable again. Fluoride replaces chloride because it is more electronegative. The cost of the ion exchange method is

high, and the treated water has a low pH and a high chloride content. Haroun et al. also studied the removal of fluoride through a two-way ion exchange cycle. This two-way system uses two anion exchange columns, and this system shows an effective process for removing fluoride from water [13]. Chubar et al. created a new ion exchanger that can simultaneously remove various anions such as fluoride, chloride, bromide, and bromate [14].

Adsorption Technology

Surface adsorption is one of the methods for removing fluoride from drinking water, which is a separation process in which some components of the fluid phase are transferred to the surface of a solid adsorbent. In surface adsorption, a porous solid is used, which creates a very large surface area. That is, we not only have the external surface but also the porosity, and the more the adsorbent surface area increases, the greater the adsorption capacity [15]. Nowadays, fluoride removal using adsorption by various materials is preferred due to the many advantages of this method, such as low cost of the materials used, easy setup conditions, high adsorption capacity, and the possibility of reusing the adsorbent materials. In recent years, the adsorption method has been more extensively studied, and this method produces high-quality water and is applicable for large volumes of water, generates a small amount of sludge, and has a high pollutant removal capacity [15]. Researchers have developed various adsorbents with high adsorption capacity and cost-effective for fluoride removal, such as activated alumina, activated carbon, calcite, activated sawdust, activated coconut shell carbon, coal, bacteria, sand adsorbents, charcoal, and other adsorbents reported in the literature. The adsorption method has a high efficiency, and activated carbon and zeolite are two materials with high surface adsorption power that can be used for fluoride separation. By passing water through a carbon filter, fluoride is adsorbed onto the pores of the activated carbon, and removal is achieved. The surface of activated carbons is gradually covered with a large amount of sludge, fluoride, and other salts, and the efficiency decreases. Therefore, the carbons must be periodically regenerated, and their surface should also be washed. Among the industrial adsorbents, compounds based on activated alumina, metal oxides, hydroxides, metal, and organic compounds, and carbon-based materials can be mentioned. Activated alumina is of great interest for fluoride adsorption due to the following characteristics: high specific surface area, specialized porous structure, excellent adsorption performance, and high adsorption efficiency. Biomasses such as dried fruit peels can be used as good adsorbents for fluoride. To improve the adsorption capacity, the combination of adsorbents can be used [16].

Sedimentation and coagulation

The precipitation method involves adding chemical agents, coagulants, and flocculants to the wastewater to form a fluoride precipitate and then achieve the goal of fluoride removal in water through the separation of the solid from the liquid. The treatment efficiency of sedimentation is mainly determined by the flocculating agents, reaction conditions, and the effect of solid-liquid separation. The sedimentation method is generally carried out in two ways: chemical sedimentation and coagulation sedimentation. For high concentrations of industrial wastewater containing fluoride, the chemical precipitation method is usually used, in which lime, calcium carbide slag, calcium chloride, and other modified compounds are added to the wastewater. Through a chemical reaction, the calcium ion and the fluoride ion combine to form CaF_2 as a precipitate, and ultimately, the fluoride is separated by the precipitation process. Currently, this treatment technology is quite significant for industrial wastewater with high fluoride content. However, complete treatment by this method is not possible because the fluoride content in the wastewater is generally less than 20-30 mg/L [17, 18]. Also, the CaF_2 precipitate particles are usually small and do not settle quickly. This is the main weakness of the precipitation method, and therefore, the use of flocculants such as polyaluminum chloride should be used for the flocculation process to achieve better separation of the solid phase [18]. The coagulation sedimentation method is mainly used for wastewater with a relatively low fluoride content in the industry. Conceptually, there is no clear difference between the coagulation sedimentation method and the chemical sedimentation method. The coagulation method mainly uses various coagulants to form a precipitate by adding a coagulant to the industrial wastewater, which stimulates the formation of more positively charged colloidal particles and effectively adsorbs the fluoride ions to form a precipitate and remove fluoride from the water. Currently, aluminum salt coagulants, mainly polyaluminum, aluminum sulfate, and aluminum chloride, are often used. In the precipitation methods for fluoride removal using calcium, it has been a successful method [19]. For wastewaters with very high fluoride levels, fluoride removal can be done in two stages. In the first stage, precipitation with lime, and in the second stage, with the addition of alum and polyelectrolyte [20]. In 1961, the National Environmental Engineering Research Institute (NEERI), Nagpur, developed an economical and simple method for fluoride removal, known as the Nalgonda technique. The Nalgonda technique involves the addition of aluminum salts, lime, and bleaching powder, followed by rapid mixing, flocculation, precipitation, filtration, and disinfection. Excess lime is used to facilitate sedimentation. Lime forms larger flocs and dense bubbles for faster sedimentation. During the flocculation process, many types of negatively charged fine particles and ions, including fluoride, are partially removed by electrostatic attachment to the flocs [21].

Discussion

The selection of the operating method for fluoride removal depends on the available facilities and practical conditions, but in any case, considering various aspects, it is considered an economically justified method. There are various methods for the treatment and reduction of fluoride, which should be cost-effective based on the need. A wide range of coagulants, oxidants, and disinfectants are used for water and wastewater treatment. An ideal and efficient chemical disinfectant should be able to inactivate microorganisms and to some extent decompose and oxidize organic and inorganic pollutants, suspended and colloidal particles, and heavy metals. The commonly used oxidizing agents and disinfectants include chlorine, bromine, iodine, hypochlorite, chlorine dioxide, and ozone, which are used to eliminate harmful organisms and control and remove odorous residues [22]. The most important coagulants used include lime, ferric sulfate, aluminum sulfate, ferrous sulfate, ferric chloride, and polyelectrolytes (synthetic organic polymeric materials). Usually, for the removal of colloidal materials from water and wastewater, metal compounds such as aluminum, iron, or some electrolytes are used. Extensive studies have been conducted on the chemical coagulants used in industries, including calcium salts, aluminum salts, and iron salts. Coagulants such as aluminum sulfate, calcium chloride, ferrous sulfate, ferric chloride, calcium hydroxide, and magnesium chloride have been studied for reducing fluoride concentration. In recent years, a new type of coagulant material called Inorganic Polymer Flocculants (IPFs) has also been prepared using the usual iron and aluminum salts, and they are increasingly being used in many parts of the world, especially in China, Japan, Russia, and Western European countries [23, 24]. In most developed countries, credible and numerous reports and studies on the impact of aluminum compounds on human health have led the water treatment industry to move away from the use of aluminum sulfate in water treatment, and the use of inorganic polymer coagulants like polyaluminum chloride (PAC) for the treatment of industrial effluents is carried out while observing environmental considerations and regulations. Compared to $AlCl_3$, the polymer aluminum chloride PAC has a stable network structure that adheres to fluoride compounds on its surface through ion exchange and neutralization processes, which is easier than replacing the hydroxyl ions, and thus enhances the ability to remove fluoride. However, these coagulants or oxidants and disinfectants also create some problems. During the use of some disinfectants, the possibility of the formation of byproducts that are somewhat toxic to humans and aquatic life has been reported [25]. The aluminum salts used as coagulants can be problematic due to the production of aluminum compounds in the water, and since their pathogenicity has been proven, they can affect the brain and bones, cause kidney disease or exacerbate it, and be hazardous for heart patients who take aspirin daily. These salts also produce a large amount of sludge, and sludge treatment is very costly. The high demand for ferrous sulfate and the need for high alkalinity, high oxygen content of the water for the use of ferric sulfate and ferric chloride, as well as the low efficiency of these coagulants in the presence of high concentrations of organic matter and color, are some of the problems that some coagulant salts face. Another major problem is the need for a high dose of iron and aluminum salts to carry out the coagulation process. The high concentration of soluble iron as a result of the use of common coagulants may cause corrosion of upstream pipes and equipment, consequently affecting the overall capital and operating costs of water treatment [26]. Research shows that excessive use of coagulants also has a negative impact on the performance of seawater reverse osmosis (SWRO) membranes. Therefore, there is a need for alternative disinfectant/coagulant materials, not only to achieve higher quality of the treated water but also with non-toxic and environmentally friendly byproducts. The aim of recent research is to search for greener chemicals that produce less sludge and, as a result, reduce sludge treatment costs. In this regard, ferrate (Fe(VI)) with its very strong disinfecting, oxidizing, and coagulating properties has received considerable attention in many studies. Ferrate (Fe(VI)) is a powerful oxidizing agent over a wide range of pH, and under acidic conditions, it is potentially stronger than all the oxidants applicable in water and wastewater, including ozone. The hexavalent ferrate ion (Fe(VI)) as a very strong oxidant has the highest redox potential among all oxidizing agents and disinfectants for water and wastewater treatment [27]. Ferrate (Fe(VI)) is also a cheaper and less hazardous oxidant compared to other current oxidants. As a result of the oxidation by ferrate (Fe(VI)), the reaction product is trivalent iron. Trivalent iron in aqueous solutions forms insoluble ferric hydroxide particles, which are considered a strong coagulant. Therefore, coagulation and flocculation occur during the use of ferrate for the oxidation and disinfection of water pollutants. This means that ferrate (Fe(VI)) can have a dual function. The expected benefits from this combined effect, which is used in advanced water and wastewater treatment operations, include higher quality of the treated water and wastewater, lower capital and operating costs [27]. The use of potassium ferrate (K_2FeO_4), which is still predominantly in the laboratory research stage, is one of the creative and promising methods in wastewater treatment. Hexavalent ferrate salts like potassium ferrate (K_2FeO_4) are chemical reagents with a high redox potential. The redox potential of potassium ferrate under acidic conditions is 2.2 V, while the redox potential of ozone is 2.04 volts and chlorine are 1.4 V. Ferrate (VI) salts like potassium ferrate have even higher redox potentials than permanganate and dichromates. Analysis of the redox potentials of various oxidants shows that potassium ferrate has one of the highest redox potential values, which consequently is accompanied by its very strong oxidizing properties. Under suitable conditions, ferrate ions can even oxidize water, and the reaction products are hydrogen protons and oxygen. Potassium ferrate is completely stable in the solid form, but it decomposes upon dissolving in water and contact with oxygen. The potassium ferrate solution is unstable under acidic conditions but stable and storable under alkaline conditions [28]. Some of the applications of potassium ferrate include water disinfection, degradation of synthetic organic pollutants, oxidation of inorganic pollutants, removal of humic substances, water and wastewater treatment, and sludge treatment. Various studies have been conducted on the use of potassium ferrate (K_2FeO_4) as an oxidant in industrial wastewater treatment, as this method has high efficiency in pollutant degradation. For example, in the decomposition of endocrine-disrupting chemicals, which are a serious threat to human and animal health, the use of potassium ferrate under appropriate conditions (pH, temperature, time, and dose) allows for complete degradation or significant reduction of these chemicals in wastewater [29]. Ferrate is also used in the decomposition of surfactants, which are surface-active agents widely used in various aspects of life, including the economy, industry, agriculture, pharmaceuticals, and molecular medicine, as well as components of detergents, cleaners, foaming agents, emulsifiers, and in formulations of plant protection products containing herbicides. Surfactants have high persistence, and their decomposition in the natural environment occurs very slowly, but the decomposition of cetylpyridinium chloride (CPC) has been achieved with 98% pure potassium ferrate [30]. Studies have also reported that the coagulation activity of potassium ferrate works well under acidic conditions. This is because potassium ferrate is a strong oxidizing agent under acidic conditions with a redox potential ($E^\circ = +2.20$ V) that is potentially the strongest oxidant realistically applicable in water and wastewater treatment [31]. As mentioned earlier, one of the main sources of fluoride pollution is the wastewater from the steel industry. Therefore, it is expected that a large amount of wastewater will be generated from the steel industries in the country in the near future, which will require planning for the treatment of the polluted effluent from this industry, as the wastewater requires fluoride removal before discharge or reuse.

Conclusion and Recommendations

There are many methods for fluoride removal, but due to the large volume of industrial effluents, a method should be used that is economical and efficient considering the existing conditions in the industry. Coagulation methods can be a suitable choice due to their easy implementation, low cost, and removal of other pollutants, among which the use of iron ferrate has received attention due to the formation of non-toxic compounds.

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