REMOVAL OF BERYLLIUM (Be²⁺) FROM WATER SAMPLES BY SORPTION PROCESS: A REVIEW

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Beryllium (Be^{2+}) is an important industrial metal because of its unusual material properties: it is lighter than aluminium and six times stronger than steel. Beryllium is a strategic metal due to its low density combined with its strength, low neutron absorption, high melting point and high modulus of elasticity. Beryllium is often alloyed with other metals such as copper and is an important component of materials used in the aerospace, automotive, energy, defense, medical, and electronics industries. However, beryllium and its compounds are very toxic, especially to the lungs, skin, and eyes. Beryllium compounds are known carcinogens based on sufficient evidence of carcinogenicity in humans from human studies. Toxic effects of beryllium include immunotoxic, allergic, mutagenic, and carcinogenic effects. Mammalian tissues do not excrete it, so the effects are cumulative and can lead to death at high concentrations. Therefore, removal of Be^{2+} is important. In this review, the removal of Be^{2+} from water samples by sorption processes using different sorbents was summarized. The effects of process parameters on the removal of Be^{2+} have been summarized. The work discussed showed that ion exchange resins, various modified biosorbents metal oxides can be used for the removal of Be^{2+} . The results showed that the pH of the solution has an important effect on the removal rate. Sorption kinetics vary from 3 minutes to 48h. When the functional groups are on the surface of the sorbent, the sorption process is rapid. However, if the surface of the sorbent is covered with oxides such as magnetite, it takes longer to reach equilibrium. Published work shows that more than 99 % of Be^{2+} can be removed from solution.

Keywords: adsorption, beryllium, toxicity, biosorbent, ion exchanger, water treatment

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1. Introduction

Beryllium (Be) is a steel-grey, brittle metal with an atomic number of 4 and an atomic weight of 9.01. Its ground-state electronic structure is given as ls^22s^2 . It has an outer shell with two s electrons, characteristic of members of group IIA of the periodic table. Beryllium's position between lithium and boron in the periodic table explains its low density of 1.85 g/cm³. The density is the second lowest of all metals. Its melting point of 1551 K (1287 $^{\circ}$ C) is consistent with its position in the periodic table [1,2].

1.1. Beryllium in the environment

Beryllium is the 47th most abundant element in the earth's crust and is widely distributed in many types of rocks. Beryllium behaves like a lithophile element in that it readily combines with oxygen to form light beryllium oxide compounds, resulting in its enrichment in the Earth's crust rather than in its core. The concentration in the primitive mantle is estimated to be about 60 parts per billion (ppb) beryllium, while estimates of the concentration of beryllium in the upper continental crust range from 1.9 to 3.1 ppm beryllium. Beryllium occurs mainly in the form of the silicate mineral beryl,

 $Be_3Al_2[Si_6O_{18}]$. It is also present in many natural minerals, including bertrandite, $Be_4Si_2O_7(OH)_2$ [3,4]. The properties of some Beryllium minerals is listed in Table 1 [3,5].

Name	Formula	Colour	BeO (percent)	Transparency
Barylite	BaBe ₂ Si ₂ O ₇	Colourless,	16	Transparent-
		white, yellow		Translucent
Bertrandite	Be ₄ Si ₂ O ₇ (OH) ₂	Colourless,	42	Transparent
		white, pale-		
		yellow		
Beryl	Be ₃ Al ₂ Si ₆ O ₁₈	White, pale-	14	Transparent-
		yellow, Green		Translucent
Chrysobery	BeAl ₂ O ₄	Green, yellow,	20	Transparent-
		red		Translucent
Helvite	Mn4Be3Si3O12S	Brown, yellow,	14	Transparent-
		grey		Translucent
Phenakite	Be ₂ SiO ₄	Colourless,	45	Transparent
		white		

Table 1. Name.	formula and	properties of some	Beryllium minerals
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Beryllium is a metallic element found in about 50 minerals. These minerals occur mainly in pegmatites, granites, syenites, and occasionally in gneisses and mica schists. When they occur in heterogeneous pegmatites, the crystals attain a large size and a good to excellent hexagonal crystal form; such crystals occasionally reach a diameter of 0.66 to 1 m, a length of several meters, and a weight of several tons. Crystals growing in cavities and druses in the parent rock are often quite transparent and of gem quality [6].

1.2 Application of beryllium

Pure beryllium and its metal alloys (e.g., Cu, Al, Mg, Ni) are an indispensable material in the manufacture of products for the aerospace, automotive, energy, defense, medical, and electronics industries. Beryllium is one-third lighter than aluminum, making it one of the lowest density metals. It is also one of the stiffest with six times the stiffness of steel. It has a high heat absorption capacity and is dimensionally stable over a wide range of temperatures.

Beryllium is a strategic metal due to its low density combined with its strength, low neutron absorption, high melting point and high modulus of elasticity, which makes it attractive for electrical components, aerospace and nuclear energy. As a result, beryllium metal is used in aircraft disk brakes, X-ray transmission windows, nuclear reactor neutron reflectors, and heat shields.

Beryllium oxide is used in high-tech ceramics, electronic heat sinks, and crucibles. Beryllium alloys are used in electrical connectors and relays, computers, underwater cable housings, aircraft engine parts, and dental applications [7–9].

1.3 Toxicity of beryllium

Beryllium exhibits a broad spectrum of toxic effects and is classified as a Group 1 carcinogen, meaning that beryllium and beryllium compounds are known carcinogens based on sufficient evidence of carcinogenicity in humans from human studies. Toxic effects of beryllium include immunotoxic, allergic, mutagenic, and carcinogenic effects. The primary health problem associated with beryllium is Beassociated lung disease (also called chronic beryllium disease or berylliosis); however, other health problems are also being considered. An important feature of beryllium toxicity is a considerable latency period, usually lasting up to 6-10 years. Because most of the attention related to beryllium has been devoted to occupational exposure, the mechanisms of beryllium toxicity are not yet clear [10–12].

Studies on the carcinogenicity of beryllium in experimental animals are available for the inhalation, intratracheal, oral, and parenteral routes of exposure. Inhalation exposure to beryllium (metal, ores, and sulfate compounds) resulted in a significant increase in lung cancer in rats and monkeys. These observations support the possible causal relationship found in the occupational studies.

Beryllium (metal, alloys and compounds) has also been shown to cause lung cancer in rats by intratracheal instillation and osteosarcoma in rabbits by intravenous and intramedullary injection [1].

WHO also reports that beryllium is rarely, if ever, found in drinking water in concentrations of concern. Therefore, it is not considered necessary to set a formal guideline value [1].

2. Removal of beryllium

Although both beryllium and beryllium compounds are classified as carcinogens, beryllium and its compounds are used in various industries. Because of the widespread use of beryllium and its known harmful effects on health and the environment, it is important to remove the potential risks. There are few studies dealing with the removal of beryllium ions from aqueous environments. In this review, the removal of beryllium from water samples by sorption methods has been summarized.

2.1. Removal of Be^{2+} by ion exchange resins

Ion exchangers are insoluble materials that carry reversibly bound ions. These ions can be exchanged stoichiometrically for other ions of the same sign. Ion exchange polymers, especially ion exchange resins, are crosslinked polymers carrying fixed functional groups or sites [13].

Othman et al. used a strongly acidic cation exchange resin, Amberlite IR-120, to remove Be^{2+} from aqueous solutions. The authors reported that the initial pH of the solution had an important effect on the removal rate and the optimum removal was achieved in a pH range of 3-3.5. In addition, increasing the solution temperature increased the sorption rate. Kinetic models showed that liquid film diffusion controlled the process in the early phase of adsorption, followed by matrix diffusion, which controlled the process in the final phase [14].

Demerdash et al. investigated the effect of flow rate and bed thickness on the removal of Be^{2+} in a fixed-bed reactor loaded with Amberlite IR-120. The authors reported that the sorption of Be^{2+} to the resin increased when the feed flow rate was decreased. The authors also found that as the bed height increased, the sorption of beryllium to Amberlite IR-120 increased at both fast and slow flow rates [15].

Ramesh et al. used inorganic ion exchangers, zeolite 4A, zeolite 13X and bentonite for the removal of Be^{2+} from aqueous solutions. The effect of sorbent dose, solution pH and contact time on the removal of Be^{2+} was studied. The optimum sorbent dose was found to be 5 g for a solution containing 3 mg-Be/L. The pH of the solution was given as another important parameter for the sorption of Be^{2+} . The removal rate of Be^{2+} was low at pH 1 and increasing the pH of the solution improved the removal rate and the optimum pH for Be^{2+} sorption was found to $be \ge 5$.

The presence of interfering ions, such as Ca, Mg and Fe, was also tested for the removal of Be²⁺ and the results showed that Mg had the greatest effect on the sorption of Be²⁺ on both zeolite 4A and bentonite. The decreasing order of effect of the counterions is Mg > Fe > Ca. However, all counterions showed less effect on bentonite than on zeolite 4A. Be- loaded zeolite 4A and benthonite were regenerated with different concentrations of NaCl. The regeneration rate of zeolite 4A is higher than that of benthonite and the optimum NaCl concentration was found to be 10 % NaCl for both ion exchangers [16].

Das and Pobi prepared a resin containing N-benzoylphenylhydroxylamine for the separation of Be^{2+} from other elements. Be^{2+} was significantly sorbed between pH 1.0 and 7.0. The loaded resin was regenerated with 2.0 M H₂SO₄ solution [17].

2.2. Biosorption

Biosorption can be defined as the removal of desired substances from an aqueous solution by biological material. The removal of contaminants occurs by various mechanisms such as absorption, adsorption, ion exchange, surface complexation and precipitation [18,19]. It may not be possible to find a sorbent that has such functional groups. Oxidation or various polymeric techniques have been used to attach functional groups to the surface of biosorbents.

Abu El-Soad et al. modified chitosan with sulfonic acid and used it to remove Be²⁺ from aqueous solutions. Chitosan was first cross-linked with glutaraldehyde to decrease the solubility of chitosan and later they used chlorosulfonic acid to introduce sulfonic acid groups into chitosan. The prepared sorbent was tested under different experimental conditions. The pH of the solution affected the removal rate of Be^{2+} and the optimum removal was achieved at pH 4. The sorption of Be²⁺ reached equilibrium in 40 minutes and the sorption kinetics obeyed the pseudoorder kinetic model. second The thermodynamic studies showed that the sorption of Be²⁺ was exothermic and spontaneous. The exhausted sorbent was regenerated with various solutions. It was found that the maximum desorption of beryllium ions from the sorbent loaded with Be with 1.5 M urea acidified with 0.6 M H₂SO₄ desorbed about 65% of beryllium ions within 30 minutes [20].

Abd El-Magied et al. prepared acetatecontaining chitosan and used it to remove Be^{2+} from aqueous solutions. The authors found that the removal of Be^{2+} was pH dependent and the maximum removal was achieved at pH 1 and pH 5. At pH values between 1-5, the removal rate decreased. The

authors explained that this difference was due to the presence of $Be(H_2O)_4^{2+}$ in acidic media and, at pH 5, to the dissociation of carboxylic acid and the electrostatic interaction of acetate groups with $[Be_3(OH)_3]^{3+}$ ions. The kinetic test showed that 80 % of the total uptake of Be^{2+} was achieved within 40 minutes and that the pseudo-second order model is most favourable for describing the biosorption process. The authors conclude that the Be^{2+} is exothermic sorption of and spontaneous. A 3 M H₂SO₄ solution can regenerate the sorbent with an efficiency of 86 % [21].

Acetate-containing cellulose was prepared by a two-step oxidation process and used to remove Be2+. NaIO4 was used to break the carbon-carbon bond of the glucopyranose ring to generate aldehyde groups, and NaClO₂ was used to oxidize the aldehyde groups to carboxylic acid groups. The authors reported that maximum removal was achieved at a pH of \geq 5. At a low pH, the removal rate was low, which was due to the conversion of the acetate groups to a molecular form so that there was no interaction between Be²⁺ and the functional groups. The sorption kinetics was very fast and reached equilibrium within 3 minutes. The authors also found that the sorption of Be^{2+} was exothermic and spontaneous. The sorbent loaded with Be can be regenerated with more than 99 % efficiency by 0.1 M HCl or H₂SO₄ solution [22].

Tokarcikova et al. prepared biochar from low-grade wheat and modified it with two different ratios of magnetite to remove Be^{2+} from aqueous solutions. The authors reported that the removal of Be^{2+} increased with increasing pH of the solution and the maximum sorption capacity was reached at pH = 6 for both sorbents. Sorption reached equilibrium in 6 h for MWBI (0.8 g FeSO₄·7H₂O was used to prepare the sorbent) and 48 h for MWBII (1.4 g FeSO₄·7H₂O was used to prepare the sorbent). The calculated kinetic parameters showed that the pseudo-second-order kinetic model fitted better to the experimental results. Regeneration tests showed that 0.01 M HCl or H₂SO₄ desorbed more than 95 % of Be²⁺ from the prepared sorbents [23].

2.3. Metal oxides and other composite sorbents

In addition to the ion exchanger and biosorbents, various types of organicinorganic composites or metal oxides have been used to remove Be²⁺.

Porschova and Parschova impregnated polymeric sorbents XAD4-A33 and XAD16-A33 with magnetite and used them for the removal of Be^{2+} from aqueous solutions. The prepared sorbents were placed in a column and the metal ion-containing solution was added to the column. The loaded sorbents were regenerated in a multistep process. First, it was rinsed with an alkaline salt solution (2 % NaOH and 2 % NaCl), then conditioned with demineralized water, and finally with demineralized CO₂-saturated water. 84 % of the Be from XAD4-A33 and 91 % of the Be from XAD16-A33 were desorbed [24].

Katsuta et al. tested Al_2O_3 , SiO_2 , TiO_2 , and CoO nanoparticles for the sorption of Be^{2+} from an aqueous solution. The authors found that the partition ratio of Be^{2+} between aqueous solution and nanoparticles increased with increasing pH. The authors also reported that the adsorption of Be^{2+} on the oxide nanoparticles was evaluated based on the surface complexation model, in which the hydroxyl groups on the surface of the oxide deprotonate and form complexes with Be^{2+} in water [25]. The capacities of the various sorbents are summarised in Table 2. The capacities of the sorbents varied from 1.5 to 45 mg/g. The variations in the capacity of the sorbent are related to the method of preparation, the functional groups available for Be^{2+} sorption, the size of the sorbent, and the nature of the functional groups. The available binding site during the preparation/modification of the sorbent.

Table 2. The comparison	of capacities	of different types of	of sorbents for the	<i>removal of</i> Be^{2+}
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Sorbent	Capacity	References
Zeolite 4A	3.3	[16]
Zeolite 13X	3.2	
Amberlite IR-120	11.9*	[14]
Aerobic granule	14.0	[26]
Ion-exchange resin with	31.2	[17]
Nbenzoylphenylhydroxylamine		
Sulfated chitosan	40.6	[20]
Magnetically Modified biochar	1.4	[23]
Acetate modified chitosan	45.0	[21]
Polystyreneazo3,4dihydroxybenzene	22.5	[27]

* obtained from the pseudo-first-order kinetic model at 31 °C

3. Effect of process parameters on sorption performance

As summarized various process parameters have been tested on Be^{2+} removal.

3.1 Sorbent dose

The removal of Be^{2+} increased with increasing sorbent dose, which can be attributed to the increase in available functional groups with increasing sorbent dose.

3.2 Solution pH

The pH of the solution affects the rate of sorption in different ways. If the functional groups are weak acid functional groups such as acetate, at an acidic pH or at a pH < pKa of a weak acid, the functional groups convert to their molecular form and the interaction between Be^{2+} and the functional groups does not occur, decreasing the removal efficiency

of the sorbent. Another parameter is the precipitation of Be^{2+} as $Be(OH)_2$. The working pH range should be lower than the precipitation pH. In addition, when metal oxides are used for the removal of Be^{2+} , the point of zero charge (pzc) of the metal oxide should be found and the pH of the solution should be adjusted to pH values at which the surface of the oxide becomes negative.

3.3 Temperature

The temperature of a solution has two main effects on adsorption. First, the rate of sorption generally increases at higher temperatures. This is primarily due to the increased rate of diffusion of the sorbate through the solution to the sorbent. Since solubility and adsorption rate are inversely related and temperature affects solubility, it also affects the extent of sorption or the capacity of the sorbent for sorbate [28]. The sorption of Be^{2+} is normally exothermic; therefore, the extent of sorption generally increases with decreasing temperature.

4. Conclusions and future work

The removal of Be^{2+} from water samples sorption by methods was investigated. The work discussed showed that ion exchange resins, various modified biosorbents metal oxides can be used for the removal of Be^{2+} . The results showed that the pH of the solution has an important effect on the removal rate. At pH 1 or 2, the removal of Be²⁺ is low, and increasing the pH of the solution increases the removal rate, however, precipitation of Be2+ can be observed at higher pH values. The optimum removal is achieved at different pH ranges and is related to the speciation of Be^{2+} as well as the nature of the functional groups of the sorbent.

Sorption kinetics vary from 3 minutes to 48 h. When the functional groups are on the surface of the sorbent, the sorption process is rapid. However, if the surface of the sorbent is covered with oxides such as magnetite, it takes longer to reach equilibrium.

The sorbent loaded with Be can be regenerated with HCl, H_2SO_4 , NaCl, or ureacontaining H_2SO_4 solution. The regeneration efficiency varied from 65 % to 99 %, which can be attributed to the nature of the functional groups.

The presence of interfering ions such as Na^+ , K^+ , Ca^{2+} and Mg^{2+} , and other cations that may be present with Be should be investigated. The effect of the concentration of interfering ions on the removal rate should also be studied in detail.

After regeneration of the sorbent, a Be-rich waste is produced. The reuse of the

Be-rich solution or the disposal of this waste should be planned.

The disposal of the spent sorbent should also be investigated in future studies.

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ВИДАЛЕННЯ БЕРИЛІЮ (Be²⁺) ІЗ ПРОБ ВОДИ СОРБЦІЙНИМ МЕТОДОМ: ОГЛЯД

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Берилій (Be²⁺) є важливим промисловим металом завдяки своїм незвичайним властивостям: він легший за алюміній та в шість разів міцніший за сталь. Берилій є стратегічно важливим металом по причині його низької густини у поєднанні з міцністю, низьким поглинанням нейтронів, високою температурою плавлення і високим модулем пружності. Берилій часто сплавляють з іншими металами, такими як мідь, він є важливим компонентом матеріалів, шо використовуються в аерокосмічній, автомобільній, енергетичній, оборонній, медичній та електронній промисловості. Однак берилій та його сполуки дуже токсичні, особливо для легень, шкіри та очей. Канцерогенний вплив сполук берилію по відношенню до людського органіму є давно доведеною і добре відомою. Токсичні ефекти берилію включають імунотоксичну, алергічну, мутагенну та канцерогенну дії. Тканини ссавців не пристосовані до виведення сполук берилію, тому спостерігається кумулятивний ефект, і, як наслідок, за високих концентраціяй це може призвести до смерті. Тому видалення Be²⁺ має велике значення. У даному огляді узагальнено дані шодо видалення Be^{2+} із водних розчинів з використанням різних сорбентів. В статті узагальнено вплив параметрів процесу на видалення Be²⁺. Встановлено, що для видалення Be²⁺ можна використовувати іонообмінні смоли, різні модифіковані біосорбенти, оксиди металів. Результати показали, що рН розчину має значний вплив на швидкість видалення. Кінетика сорбції змінюється від 3 хвилин до 48 годин. Коли функціональні групи знаходяться на поверхні сорбенту, процес сорбції відбувається швидко, однак, якщо поверхня сорбенту покрита такими оксидами, як магнетит, для досягнення рівноваги потрібно більше часу. Згідно опублікованих робіт, з розчину можна видалити більше ніж 99 % Be²⁺.

Ключові слова: адсорбція, берилій, біосорбент, іонообмінник, водопідготовка