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Performance of removing aqueous contaminant by zirconium based adsorbents: a critical review

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The studies on materials for decontamination in aqueous solutions have increasingly received greater attentions. Such contaminants as heavy metals, arsenic, fluoride and phosphate are harmful to humans and aqueous species due to higher toxicity. Zirconium based adsorbents have become more attractive due to outstanding performance in decontamination. This article provides a comprehensive review of the performance and mechanisms of five types adsorbents: zirconium (hydro)oxides, zirconium hydrogen sulfate, zirconium based multiple metal typed adsorbents and zirconium impregnated complexes. The pseudo-first order and pseudo-second order equations and the intraparticle diffusion model can be applied in describing the adsorption kinetics, while Langmuir and Freundlich equations are the most commonly used adsorption isotherms. The important mechanisms for uptake of contaminants are: ligand exchange between adsorbate and adsorbent, surface complexation formation, and Lewis acid–base and electrostatic interactions. A series of successful studies demonstrate that the adsorbents are promising for removing aqueous contaminants.

KEYWORDS
zirconium, adsorption, heavy metal, anions, ligand exchange

1 Introduction

Zirconium is a metallic element with the atomic number of 40. Its elementary substance is greyish-white and soft with ideal ductility. It is a malleable metal which is solid at room temperature, and would become hard and fragile under lower purities (Kloprogge et al., 2020). Zirconium can form various compounds and be used as promising engineering materials in many industrial processes, such as manufacturing of photoflash bulbs, surgical equipment, and tanning of leather (Lee et al., 1988; Akhtar et al., 2008). In recent years, zirconium-based materials have widely been studied for their applications in water treatment. Some of them have great potentials to be used as adsorbents in water purification due to the promising performances.

In recent studies, zirconium-based materials have attracted more attentions due to their higher binding capacities towards important anionic and cationic contaminants, such as copper, lead, arsenic, fluoride and phosphate (Wei et al., 2011; Chowdhury et al., 2019; Mukherjee and Singh, 2020; Khatun et al., 2022; Helleckes et al., 2023). Heavy metals are

TABLE 1 Effects of toxic heavy metals and anions on health and MCL (US Environmental Protection Agency, 2012).

Contaminants	Toxicity and effect	MCL (mg/L)
Arsenic	skin manifestations, visceral cancers, vascular disease	0.01
Lead	fetal brain damage; kidney, circulatory system, and nervous system diseases	0.015
Cadmium	kidney damage, renal disorder, human carcinogen	0.005
Chromium	headache, diarrhea, nausea, vomiting, carcinogenic	0.1
Copper	liver damage, renal disorder, human carcinogen	1.3
Mercury	kidney damage	0.002
Fluoride	bone disease, pain and tenderness of the bones, mottled teeth	4.0

harmful to humans and aqueous species as they are highly toxic and non-degradable. Such pollution has become a great global environmental thrust, as they can enter the human body through several pathways and can cause irreversible damages to human organs. Discharge of too much heavy metals can destroy the ecological balance. Fluoride contamination in groundwater is a serious environmental problem as excessive intake would cause skeletal fluorosis to humans. Arsenic contamination can cause several diseases, such as liver cancer. As a result, the World Health Organization (WHO), U.S. Environmental Protection Agency (USEPA) and many governmental environmental protection agencies have set the maximum contaminant levels (MCLs) for the heavy metals, arsenic, and fluoride in drinking water and the effluent of wastewater. The MCL of main heavy metals and fluoride are shown in Table 1.

The main concern of excess phosphorous in environment is the risk of eutrophication, since too much phosphorous can accelerate biological growth in waters, leading to the destruction of aquatic ecological system. The regulated phosphorus concentration is set as 0.02 mg/L.

Adsorption is one of better technologies for water and wastewater treatment due to its low cost, high efficiency, and easy handling (Bavi et al., 2023; Filho et al. 2023; Scanlon et al., 2023; Waghmare et al., 2023; Yang et al., 2023). A number of studies were carried out on zirconium-based adsorbents. The typical ones are zirconium oxide/hydroxide based adsorbents, zirconium based hydrogen sulfates, zirconium based multiple metal oxide, zirconium ion impregnated materials and zirconium phosphate adsorbent. Compared with traditional adsorbents such as activated carbon and ferric oxides, zirconium-based adsorbents have advantages like higher adsorption capacity, and ease in operation and regeneration.

In this article, the adsorption performances and mechanisms of different types of zirconium-based adsorbents are reviewed and discussed in details. It is anticipated that it would provide a summary of current scientific and technological studies for scientists and engineers in fields of environmental engineering. The review work will be useful for future development of innovative and highly effective adsorbent(s) for water and wastewater treatment and resource recovery.

2 Zirconium based Adsorbents

Generally, Zirconium based Adsorbents can be classified into five types, including Zirconium (hydro)oxide, Zirconium hydrogen sulfate, Zirconium based multiple-metal adsorbents, Zirconium impregnated complex and Zirconium phosphate. Their synthesis procedures are given in Table 2.

2.1 Zirconium oxide and hydroxide adsorbents

Zirconium oxide/hydroxide is the most traditional zirconium adsorbent owning many ideal properties like chemical stability and unique surface chemistry. Their performance in removal of mercury, chromium, phosphate and fluoride has been extensively studied, and some of the key results are summarized in Table 3.

Zirconium oxide (ZrO_2) (called as zirconia) is a white crystalline oxide form of zirconium. It is the most natural form of zirconium which can be found in mineral baddeleyite with a monoclinic crystalline structure. Zirconium hydroxide, known as hydrous zirconia, has an uncertain chemical formula which is mostly described as $ZrO_2.nH_2O$. It is also written in the form of common hydroxide description as $Zr(OH)_4$. The hydrous zirconia can be prepared facially by precipitation method. Zirconium oxychloride is first diluted in HCl solution, and then ammonia is added dropwise with constant stirring at room temperature until precipitate produces. The hydrous zirconia can be obtained after the precipitate is filtered, washed and dried (Rashad et al., 2018). On the other hand, the zirconium oxide can be obtained by heating the zirconium hydroxide for the removal of chemically bound water.

Mesoporous, micro or even nano sized zirconium oxides with large surface area and specific functionality are synthesized. The structure and morphology of zirconium oxides depends on different synthesis routes. Typically, they have high porous structure and high surface area (Ho, 1982; Saridag et al., 2013; Kwaśny and Balcerzak, 2017). The nano-sized zirconium oxide can be produced by the hydrothermal reaction; zirconium hydroxide can be heated at 200°C for 8 h for the production. This hydrothermal process at high temperatures/pressures can convert the hydroxide into the oxide ($Zr(OH)_4 \rightarrow ZrO_2 + 2H_2O$) (Yoshimura and Sōmiya, 1999; Lupo et al., 2004).

The adsorption capacities of several types of zirconium (hydro) oxides are summarized in Table 3. Generally speaking, zirconium oxide adsorbents have higher adsorption efficiency for anions (e.g., fluoride and phosphate). The zirconium (hydro)oxides with smaller sizes have higher adsorption capacity due to larger specific surface

TABLE 2 Summarization of synthesis methods of Zirconium based adsorbents.

Adsorbent	Synthesis procedure	Ref.
Zirconium hydroxide	Precipitation	Peterson et al. (2010), Cheng and Chuah (2020)
Zirconium oxide	Precipitation and heating	Rashad et al. (2018)
	Hydrothermal	Yoshimura and Sōmiya (1999), Lupo et al. (2004)
Zirconium hydrogen sulfate	Coprecipitation	Ma et al. (2011a), He et al. (2014)
Zirconium based multiple-metal adsorbents	Simultaneous oxidation and coprecipitation	Sonal and Mishra (2021)
	Co-precipitation	Ogata et al. (2020)
Zirconium impregnated complex	Chemical combination	Singh et al. (2020), Liu et al. (2021)
Zirconium phosphate	Precipitation	Cheng and Chuah (2020)

areas. The hydrous zirconium oxide has better adsorption capacity for fluoride than zirconium oxide (124 mg/g or 6.53 mmol/g); the adsorption of phosphate by zirconia-functionalized graphite oxide was as high as 131.6 mg/g.

2.2 Zirconium based hydrogen sulfate nanoparticles

Apart from the zirconium (hydro)oxide particles, newly zirconium based hydrogen sulfate nanoparticles were successfully synthesized; the thermal gravimetric and elemental analyses indicated that its molecular formula was $Zr_2(OH)_6SO_4 \cdot 3H_2O$ (Ma Y. et al., 2011; He et al., 2014). This nanoparticle was prepared by coprecipitation method by mixing the zirconium oxychloride solution and sulfate acid solution. This nano-sized zirconium particle has unique physical and chemical properties. The shape of the adsorbent is irregular, which indicates it is not crystalline. The size of the sorbent ranges from 60 to 90 nm when it is well dispersed in solution (Ma Y. et al., 2011). Unsaturated electron orbits in the nano-sized particles make it more efficient to combine other atoms. Additionally, the extremely large specific area of the nanoparticles promises their excellent performance on pollutant removal. Compared with the traditional nano-sized zirconium (hydro)oxide adsorbents, this nanoparticle has better performance for arsenic and fluoride with adsorption capacity of 256.4 and 78.6 mg/g, respectively. Most uptake occurred in the first 3–6 h and the optimal pH was 2.5–3.5.

The adsorbent was incorporated in the adsorptive hollow fiber membrane for water treatment. It overcame the limitation of the nanoparticles and demonstrated effectiveness for fluoride and arsenic removal. The working mechanism was the ion exchange between arsenic ion and hydrogen sulfate ions (Wiechert et al., 2023). Through membrane filtration, the anionic contaminants can be treated by just one step operation and no sludge is generated.

Various zirconium modified materials were fabricated for arsenic removal from water, such as zirconium modified pomegranate peel, zirconium oxide immobilized alginate beads and zirconium-chitosan modified spherical sodium alginate composite. In such materials, zirconium ions are grafted onto support materials, which are easy to collect after adsorption process. In recent years, some metal-organic frameworks (MOFs) have been applied for arsenic removal from water. The UiO-66 MOF was first studied for arsenate removal from water. The adsorption capacity was as high as 303.34 mg/g, which was much higher than other zirconium-based adsorbents for arsenic removal (Wang Y. et al., 2015). The UiO-66(Zr)-derived t-zirconia was developed based on UiO-66, which shows very high adsorption capacity for both arsenate and arsenite (Qu et al., 2022).

2.3 Zirconium based multiple–metal typed adsorbents

In recent studies, the work on synthesis and applications of multiple-metal typed adsorbents (namely, Zr with low-cost metals) increasingly has more attention. The zirconium based multiple-metal adsorbents can not only have advantages of each single metal oxide/hydroxides but also show significant synergistic effects through each component (Zhang et al., 2013). Iron (Zeng, 2003), titanium (Bekkouche et al., 2004), manganese (Gadde and Laitinen, 1974), and aluminum (Davis and Gloor, 1981) were reportedly used. These had much higher adsorption capacities than traditional mono-metallic adsorbents due to newly created active sites by the incorporation of new metals. These metals (Zr and associated metals used in fabrication of adsorbents) may work as Lewis acidic centers and form a list of complexes with target compounds (e.g., arsenic) (Fukuzaki et al., 1996). Because of the high electro-negativity properties, the anions (e.g., arsenate, arsenite, fluoride and phosphate) have strong affinities towards multivalent metal ions (e.g., Al (III), Fe (III) and Zr (IV)) (Luo and Inoue, 2004), leading to much better adsorption performances.

In the multiple-metal typed adsorbents, the surface areas vary substantially with zirconium content. Crystallization generally results in crystal growth, which in turn leads to larger interparticle pores. The zirconium titanium oxides were mainly X-ray amorphous (Sizgek et al., 2008). The pure metal oxides contained a larger mean mesopore diameter and a wider distribution than the mixed oxides. This may be due to the agglomeration of nanoparticles. In the iron-zirconium binary oxide, both morphologically heterogeneous, amorphous aggregations and crystalline domains with well-defined lattice fringes exist. Two sets of well defined peaks were identified. One was assigned to goethite and the other to lepidocrocite. They are slightly diverted from the standard references. The shifting of the diffraction positions

TABLE 3 Adsorption performance of Zirconium hydro (oxide) adsorbents.

Adsorbent	Adsorbate	Maximum adsorption (mg/g)	Optimal pH	Ref.
Hydrous zirconium oxide	Fluoride	124	4.0	Dou et al. (2012)
Zirconium oxide	Fluoride	19	4.75	Blackwell and Carr (1991)
Hydrated zirconium oxide	Fluoride	66	4.0	Biswas et al. (2008)
Mesoporous zirconium oxide	Phosphate	29.71	3.0	Liu et al. (2008)
Hydrous zirconium oxide	Phosphate	53	2.0	Rodrigues et al. (2012)
Amorphous zirconium hydroxide	Phosphate	30	5.0	Chitrakar et al. (2006)
Zirconium mesostructured immobilized calcium alginate	Phosphate	67.99	2.1–7.2	Yeon et al. (2008)
Zirconia-functionalized graphite oxide	Phosphate	131.6	2.0	Zong et al. (2013)
Lanthanum carbonate modified microfibrous composite	Phosphate	38.5	--	Yang (2023)
Goethite	Phosphate	8.55	--	Zhong et al. (2007)
Hydrous zirconium oxide	Chromium	66	2.0	Rodrigues et al. (2010)
Zirconium oxide	Mercury	0.69	7.0	Rauf et al. (1989)
Hydrous zirconium oxide	Mercury	16.11	5.62	Mishra et al. (1996)
Carbon nanosheets coated zirconium oxide nanoplate	Zinc	606.06	8.0	Fouda-Mbanga et al. (2023)
Vermiculite-based nanoscale hydrated zirconium oxides	Nickel	90.21	8.0	Liu et al. (2019)
Zirconium oxide intercalated sodium montmorillonite scaffold	Chromium	64.57	3.0	Rathinam et al. (2021)
	Phosphate	52.46	6.0	
Amorphous zirconium oxide	Arsenate	32.4	7.0	Cui et al. (2012)
	Arsenite	83	7.0	
Zirconium based nanoparticle	Arsenate	256.4	2.5–3.5	Ma et al. (2011a)
Zirconium metal-organic framework UiO-66	Arsenate	303.34	2.0	Wang et al. (2015b)
Oxygen-rich poly-bisvanillonitrile embedded amorphous zirconium oxide	Arsenite	115	3.0–5.0	Seynnaeve et al. (2021)
	Arsenate	245	3.0–5.0	
Zirconium modified pomegranate peel	Arsenate	83.33	4.0	Poudel et al. (2021)
UiO-66(Zr)-derived t-zirconia	Arsenate	352.1	3.0	Qu et al. (2022)
	Arsenite	147.5	11.0	
Zirconium oxide immobilized alginate beads	Arsenate	28.5	2.0	Kwon et al. (2016)
	Arsenite	32.3	7.0	
Zirconium-chitosan modified spherical sodium alginate composite	Arsenate	76.78	3.0	Lou et al. (2021)
	Arsenite	43.19	3.0	

was likely due to the doping state of the aggregated nanoparticles of the components (Dou et al., 2018).

A number of studies was conducted on these adsorbents, especially for removal of fluoride, arsenic, natural organic matters (NOMs) and phosphate. They can be prepared by simultaneous oxidation and coprecipitation techniques, like zirconium–manganese binary hydrous oxide (Zhang et al., 2013), zirconium-iron oxide (Dou et al., 2011) and magnetic Fe-Zr binary oxide (Long et al., 2011). In some cases, the adsorbent consisting of several metals can be synthesized through the co-precipitation approach (Ogata et al., 2020). Typically, zirconium (IV) solution is mixed with a multiple-metal containing solution, then sodium hydroxide solution or ammonia solution is added into the mixture to produce the fine-sized adsorbents. The performances of adsorbents summarized in Table 4 are much better than the commercial adsorbents.

Due to the characteristic of the metallic species as Lewis acid during adsorption, the optimal pH of most zirconium based multiple metal oxide adsorbents is still in acid range, as electrostatic attraction plays an

important role during the pollutant removal process. It should be noted that after addition of different metals like titanium and manganese, the mixed metal oxide adsorbents can remove metal cations, like vanadate, uranium and strontium which could not be removed by single zirconium metal. Molar ratio between added metal(s) and Zr is important for adsorption performance. Hence, the optimization should be conducted to obtain better uptake.

2.4 Zirconium impregnated complex adsorbents

Hard Lewis metals such as zirconium, iron, aluminum, and lanthanum have higher affinities towards the anions through the ligand exchange mechanism (Awual et al., 2011). As a strong Lewis acid, zirconium ions can be combined with other support materials to form hybrid complexes to enhance the performance as shown in Table 5. The most common support material is carbon-typed materials, e.g., biochar (e.g., cashew nut shell carbon (Alagumuthu and Rajan, 2010), coconut shell carbon (Sathish et al., 2007), pea peel waste (Swain et al., 2012), peanut shell (Huang et al., 2022) sludge (Wang et al., 2018) and activated carbon (Schmidt et al., 2008).

The zirconium ions can combine with the carbon through chemical reactions with the unsaturated bonds on carbon surface. This combination remarkably generates larger surface area and improves the surface acidity and pore structure of carbon materials such as carbon nanotube (CNT) (Ntim and Mitra, 2012), which is beneficial for metal uptake as shown in Table 5. In activated carbon, the adsorption sites for metal can be divided into two major groups: hydrophobic surfaces containing the graphene layers and hydrophilic surface comprising of oxygen functional groups (Sathish et al., 2007). When combining with zirconium, there strong interaction and synergistic effect caused by the motion of the free electrons between the support and the metal further improve the adsorption capacity (Yang Y. et al., 2023; Yang et al., 2024). Different types of activated carbon contain different adsorption sites, making their affinity various towards different contaminants.

Zirconium can be combined with ion exchange resin(s) to form effective adsorbents like Zr-loaded lysine diabetics acid resin (Balaji et al., 2005), zirconium loaded EDTA polymer resin (Suzuki et al., 2000), and zirconium (IV) complexes of the chelating resin (Tanaka et al., 2002). These resins contain groups like EDTA, phosphoric acid, hydroxyl and epoxy groups, which show much higher affinities to metal ions. Reported support materials include orange waste gel (Biswas et al., 2008), collagen fiber (Liao and Shi, 2005), fibrous adsorbent (Awual et al., 2011), zeolite (Xie et al., 2017; Ma et al., 2019), montmorillonite (Ma Y. et al., 2011; Zou et al., 2020), and bead (Qiusheng et al., 2015; Qing et al., 2022). Due to their abundant functional groups, they can be great support for zirconium, leading to the better uptake as shown in Table 5.

2.5 Zirconium phosphate adsorbents

Zirconium based adsorbents are widely used for phosphate removal, leading to formation of zirconium phosphate. It has low solubility in water and can further be used as an adsorbent/cation exchanger for heavy metals (e.g., lead, copper, cadmium and zinc). In practical applications, the zirconium phosphate adsorbent can be prepared by adding phosphate acid into zirconium solution (Azeroual et al., 2003; Gorbunoff, 1984; Nawrocki et al., 1993).

Similar to zirconium (hydro)oxide, zirconium phosphate can be developed into small sized particles which have larger surface area and specific functionality. The composite is usually grafted onto other engineering materials for column filtration study and for better industrial applications. Zirconium phosphate immobilized silica gel, α-zirconium phosphate crystals resin, and polymer-based zirconium phosphate materials are developed for lead and copper removal. The zirconium phosphate materials are summarized in Table 6.

As shown in Table 6, the adsorption capacities of zirconium phosphate for lead are different. The adsorption was strongly relative to the adsorbent structure. Normal crystalline zirconium phosphate adsorbent has an adsorption capacity of 21 mg/g, while that by the nano-sized one can be as high as 672.5 mg/g. Other heavy metals, like zinc, copper, cadmium, chromium and strontium can well be removed by the adsorbents.

3 Adsorption kinetic models

Adsorption kinetic study is critical in evaluating performance of an adsorbent. It plays an important role in selection and design of adsorption reactors. The commonly used kinetics models for adsorption process of zirconium adsorbents are given in Table 7.

To better understand the modes of adsorption kinetics, several models are used, e.g., pseudo-first order, pseudo-second order, double constant, and Elovich models (equations); among them, pseudo-first order and pseudo-second order equations are commonly used due to their simplicity (Ali et al., 2023):

$$\ln\left(q_e - q_t\right) = \ln q_e - k_1 t \qquad (1)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad (2)$$

where q_e and q_t are the adsorption capacities (mg/g) of the adsorbent at equilibrium and at time t (h), respectively; k_1 (h^{-1}) and k_2 (g/mg h) are the pseudo-first order and pseudo-second order adsorption rate constants, respectively. The suitability of equation is evaluated by the values of correlation coefficient (r^2); the higher r^2 value means the more suitable model.

As shown, most adsorption processes follow the pseudo second order kinetic model. This suggests the importance of chemical adsorption reaction(s) (Yang et al., 2020; Yang et al., 2021b).

Unlike the pseudo-first and second order equations, the intraparticle diffusion model is based on theoretical considerations and provides mechanisms in adsorption kinetics. It is more meaningful and theoretically sound to use it to simulate uptake history.

The model is based on an assumption of "two-step mass transport mechanism" where adsorbate first transfers through the external liquid film from the bulk solution to the adsorbent surface,

TABLE 4 Adsorption performance of zirconium based multiple-metal adsorbents.

Adsorbent	Adsorbate	Maximum adsorption (mg/g)	Optimal pH	Ref.
Zirconium titanium oxides	Vanadate	24.96	pH = 10.5	Sizgek et al. (2008), Drisko et al. (2009)
Mesoporous Zirconium Titanium Oxides	Vanadate	13.2	pH > 9	Griffith et al. (2008)
Mesoporous Zirconium Titanium Oxides	Uranium	35.7	3.8	Sizgek et al. (2009)
Zirconium-iron oxide	Fluoride	26.54	3.5	Dou et al. (2011)
Iron(III)- zirconium(IV) Hybrid oxide	Fluoride	7.454	4.0–7.0	Biswas et al. (2008)
Granular zirconium-iron oxide	Fluoride	26.54	3.5	Awual et al. (2011)
Magnetic Fe-Zr binary oxide	Phosphate	17.87	3.0	Long et al. (2011)
Zirconium iron binary oxide	Phosphate	33.4	5.5	Ren et al. (2012)
Mixed hydrous oxides (Zr, Fe, Al)	Phosphate	22.9	4.0	Chubar et al. (2005)
Zirconium-aluminum hybrid adsorbent	Fluoride	65.07	5.0–9.0	Wu et al. (2018)
Nickel-aluminum-zirconium complex hydroxide	Phosphate	157.2	—	Ogata et al. (2020)
Iron-zirconium bimetal oxide	Hexavalent chromium	59.88	2.0	Wang et al. (2015a)
Zirconium-iron oxide nanoparticle	Phosphate	63.9	1.5	Zhang et al. (2017)
Calcium-alginate hydrogel-sphere encapsulated with Fe–Mn–Zr ternary metal composite	Copper	103.1	4.0	Yuan et al. (2023)
Fe–Mn–Zr ternary magnetic sorbent	Arsenic	81.3	2.0	Zou et al. (2022)
Activated charcoal coated zirconium-manganese nanocomposite	Arsenate	95.6	6.0	Yin et al. (2019)
	Arsenic	132.28	10.0	
Iron and zirconium modified luffa fibre	Arsenate	2.89	7.0	Nguyen et al. (2020)
Iron-zirconium microwave-assisted modification of small-pore zeolite	Arsenate	42.31	3.0–9.0	Abdellaoui et al. (2021)
La-Zr bimetallic MOFs	Arsenic	83.4	2.0	Han et al. (2022)
Bowknot-like Zr/La bimetallic organic frameworks	Arsenate	102	5.0	Kong et al. (2022)

and subsequently diffuses into the adsorbent before finally being adsorbed by functional groups. Depending upon the structures of adsorbents, the diffusion inside of adsorbent can be either surface diffusion or pore diffusion when specific surface area is either larger or smaller than 100 m³/g-adsorbent (Choy et al., 2004). The mathematical equations and corresponding initial and boundary conditions with the assumption of surface diffusion are expressed as follows (Chen, 2012):

$$D_s \frac{\partial}{\partial r} \left(r^2 \frac{\partial q}{\partial r} \right) = r^2 \frac{\partial q}{\partial r}, 0 \le r \le a_p, t \ge 0 \qquad (3)$$

$$q = 0, 0 \le r \le a_p, t < 0 \qquad (4)$$

$$\frac{\partial q}{\partial r} = 0, r = 0 \qquad (5)$$

$$D_s \rho_p \frac{\partial q}{\partial r} = k_f (C - C^*) \qquad (6)$$

where C and q are the concentrations of the adsorbate in bulk and in solid phases, respectively. C^* is the aqueous phase concentration at

the particle surface in equilibrium with the corresponding concentration in the solid phase q^*. D_s is the surface diffusivity within the particle, ρ_p is the particle density, r is radius distance measured from the center of particle, ∂_p is the particle radius, k_f is the external mass transfer coefficient. The porous diffusion model can be found in the literature (Chen, 2012).

4 Adsorption isotherms

Several adsorption isotherms are extensively used to evaluate the maximum adsorption capacity, predict the concentrations of treated effluent and estimate the dosage of adsorbent to be used. The distribution of targets (e.g., arsenic, fluoride and phosphorous) in the bulk solution and on the adsorbents can be described by one or even a few isotherm equations: Gaseous slip, Langmuir, Freundlich, Tempkin, Dubinin-Radushkevich and Brunauer-Emmet-Teller (BET) equations. Among them Langmuir and Freundlich isotherms given below are the most

TABLE 5 Adsorption performance of zirconium impregnated adsorbents.

Adsorbent	Adsorbate	Maximum adsorption (mg/g)	Optimal pH	Ref.
Zirconium impregnated cashew nut shell carbon	Fluoride	162	3.0	Alagumuthu and Rajan (2010)
Zirconium impregnated collagen fiber	Fluoride	41	5.5	Liao and Shi (2005)
Zirconium carbon hybrid sorbent	Fluoride	7.4	-	Velazquez-Jimenez et al. (2014)
Zirconium-modified-Na-attapulgite	Fluoride	24.55	4.13	Zhang et al. (2012)
Zirconium complexes of chelating resins	Fluoride	109	2.0–4.0	Tanaka et al. (2002)
Zirconium Impregnated coconut shell carbon	Fluoride	9.11	8.0	Sathish et al. (2007)
Hybrid sorbent of Zr(IV) ethylenediamine	Fluoride	68.25	7.0	Swain et al. (2012)
Zirconium Ion Modified MgAl-layered Double Hydroxides	Phosphate	17.7	-	Miyauchi et al. (2009)
Zirconium loaded orange waste gel	Phosphate	57	3.0	Biswas et al. (2008)
Zirconium-modified bentonite	Phosphate	40.2	4.0	Lin et al. (2018)
Surfactant modified zeolite/hydrous zirconium oxide	Phosphate	18.14	7.5–9.8	Xie et al. (2017)
Zirconium-pillared montmorillonite	Trivalent chromium	129.63	8.0	Ma et al. (2011a)
Zirconium-loaded Ca-montmorillonite	Phosphate	22.37	4.0–8.5	Zou et al. (2020)
Zirconium alginate beads	fiuoride	32.797	2.0	Zhou et al. (2014)
Sodium alginate/zirconium hydrogel	Phosphate	256.79	3.0	Qing et al. (2022)
Zirconium-Modified Activated Sludge	Phosphate	82.65	2.0	Wang et al. (2018)
Zirconium-modified biochar	Phosphate	58.93	2.0	Huang et al. (2022)
Lanthanum carbonate modified microfibrous composite	Phosphate	38.5	--	Yang (2023)

commonly used due to simplicity and accuracy (Chen, 2012; Ezzati, 2020).

$$q_e = \frac{q_{max} b C_e}{1 + b C_e} \qquad (7)$$

$$q_e = K_f C_e^{1/n} \qquad (8)$$

where, q_e (mg/g) and C_e (mg/L) are amount of adsorbate adsorbed and residual adsorbate concentration; q_{max} (mg/g) is theoretical maximum adsorption capacity of adsorbent; b (L/mg) is adsorption reaction constant related to affinity of binding sits; K_f is a constant for relative adsorption capacity, and higher K_f indicates higher maximum adsorption capacity; n is heterogeneity factor and a lower value indicates more heterogeneous surface.

Langmuir isotherm assumes that the adsorption takes place on a homogeneous surface and monolayer adsorption occurs on the surface. It can be used to evaluate the maximum adsorption capacity of an adsorbent (q_{max}). On the other hand, Freundlich isotherm is for multilayer adsorption process on heterogeneous surfaces.

Redlich-Peterson isotherm given below combines the characteristics of both Langmuir and Freundlich isotherms, and can describe adsorption with both uniform and non-uniform adsorption (Chen, 2012).

$$q_e = \frac{A C_e}{1 + B C_e^n} \qquad (9)$$

where A (L/g) and B ((L/mg)^n) are Redlich-Peterson model constants and n is Redlich-Peterson model exponent.

Table 7 provides the models for adsorption isotherm and constants from several reported studies. In general, there is no common rule on selection of suitable model(s). Based on the observation of uptake kinetics, it is widely accepted that the adsorption of metals and anions is due to chemical interactions/reactions. In the processes, both physical process and chemical reactions are involved. For example, the electrostatic attraction first occurred between the adsorbents and the phosphate, by which the later was attracted onto the surfaces of adsorbents; the surface complexation and ligand exchange subsequently occurred, leading to the uptake (Yang et al., 2020; Yang et al., 2021b).

The selection of equilibrium mode(s) is mainly dependent on properties of adsorbents (e.g., type, size and specific surface area) and adsorbates (e.g., type, pH, ionic strength, and concentration). It should be noted that these models can only describe adsorption of

TABLE 6 Adsorption performance of zirconium phosphate adsorbents.

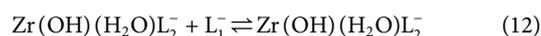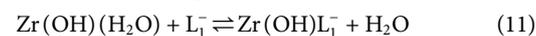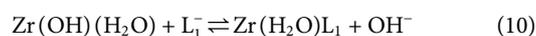| Adsorbent	Adsorbate	Maximum adsorption (mg/g)	Optimal pH	Ref.
Amorphous zirconium phosphate	Lead	621	5.5	Pan et al. (2007a)
	Cadmium	190.6	5.5	
	Zinc	91.5	5.5	
Polymer-based zirconium phosphate	Lead	410	3.5	Pan et al. (2007b)
Amorphous zirconium phosphate	Lead	155	5.5	Jiang et al. (2008)
Crystalline zirconium phosphates	Lead	21	6.0	Jiang et al. (2008)
Polysulfone capsules containing zirconium phosphate	Lead	149.7	5.75	Ma et al. (2011a)
Polymer-supported zirconium phosphate	Lead	556	6.0	Pan et al. (2006)
Zirconium phosphate nanoparticles	Lead	672.5	-	Zhang et al. (2011)
Silicon-based zirconium phosphate	Strontium	100.77	6.0	Jiao et al. (2021)
Polypyrrole modified zirconium (IV) phosphate nanocomposite	Hexavalent chromium	62.5	2.0	Behera et al. (2022)

TABLE 7 List of models for kinetics and isotherm study.

Adsorbents	Adsorbate	Kinetics model	Isotherms	Ref.
Zirconium oxide	Fluoride	Pseudo second order	Freundlich	Dou et al. (2012)
Hydrous zirconium oxide	Phosphate	Pseudo second order	Langmuir	Rodrigues et al. (2012)
Zirconium hydrogen sulfate nanoparticle	Fluoride	Pseudo second order	Langmuir	Zhang et al. (2013)
		Intraparticle diffusion		
Zirconium iron hybrid oxide	Fluoride	Pseudo second order	Redlich Peterson	Suzuki et al. (2000)
Zirconium-iron oxide	Fluoride	Intraparticle diffusion	Freundlich	Huang et al. (2022)
Zirconium impregnated cashew nut shell carbon	Fluoride	Pseudo second order	Redlich Peterson	Alagumuthu and Rajan (2010)
Zirconium loaded orange waste gel	Phosphate	Pseudo second order	Langmuir	Biswas et al. (2008)
Polysulfone capsules containing zirconium phosphate	Lead	Intraparticle diffusion	Langmuir	Ma et al. (2011b)

single-component contaminant (e.g., arsenic or fluoride) and fail to do in cases of multiple-component ones (e.g., co-existence of arsenic and fluoride, or that of copper and zinc). Therefore, such theoretical models as surface complex formation (SCF) models and ion exchange-SCF model can be used (Lim et al., 2008; Chen, 2012).

5 Adsorption mechanisms

Zirconium is a typical Lewis acid and its electronic orbit is unsaturated. It has available coordination sites to accept electron pairs. In order to satisfy the coordination sites of the zirconium ions, electron pair donors could be coordinated to the available sites. In aqueous solutions, these donors would be water molecules, hydroxide ions and other Lewis bases (Blackwell and Carr, 1991; Blackwell and Carr, 1992). For example, fluoride and phosphate ions can provide electron pairs. Target contaminants are therefore strongly coordinated with zirconium ions and better uptake is achieved (Blackwell and

Carr, 1992). The following chemical reactions can be used to describe the interaction between various species and different active sites on zirconium adsorbent surface (Blesa et al., 1988):

$$Zr(OH)(H_2O) + L_1^- \rightleftharpoons Zr(H_2O)L_1 + OH^- \quad (10)$$

$$Zr(OH)(H_2O) + L_1^- \rightleftharpoons Zr(OH)L_1^- + H_2O \quad (11)$$

$$Zr(OH)(H_2O)L_2^- + L_1^- \rightleftharpoons Zr(OH)(H_2O)L_2^- \quad (12)$$

where L_1 and L_2 represent Lewis base in the solution and on the zirconium ions, respectively. The zirconium ion combines with H_2O and OH^- ligand in aqueous solution.

Equations (10)–(12) describe the structural change in the surface sites for the zirconium ion impregnated complex adsorbents or zirconium (hydro) oxide. The ligand exchange happens between H_2O/OH^- with L_1. For the zirconium based hydrogen sulfate nanoparticles or other zirconium combined adsorbent, Eq. 12 can be used to describe the structural change of the surface sites, on which the ligand exchange happens between L_1 and L_2.
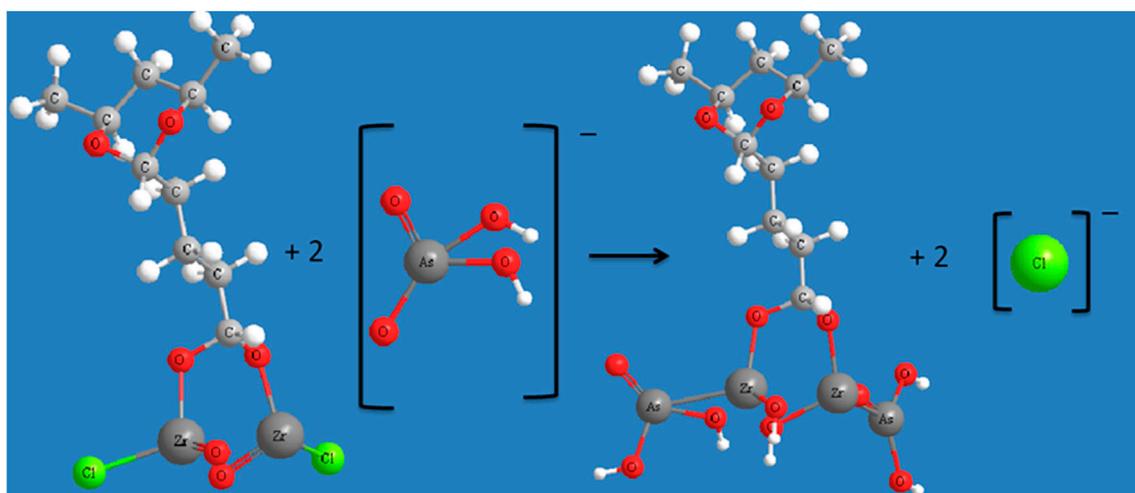
FIGURE 1
Illustration of ligand exchange between arsenate and zirconium based adsorbent (Zhao et al., 2023).
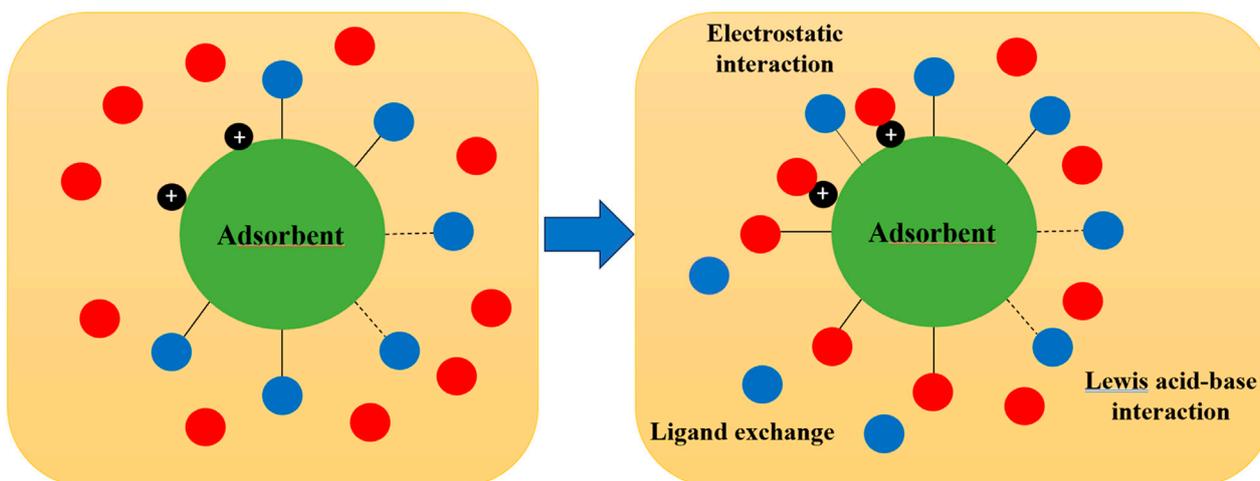

FIGURE 2
Mechanisms for removal of aqueous contaminants by zirconium based adsorbents.

Single-component zirconium metal adsorbent is usually less efficient in uptake of contaminants due to the similarity in Lewis acid characteristics between adsorbent and adsorbate. When the zirconium is incorporated with other materials such as metals and anions (e.g., zirconium phosphate), the roles of ion exchange and electrostatic attraction become more significant, leading to better uptake (Zhao et al., 2016; McGowan et al., 2023).

The zirconium based adsorbents may have exchangeable anions (e.g., SO_4^{2-}, and Cl^-) or cations (e.g., H^+). They can exchange of the targe compounds based on the characteristics of charges. Such ligand exchange commonly occurs in uptake process. Various evidences demonstrate that ion exchange plays a key role in uptake of contaminants.

The zirconium based nanoparticle can treat arsenic and fluoride with excellent performace due to the enriched content of sulfate ions in the adsorbent (Ma Y. et al., 2011; He

et al., 2014). The arsenic adsorption was studied by using Zirconium/polyvinyl alcohol modified flat-sheet polyvinylidene fluoride membrane (Zhao et al., 2016). Through the batch experiment and the spectrum analysis, it was found that the ligand exchange between chloride and arsenate lead to the arsenate uptake as illustrated in Figure 1.

The point of zero charge (PZC) of adsorbent is an important factor in adsorption of either anions or cations. The PZC not only determines the surface charge property of adsorbent in different solutions, but also affects the chemical reactions and the electrostatic interaction between the ions and the adsorbent (Yang et al., 2021a; Yang et al., 2022).

Adsorbent surface becomes positively charged at solution pH < PZC, while it is negatively charged at pH > PZC. Electric attraction contributes to the pollutant as this interaction has some similarity with ionic bonds, and the ion will be attracted to the oppositely

charged adsorbent surface. On the other hand, electrostatic repulsion occurs when the adsorbent surface is charged the same as the ions and causes an adverse effect on the performance.

The important mechanisms are illustrated in Figure 2 and summarized as follows: ligand exchange between adsorbate and adsorbent, surface complexation with electron donation from adsorbate, and Lewis acid–base and electrostatic interactions. These were confirmed through energy dispersive X-ray spectroscopy (EDX), X-ray photoelectron spectroscopy (XPS), Fourier transform infrared spectroscopy (FTIR), nuclear magnetic resonance (NMR) and zeta potential analyses in a larger number of published research articles (Wei et al., 2011; Sonal and Mishra, 2021; Yang L. et al., 2023; Zhang et al., 2023).

6 Conclusion

The findings from the studies on zirconium based adsorbents for removal of cationic heavy metals, and anionic contaminants are discussed and summarized. The adsorbents with better performances are zirconium oxide/hydroxide, zirconium hydrogen sulfate nanoparticles, zirconium based multiple metal oxide, zirconium ion impregnated complex and zirconium phosphate. The adsorption capacities and kinetics for aqueous contaminants are much better than many reported adsorbents; the adsorption capacities are as high as several micromoles per gram of adsorbent and uptake can be completed within several hours. The Lewis acid-base interaction and ligand exchange are main mechanisms for the better uptake. The adsorption isotherm can be well described by either Langmuir and Freundlich equation, while the intraparticle diffusion model as well as the empirical equations (pseudo-first and second order equations) can be used to describe adsorption history. This review work clearly indicates that zirconium-typed adsorbents are promising for treatment of heavy metal and anionic contaminants and the recovery.

Author contributions

DZ: Writing–original draft, Writing–review and editing. YY: Conceptualization, Writing–review and editing. JC: Conceptualization, Writing–review and editing, Funding acquisition, Supervision.

Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

The author JC declared that he was an editorial board member of Frontiers at the time of submission. This had no impact on the peer review process and the final decision.

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Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fceng.2024.1282076/full#supplementary-material

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