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Adsorption of various inorganic and organic pollutants by natural and synthetic zeolites: A critical review

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ABSTRACT

Zeolites are microporous crystalline aluminosilicates with high surface area and uniform pore size. Natural and synthetic zeolites have been used to adsorb organic and inorganic compounds in aqueous media due to thier particular physicochemical properties and the low cost of the process. The flexibility of zeolites to remove pollutants from water, such as dyes, heavy metal ions, and phenolic compounds, is discussed in this document in the context of contemporary research. This review briefly consolidates the currently available literature to comprehend the structure of zeolites and their synthesized by hydrothermal method. Later, this manuscript is present different parameters to study the adsorption mechanisms of organic and inorganic contaminants using the zeolites. The main adsorption processes using zeolites as adsorbents include chelation, surface adsorption, natural processes, diffusion, electrostatic interaction and complexation. In addition, the research demonstrates that the dominant models in the isothermal and kinetic study of adsorption are the Langmuir and the pseudo-second-order models. We can assess the beneficial applicability of zeolite materials for real wastewater treatment in the future by comparing their adsorption capacities for removing harmful substances from water to those of other adsorbents and crude zeolites.

1. Introduction

The question of water treatment, conservation, and quality could be a universal issue. Pollutants discharged into springs, and aquatic systems have evolved considerably in quantity and quality(Bhaskar et al., 2020). More complex products enrich simple domestic discharges, and also the sewerage networks have very diverse characteristics since they now collect industrial, commercial and artisanal discharges (Akani and Nwankwo, 2018; Bhaskar et al., 2020; Ma et al., 2016a). Pollution, a fancy and taxing phenomenon of world development, could be a degradation of a natural environment by chemical substances and industrial wastes affecting, to varying degrees, all urban, industrial, and rural areas and, therefore, the aquatic environment. It impacts the soil, water, and air and may reach ditches, rivers, canals, marshes, lakes, the ocean, and groundwater (Rong et al., 2015). Among all the harmful and priority chemical pollutants, aromatic compounds, including substituted phenolic compounds (nitrophenols, chloroprenes…), are detected in leachates at alarming concentrations. Organic and inorganic compounds that cause unpleasant tastes and odors are detected in surface water, groundwater and wastewater. These are the highest ten micropollutants on the list of organic and inorganic compounds classified by the U.S. (Akani and Nwankwo, 2018; Ismagilov et al., 2007; Luis, 2006; Ma et al., 2016a; Sun et al., 2015). The main techniques include biological degradation, membrane filtration (El-Kordy et al., 2022b), advanced

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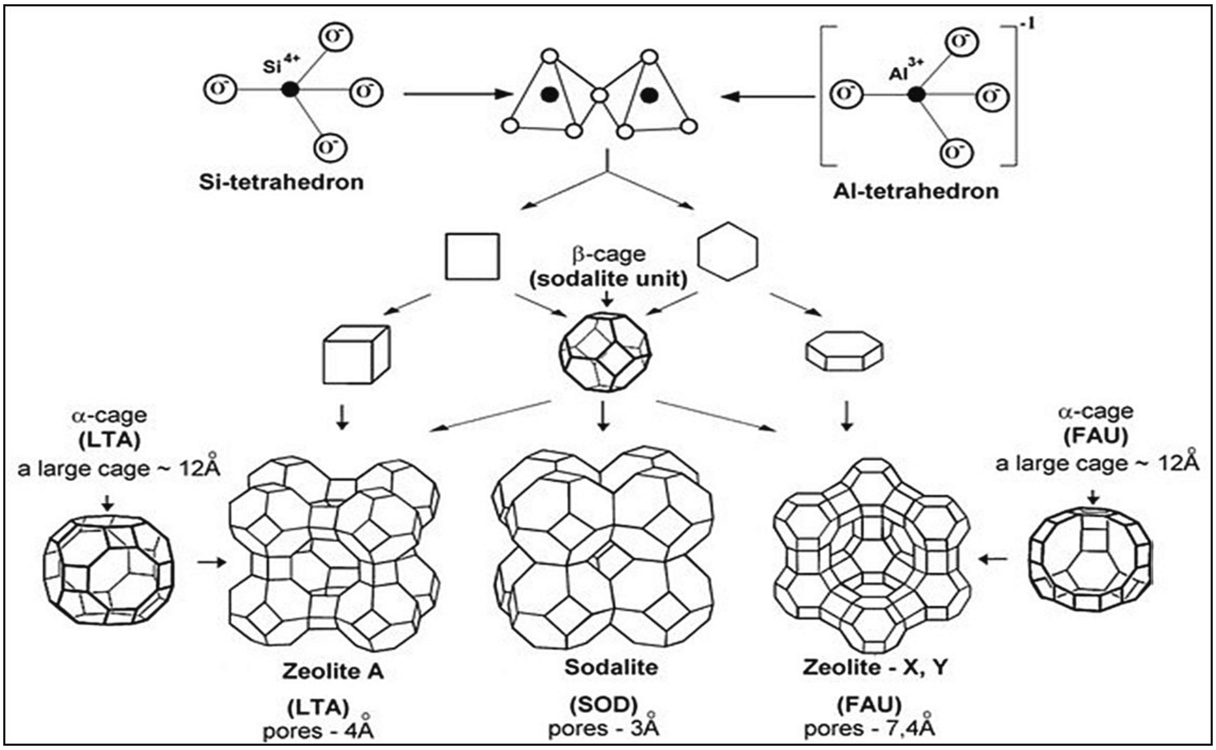

Fig. 1. Structural unit of zeolite, exemples zeolite A (LTA), sodalite (SOD), and faujasite (FAU) (Scientific, 2017).

oxidation, photocatalytic degradation, electrochemical oxidation, and particularly those supported adsorption phenomena (El-Kordy et al., 2022a). Of all the techniques listed, solid-state adsorption processes are highly effective in removing organic compounds from wastewater. Adsorption processes provide many advantages over other techniques, like low capital and operating costs, easy use, the potential use of naturally available solid supports, high efficiency, and simple apparatus in eliminating pollutants, even at trace levels(Abdellaoui et al., 2020; Abou Oualid et al., 2020; Afiqah et al., 2019; Ahmad et al., 2021; Eltaweil et al., 2020; Hosseini et al., 2021; Moosavi et al., 2020). Adsorption methods are widely used to separate organic and inorganic pollutants from domestic and industrial wastewater. Therefore, very important research efforts are devoted to developing varied alternative adsorbents with high sorption capacities to uptake pollutants from wastewater(Alshabib and Onaizi, 2019; Erkonak et al., 2008; Lin et al., 2020; Mandal and Das, 2019; Mirmohamadsadeghi et al., 2012; Petrie et al., 2015; Sun et al., 2015; Vuppala et al., 2019). Many types of adsorbents exist, the most common being zeolite. It is a superior adsorbent because of its large specific area, various surface functions, and porosity. Nevertheless, the disadvantage of zeolite is its high preparation and regeneration cost and, thus, the limit of its adsorption capacity. Nevertheless, recent data from the scientific literature show that zeolites have proven to be very effective in the field of liquid effluent treatment.

Nevertheless, the bibliographic data on the microstructure and the rendering of removal in environmental reactions remains insufficient. Therefore, this paper aims to provide preliminary knowledge on the technical feasibility of the zeolite adsorption process by evaluating the results of published studies. Several studies and reviews of zeolite adsorption have investigated the elimination of organic pollutants from aqueous solutions (Chaouati et al., 2013; Cheng et al., 2016; Kragović et al., 2019; Lin et al., 2020; Merrikhpour and Jalali, 2013; Na chat et al., 2022; Razavi et al., 2021), while others have focused on removing inorganic pollutants (Elhakim et al., 2021; Hailu et al., 2017; Ma et al., 2016b; Marszałek et al., 2022; Mehraban et al., 2007; Yang et al., 2019a). Accordingly, The present literature review focuses simultaneously on the adsorption of organic and inorganic pollutants on natural and synthesised zeolites.

The essay examines the essential structural characteristics of zeolites and their industrial uses. Isotherms, kinetics, mathematical models, and adsorption mechanisms typically used to describe processes are all covered in-depth. Studies on equilibrium, thermodynamics, and characterization are also covered. There is also a summary of how these models have been used in zeolite adsorption studies already performed. One of the strengths of this review is its thorough synthesis of recent research results on adsorption of various organic and inorganic pollutants using natural and synthetic zeolites. This makes it easier to identify the crucial variables that would improve adsorption efficiency. The effect of the best-performing parameters such as pH, initial concentration, zeolite properties, nature of the organic pollutants.

The first zeolite was identified and named by Axel F. Cronstedt in 1756. When he noticed that this particular sort of rock could hold onto water within its structure and release it when heated, he came up with the term “zeolite” from the Greek words “zeo” to boil and “lithos” to stone,which had previously been used in literature.(de Aquino et al., 2020; Kim et al., 2022a; Madhu et al., 2022; Ramezani Shabolaghi and Irani, 2022; Wang and Chen, 2021). Zeolites are members of the mineral family and are more precisely categorized as alumino-silicates having a distinct crystalline structure. Al, Si, and O are the three primary components of them. The arrangement of these components in tetrahedral SiO_4 and AlO_4 configurations creates various zeolite kinds. Four oxygen atoms are covalently linked to the center Al or Si atom. The formula TO_4 is frequently used to denote the core atom along with the oxygen atoms. The central atom is also known as the [T] atom(Alborzi et al., 2022; Feng et al., 2022; Li et al., 2022b; Wang and Chen, 2021).

The Structure Commission of the International Zeolite Association (SC-IZA) is responsible for assigning codes for the proper nomenclature of zeolite frameworks; IUPAC accepts these codes (Mccusker and Baerlocher, 2019). The three capital Roman letters that make up the nomenclature code define the topology of the atomic lattice T, regardless of its chemical makeup or symmetry. Take LTA, MOR, or FAU as examples. In addition, a hyphen, such as -CLO, -LIT, or -PAR, is inserted before the code in the scenario where there is a broken framework, one

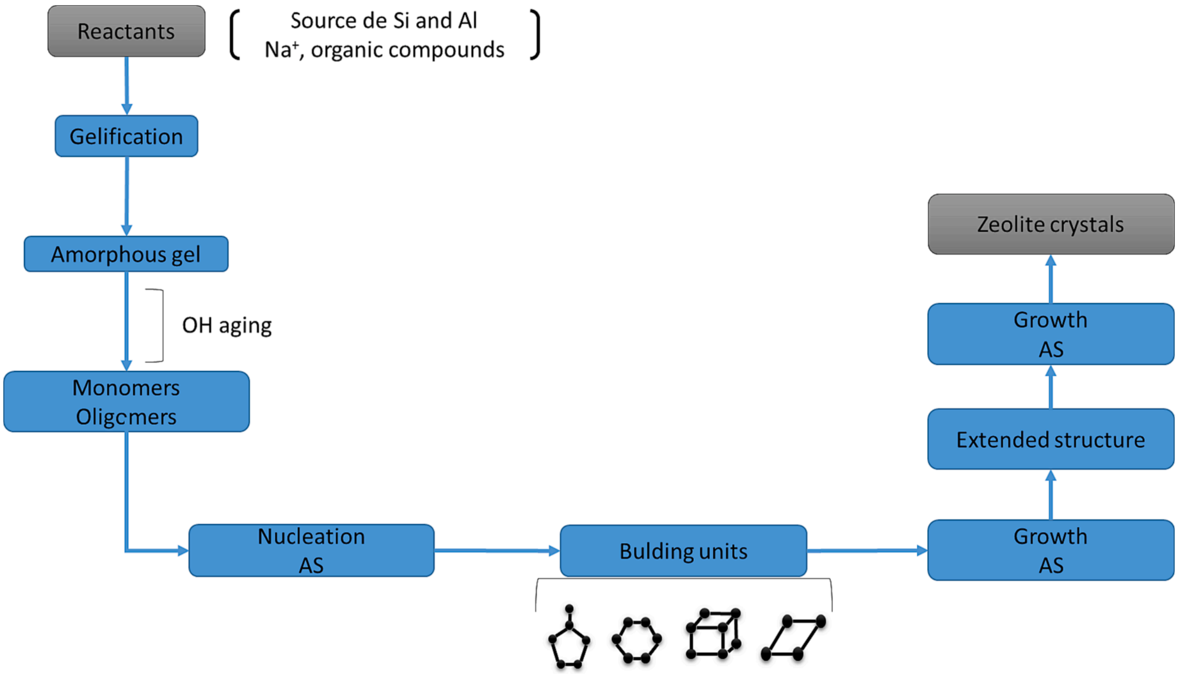

Fig. 2. Diagram of the hedrothermalsynthesis of zeolites.

whose periodicity at a certain time is not constant. (Li et al., 2022b; Wang et al., 2021a).

The location where the material was found or discovered, a researcher's name, a company's name, or whether the material has a particular property are all frequent factors that affect the letters that are utilized(El Alouani et al., 2021), and so forth 218 zeolite frames had been given codes by the IZA-SC before July 2014(He et al., 2021; Ji and Zhang, 2022).

Zeolites have desirable characteristics that are governed by their structural makeup. The basic tetrahedral structure known as the Primary Building Unit (PBU) serves as the foundation for the zeolite framework. Due to the T atom's position at the PBU's center and its function as a connector between PBUs, the oxygen atoms are positioned at the corners of each unique tetrahedron. The frame type explains the connections and distribution of the T atoms throughout the three-dimensional network.

Each zeolite has a specific kind of structure, which also establishes characteristics like topology and pore size(He et al., 2021; Khan et al., 2021). Regardless of the zeolite's chemical makeup, the framework establishes its identity. However, there is a limitation when the $[TO_4]$ tetrahedra are bound to one another. Due to the negative charge contained in each PBU, which causes unfavorable interactions during the creation of zeolites, Lowenstein's rule (Rong et al., 2015)states that there cannot be Al—O-—i bonds, i.e., two nearby AlO_4tetrahedra. For this reason, in any zeolite framework, the minimum value of Si/Al is always 1(Chaibi et al., 2021; Razavi et al., 2021). Linking multiple PBUs leads to more complex secondary building unit (SBUs) structures. Rings are the most basic SBU types and can be divided into several categories based on how many T atoms they contain. For instance, a ring is a 4-ring if it contains four T atoms, and so on (Lv et al., 2022; Mauer et al., 2022). The pores of zeolites surrounded by several rings can be classified as very large (consisting of 14 rings), large (12 rings), medium (10 rings), and small (8 rings)(Lee et al., 2019). When the ring defines an oriented face of the three-dimensional frame, the word "window" is used. Zeolites can have frameworks with anything from four to twenty rings. (dos Santos et al., 2021; Kiwaan et al., 2021; Wang et al., 2021b)).

A more intricate SBU cage construction is created when these rings are combined. Meier first provided eight kinds of cages (Lepreux, 2005), but as characterization techniques advanced, it became possible to identify other types of cages. Cavities are categorized according to the type and quantity of rings (Zhang et al., 2019b). For instance, a cage [4662] has 8 windows: 6 are created by the 4 rings, and the 6 rings create 2. Fig. 1 shows examples of construction units and pore/cage sizes of three zeolite structures such as zeolite A (LTA), sodalite (SOD) and faujazite (FAU). Each type of zeolite is classified according to the number and type of rings. The Formation of the LTA, SOD (sodalite) and FAU (faujasite) structures from 4R and 6R PerBUs, with rings of 4 and 6 tetrahedra, respectively. In the structural models, the T elements in the framework are located at the intersection of edges, with oxygen atoms in the middle of the edges.

The oxygen atoms, which act as linking atoms between the T atoms, are absent from the graphical representation of the cages, as can be seen, to grasp the overall structure better. Greek letters are also used to identify some Zeolite structures. The latter is well-known for its inclusion in the popular A, X, and Y zeolites (Kim et al., 2021). In reality, X and Y zeolites have different chemical compositions but share the same structural element, FAU. Finally, cages and annular connections can form channels and pores in one, two, or three dimensions,with the junction of the channels resulting in the formation of the complete zeolite structure that we showed previously in the Fig. 1 (Ouyang et al., 2021).

2. Synthesized zeolites

Scientists have created synthetic methods in response to the first production of natural zeolites. The procedures used to create the synthesized zeolites have been repeated and altered to produce the same or different materials, occasionally with considerably different properties from the final output. Regardless of their origins, zeolitic minerals often develop under hydrothermal settings in nature (Kim et al., 2021). The goal of the earliest synthetic methods was to faithfully mimic the conditions that led to the production of natural zeolites, according to Fig. 2.

According to its definition, hydrothermal refers to heterogeneous reactions that dissolve and recrystallize materials that are typically rather intractable (Sadki and Saidi, 2014). As the name implies, hydrothermal techniques typically involve the use of water at relatively

Table 1
Organic and inorganic pollutants adsorption by zeolite-based materials from wastewater.

	Q_{max} (mg/g)	R (%)	Kinetic mode	Isotherm model	Ref
Oxytetracyclinehydrochloride	83.3		PSO	Langmuir	(Başkan et al., 2022)
CO_2	212.9			Langmuir	(Jedli et al., 2022)
CO_2	167.2				(June Choi et al., 2022)
CO_2	27.2				(Kwon et al., 2022)
Ammonium	17.8		PSO and Elovich	Sips	(Nguyen et al., 2022)
NH_4^+ and NO_3^-		98			
Rhodamine 6G		71	Thomas et Adams-Bohart		(Ameh et al., 2022)
Metal ions Pb^{2+}	487.8		PSO	Langmuir	(Ameh et al., 2022)
Metal ions Cd^{2+}	194.0		PSO	Langmuir	[92]
Methyleneblue	48.7	91	PSO	Freundlich	(Cheng et al., 2021)
Ammonium	11.8				(Muscarella et al., 2021)
CO_2	132.0				(June Choi et al., 2022)
CO_2		44			(Kim et al., 2022b)
Nickel		97	Avramikinetic	Redlich-Peterson	[43]
Ammonia and		88			(Ma et al., 2022)
Phosphate		99			[96]
Propionicacid	516.0		PSO		(Na chat et al., 2022)
VOCs		3 % CO_2 and 97 % CH_4			(Sun et al., 2022)
Co(II)	10.8		PSO	Langmuir	(Gupta et al., 2011)
Cu(II)	38.1		PSO	F and L	(Zhan et al., 2013)
As(V)	38.2		PSO	Freundlich	(Suazo-Hernández et al., 2019)
Cd(II)	78.1		PSO	Langmuir	(Ahali Abadeh and Irannajad, 2017)
Ni(II)	76.3		PSO	Freundlich	(Ahali Abadeh and Irannajad, 2017)
Pb(II)	123.0		PSO	Freundlich	(Nah et al., 2006)
Zn(II)	30.0		PSO	Langmuir	(Fungaro and Graciano, 2007)
Ga(III)	86.9		PSO	Langmuir	(Zhao et al., 2015)
In(III)	94.7		PSO	Langmuir	
Cs(II)	77.5			Freundlich	(Eljamal et al., 2019)
Cs(II)	81.4		–	Tóth	(Rahman et al., 2021)
Cr(VI)	48.3		PSO	Langmuir	(Angaru et al., 2021)
Cd(II)	196.0			Langmuir	(Elwakeel et al., 2017)
Cu(II)	118.1		PSO	Langmuir	(Li et al., 2020)
Cu(II)	77.8		PSO	Langmuir	(Kouznetsova et al., 2020)
Th(IV)	9.2		PSO	Freundlich	(Kaygun and Akyil, 2007)
Sr(II)	0.9		PSO	Temkin	(Yusan and Erenturk, 2011)
Cu(II)	719.3		PSO	Langmuir	(Dinu and Dragan, 2010)
Co(II)	467.9		PSO	Langmuir	(Humelnicu et al., 2011)
UiO_2(II)	536.3		PSO	Sips	
Th(IV)	438.5		PSO	Sips	(Humelnicu et al., 2011)
V(V)	277.7		Elovich	Freundlich	(Salehi et al., 2020)
Cr(VI)	344.8		PSO	Langmuir	(Mthombeni et al., 2015)
V(V)	74.9		PSO	Langmuir and Sips	(Mthombeni et al., 2016)
Pb(II)	138.8		–	Langmuir	(Nakamoto et al., 2017)
Cd(II)	56.2		–	Langmuir	(Nakamoto et al., 2017)
ReactiveOrange5	239.3		PSO	Freundlich	(Piri et al., 2019)
Reactive orange16	90.0		PSO	Freundlich	
Congored	40.6		PSO	Freundlich	
Methyleneblue	222.7		PSO	Langmuir	(Lin et al., 2016)
Methyleneblue	28.4		PSO	Langmuir	(Majid et al., 2019)
Methylene blue	49.0		PSO	l	
Congored	161.3		PSO	Langmuir	(Madan et al., 2019)
Methyleneblue	53.2		PSO	Langmuir	(Zhu et al., 2014)
Malachite green	48.5			Langmuir	
RhodamineB	64.4		PSO	Langmuir	(Cheng et al., 2017)
Methyleneblue	94.4		PSO	Freundlich	(Huang et al., 2019)
RhodamineB	32.7			Langmuir	(Li et al., 2020)
Methyleneblue	555.5		PSO	Langmuir	(Shui et al., 2020)
ReactiveRed 120	19.6		PSO	HillandSips	(Dehghani et al., 2017)
ReactiveRed 196	39.0			Jovanovic	
Brilliantgreen	90.0		PSO	Langmuir	(Shamsudin and Shahadat, 2019)
Methylene Orange	8.3		PSO	Langmuir	(Yang et al., 2019b)
Methyleneblue	151.5		PSO	Freundlich	(Khanday et al., 2017)
Acidblue29	212.7		PSO	Freundlich	
Methylene Orange	287.0		PSO	Freundlich	(Radoor et al., 2021)
Congored	19.0		PSO	Freundlich	(Radoor et al., 2020)
Malachite green	29.5		PSO	Langmuir	(Radoor et al., 2021)
Brilliantgreen	1461.3		PSO	Langmuir	(Mittal et al., 2020b)
Methyleneblue	661.9		PSO	Langmuir	(Mittal et al., 2020a)
Basicyellow 28	180.0		PSO	Freundlich	(Panic and Velickovic, 2014)
Basicyellow 28	117.0		PSO	Langmuir	(Panic and Velickovic, 2014)
Methyleneblue	81.9			Freundlich	(Sabarish and Unnikrishnan, 2018a)
MethylOrange	76.4			Freundlich	(Sabarish and Unnikrishnan, 2018b)
Methylene Orange	153.0			Freundlich	(Habiba et al., 2017)
AcidBlue74	16.6			Freundlich	(Ghanavati et al., 2021)

(*continued on next page*)

Table 1 (*continued*)

	Q_{max} (mg/g)	R (%)	Kinetic mode	Isotherm model	Ref
Hexavalentchromium	25.6		PSO	l	(Liu et al., 2022)
Phenol			PSO	l	(Ba Mohammed et al., 2019a)
Zn(II)	80.4		PSO	Langmuir	(Zhang et al., 2020)
Co(II)	9.1				(Lin et al., 2020)
Mn(II)	8.6				(Lin et al., 2020)
Cd (II)	92.4				(Zhang et al., 2019a)
Phenol			PSO	Freundlich	(Yousef et al., 2011)
Phenol	40.7		PSO	Redlich-Peterson	(Cheng et al., 2016)

high pressure (between 1 and 100 MPa), typically autogenous, and temperatures (ranging from 100 to 1000 °C) (Mthombeni et al., 2015). At the same time, a more current definition specifies that a hydrothermal process is one that is performed above ambient temperature and one bar (Lin et al., 2016). In this process, the pressure in the reactor increases as a function of temperature, but it is not common to know the pressure in the reactor. Therefore, the vapour pressure at this temperature is used as a reference.

The process is occasionally referred to as solvothermal when an alternative solvent is used in place of water. However, some authors have gone one step further and given the process a name based on the solvent it uses, such as glycothermal and ammonothermic(Liu et al., 2022).

A hydrothermal environment needs the presence of alkaline media in the form of basic solutions (Li et al., 2020). As a result, an alkaline media, a solvent, a supply of silica (SiO_2), and an alumina (Al_2O_3) are required to create zeolites. They are finally put together to react at high temperatures to achieve crystallization after aging. According to Cundy (Pei et al., 2022), the hydrothermal approach can be used to produce zeolite minerals in the following manner:

1. Silica and alumina from amorphous sources are dissolved and packed in extremely alkaline solutions with Na + or K + cations (NaOH or KOH).
2. The already-formed mixture is heated to increase pressure inside the autoclave and speed up the reaction product.
3. Before the reaction temperature of the amorphous gel. After that, it experiences what is known as induction time (the precursors remain amorphous, and no crystallographic phase is detected). When the first crystals form, this phase concludes.
4. The crystallization period, which follows the induction period, is when nearly all of the material progressively transforms into crystals
5. Following crystallization, recovery is accomplished through filtration, washing, and drying. In the event that use has taken place, the biological skeleton is eliminated using a high-temperature calcination procedure.

All scientific and technological advancements in the zeolites field have been made possible by manipulating and researching some important variables. Composition, time, temperature, agitation, and reaction kinetics are the primary determinants of zeolite synthesis (mechanisms).

3. Adsorption of various pollutants

Many scientific works on adsorption in zeolite systems have been carried out in different laboratories from different backgrounds. This is how several types of adsorbents of natural (zeolites, natural clays, etc.) or synthetic (activated carbons, bridged clays, silica gels, resins, etc.) origins have been prepared and tested in the adsorption of several organic (phenols, humic acids, dyes, pesticides, etc.) and inorganic micropollutants, in particular heavy metals.

On the other hand, the scientific literature offers us very few critical works on the adsorption of organic and inorganic compounds(Ahmed and Hameed, 2020; El Alouani et al., 2021; Elhakim et al., 2021; Jia et al., 2022; Marszałek et al., 2022; Sharma et al., 2022). Generally, the various works relating to adsorption (mainly organic and inorganic compounds) have emphasized the influence of certain parameters such as solubility, pH, temperature, the adsorbate nature, and the hydrophobic and organophilic properties of the different adsorbents used (Dehmani et al., 2020a, 2020b; Feng et al., 2020; Lütke et al., 2019; Nakhjiri et al., 2021; Wang et al., 2020; Yao et al., 2020; Zhang et al., 2021). Furthermore, in comparison with conventional adsorbents and to enhancenatural materials, several scientific kinds of research have focused on using zeolites to improve their surface adsorption properties.

These modifications, which are of physicochemical types based essentially on ion exchange, generally lead, depending on the nature of the modification, to obtaining hydrophobic adsorbents and heterogeneous catalysts. In the field of adsorption and despite their thermal instabilities, hydrophobic and organophilic, have been widely used in the depollution of water contaminated by certain organic micropollutants, in particular phenols, pesticides, dyes, etc.(Ba Mohammed et al., 2021, 2020; Beauchet et al., 2010; Sellaoui et al., 2021).

Since the 1990s, many studies have been performed on the adsorption of organic and inorganic pollutants on natural and modified zeolites (Armaroli et al., 1999; Clark and Snurr, 1999; Du et al., 1999; Hernández-Huesca et al., 1999; Huber and Knözinger, 1999; Ivanov et al., 1999; Khelifa et al., 1999; Mello and Eić, 1999; Rouquerol et al., 1999; Tatlier and Erdem-Şenatalar, 1999; Timonen and Pakkanen, 1999). The Table 1 presentsmost of the published works on using zeolites and zeolite-based materials in adsorption of organic and inorganic pollutants. The adsorbed quantity of pollutants on zeolite materials is very important in toxic and harmful amount of about 40.7 mg/g in the work of Wen PingCheng (Ping et al., 2016). A rate exceeds 98 % with an adsorbed amount exceeding 70 mg/g in the work of Bochera and his colleagues (Ba Mohammed et al., 2019a).

RushdiI. Yousefutilized a natural zeolite in the removal of phenol, the results in squash utilized the zeolite in adsorption with a removal rate greater than 90 % and a $Q_{max} = 356.5$ mg/g (Yousef et al., 2011). Zeolites are good adsorbents for dyes elimination, analysis of literature data shows that the adsorption capacity of Reactive Orange 5 is about 239 mg/g, Reactive orange 16 is about 90 mg/g and Congo red at a Q = 40 mg/g in the work of F. Piri(Piri et al., 2019)under normal conditions. An adsorbed quantity of methylene blue on a zeolite-based material in the work of Z. Shui(Shui et al., 2020)exceeds 555.5 mg/g. Finally, all published works show that zeolitic materials present a good and efficient choice for wastewater treatment.

pollutants for the environment. For example, a rate exceeds 99 % for cadmium in the works of D. Li (Li et al., 2022a), Z. AhaliAbadeh(Ahali Abadeh and Irannajad, 2017), K. Z. Elwakeel(Elwakeel et al., 2017), and K. Nakamoto(Nakamoto et al., 2017), very important results for a pollutant more dangerous for human health and the environment, phenol with low concentrations poses serious environmental questions. For this reason, the scientific research carried out in the last years on eliminating this pollutant, especially the adsorption, keeps a great place in the treatment methods used to eliminate phenols and phenolic compounds. This study exhibits the effectiveness of zeolite and zeolite-based materials in the adsorption of phenol. An adsorbed amount of about 40.7 mg/g in the work of Wen PingCheng(Ping et al., 2016). A rate exceeds 98 % with an adsorbed amount exceeding 70 mg/g in the work

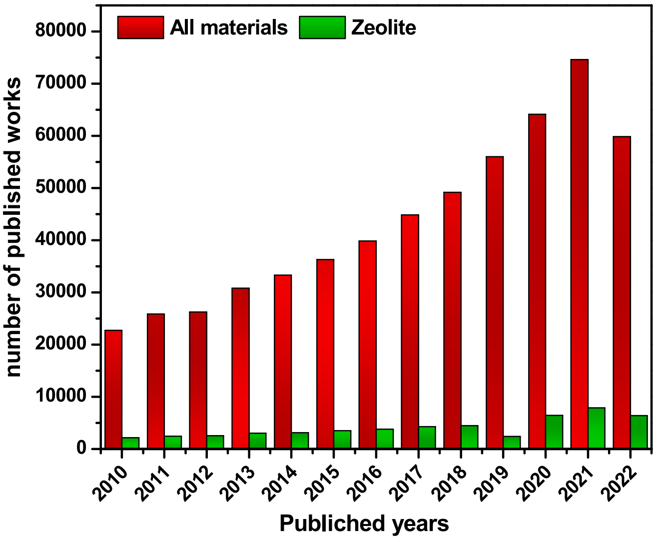

Fig. 3. Evolution of the number of works published in the journal on the adsorption of organic and inorganic pollutants.

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RushdiI.Yousef utilized a natural zeolite in the removal of phenol; the results in squash utilized the zeolite in adsorption with a removal rate greater than 90 % and a $Q_{max} = 356.5$ mg/g (Yousef et al., 2011). Zeolites are good adsorbents for dyes elimination; analysis of literature data shows that the adsorption capacity of Reactive Orange 5 is about 239 mg/g, Reactive orange 16 is about 90 mg/g and Congo red at a $Q =$ 40 mg/g in the work of F. Piri(Piri et al., 2019)under normal conditions. An adsorbed quantity of methylene blue on a zeolite-based material in the work of Z. Shui(Shui et al., 2020)exceeds 555.5 mg/g. Finally, all published works show that zeolitic materials present a good and efficient choice for wastewater treatment.

Prior to being released into aquatic systems, wastewater must be cleaned of organic and inorganic pollutants. A crucial topic of scientific inquiry for a long time has been the creation of efficient methods for this goal. Adsorption, precipitation, advanced oxidation, ozonation, coagulation-flocculation, ion exchange, solvent extraction, membrane filtration, and reverse osmosis have been tested to treat water that contains organic pollutants. However, the solid-state adsorption method has shown to be the most successful at removing organic and inorganic contaminants from wastewater of all the technologies discussed.

Compared to the other methods outlined, the adsorption process has several benefits, including low capital and operational costs and the potential use of a variety of naturally occurring solid adsorbent media. In order toeliminate organic and inorganic contaminants from home and commercial wastewater, adsorption technology has been extensively used. Many studies have focused on finding low-cost adsorbents with high adsorption capacities for typical contaminants and increasing their effectiveness for uptaking. Adsorbents have historically been used to treat wastewater, including activated carbon, clay minerals, agricultural wastes, and zeolites (Sharma and Feng, 2017). As efficient adsorbents for inorganic and organic contaminants in wastewater, zeolites have drawn much interest. The pore size and cages that result in size selectivity make up the empty space in zeolites. Zeolite utilized as an adsorbent is sometimes referred to as a molecular sieve because, from a molecular perspective, the zeolite lattice behaves to solid particles like an ordinary sieve.

For a given structure, the pore diameter and the diameter of the cages connected by the windows are fixed, but the size and number of cations present can still modify them. For example, the window diameters of zeolite A exchanged with K^+, Na^+ or Ca^{2+} are 0.3, 0.4, and 0.5 nm, respectively.

Much research was carried out on the organic and inorganic pollutants uptaking by using inexpensive adsorbents, including charcoal, zeolites, clay, and rock minerals (Fig. 3). The results in Fig. 3 show the increase in the number of works interested in the use of the adsorption process in the treatment of organic and inorganic pollutants. The strong point of this study is that it tells about the evolution of works based on zeolites and materials based on zeolite in adsorption. Moreover, a great percentage of these materials are compared with other solids.

Zeolites have been used in several research chapters and review papers to absorb both inorganic and organic contaminants (Abbasi, 2019; Aksu and Yener, 1999; Ioannou and Simitzis, 2009; Li et al., 2017, 2019; Liu et al., 2019; Mrunal et al., 2019; Sheela and Nayaka, 2012; Vikrant et al., 2019; Zhang et al., 2017). Work detailing the interactions between zeolites and organic and inorganic contaminants has been published (Guan et al., 2021; Mthombeni et al., 2016, 2015; Nakamoto et al., 2017; Piri et al., 2019). Some review articles target very specific organic substances, like phenols (Shakir et al., 2008) and alcohols (Elhakim et al., 2021). While some zeolite adsorption papers focused on the adsorption of heavy metals (Dehmani et al., 2020a), others have examined the removal of the dye from aqueous solutions (Abbas-Ghaleb et al., 2019; Choi et al., 2018; Huang et al., 2021; Jodeh et al., 2015). The subject of this criticism is the adsorption of organic and inorganic contaminants onto natural and modified zeolites. The adsorption capacity prediction of a system is not easily envisaged since it is strongly dependent on many factors, such as the properties of the adsorbent (particle size, porosity, surface area) and the adsorbate (structure, ionic charge, functional groups, water solubility, pKa, polarity, functionality). The adsorbent molecular weight and size, solution circumstances (such as the solvent, temperature, solute concentration, pH, ionic strength, and solute competition),the solid–liquid interface interactions, and the experimental apparatus type are some of these factors.

4. Effects of adsorption parameters

The pH is a significant factor in any adsorption study because it affects the adsorbent structure and the adsorption mechanism. In addition, this factor is important in the electrostatic mechanism. This factor depends on the origin of the water to be treated and its treatment process (coagulation, flocculation, oxidation). It is, therefore, useful to know the adsorption efficiency at different pH. We have represented some pH favorable to the adsorption of some organic and inorganic pollutants on different zeolites(Alkaim et al., 2014; Frost and Griffin, 1977; Griffin et al., 1977; Johnson, 1990; Miller et al., 1983; MULJADI et al., 1966).

For example, the variations of the quantity adsorbed as a function of pH from acidic to basic medium, from an initial concentration of phenol 60 mg/L and zeolite in the case of the adsorption of phenol go from 75 to 11 mg/g in the work of Bouchra Ba Mohammed. Three regions of the pH effect on the adsorption of the lord metals (Cd, Ni, Cu, and Pb) were noticed by HajarMerrikhpou(Yousef and El-Eswed, 2009);zeolite adsorption capacity increased as the initial pH of the solution increased from 2 to 4 (region 1). However, the zeolite adsorption performance remained almost unchanged in region 2 (pH between 4 and 7). These results were justified by the effect of pH on the structural properties of zeolite, an effect dependent on the dominant charge on the solid surface, whichis dependent on zero charge pH. It may be noted that the phenol adsorption capacity remains almost unchanged at pH between 3.3 and 8.6;however, it strongly decreases when the pH becomes higher than 8.6. Thus, at pH higher than phenol pK_a (9.89), phenol dissociates and appears as phenolate ions ($C_6H_5O^-$); these arguments are given by Wen Ping Cheng (Cheng et al., 2016)when studied the pH effect on the phenol adsorption on the composite zeolite X/activated carbon.

In his research, Rushdi I. Yousef (Yousef and El-Eswed, 2009) investigated the adsorption of phenol and chlorophenols on Jordanian zeolite tuff in buffered solutions with pH values of 4.0, 6.0, 10.5, and the pKa of phenols. The pH range was selected so that two values (4.0 and 6.0) were below the pK_a of phenols, one was at the pKa of phenols, and

Table 2
Effect of textural parameters on the amount adsorbed by zeolite(Jiang et al., 2020)(Chaouati et al., 2013).

Zeolite name	Surface area ($m^2\ g^{-1}$)	Pore volume ($cm^3\ g^{-1}$)	Si/Al ratio from XRF analysis	QLF (μmol/g)
FAU250	727	0.5136	409	1304
FAU50	698	0.5160	43	1271
FAU40	606	0.4819	32	825
FAU30	789	0.5233	31	837
BEA250	516	0.3022	286	414
BEA150	524	0.3243	107	420
BEA75	563	0.3720	78	530
MOR120	431	0.2687	113	529
HFAU (5)	690	0.357	5	13.610
HFAU (5) modified	670	0.360		4.239
HFAU (60)	906	0.485	60	24.512
HFAU (60) modified	821	0.473		21.958

Table 3
Adsorbed quantity of zeolitic materials. **MB**: Methylene bleu, and **MG:**Malachite green.

Adsorbedquantity mg/g		
Cd(II)	**MB**	**MG**
78.25	48.7	48.59
196	222.78	29.58
52.21	28.41	
92.47	49.02	
	94.481	
	555.56	
	151.51	
	661.91	

one was at a pH higher than the pK_a (10.5). The impact of phenol dissociation on adsorption has been thus investigated. The pH of the Jordanian zeolite tuff (6.9, Section 3.1), where the zeolite surface charge is positive and negative, respectively, was lower and higher than these pH values.

The amount adsorbed is proportional to the adsorbent surface area that is accessible to the adsorbate, which is frequently different from the material's total specific surface area. As a result, it depends on the material's accessible surface area, which might be challenging to determine. The porosity is influenced by how the pores' sizes are distributed. It reflects the microporous adsorbents' internal structure. (Haffane et al., 2016; Jeguirim et al., 2018). The porosity of adsorbent material is crucial when it comes to the adsorption of aromatic organic compounds.

Macroporous carbon has a better adsorption capacity than mesoporous carbon in the case of macromolecules. Indeed, the smaller size involvesa larger contact surface. This will then increase its retention power. Moreover, this surface increases when the support is porous. Industrial adsorbents (mainly zeolites) develop huge specific surfaces (600 to about 1200 m^2/g). Several works have shown the relationship between the adsorbed quantity and the textural parameters (Aazza et al., 2017; de Aquino et al., 2020). As an example, Nan Jiang shows an important difference in the quantity adsorbed of phenol of zeolite modefié one passes from 837 to 1304 mg/g for a zeolite faujasite of textural parameters (727 m^2/g and 0. 5136 cm^3/g) to another of parameters (789 m^2/g 0.5233 cm^3/g)(Jiang et al., 2020). The same result to the work of NourrdineChaouati;Briefly, the specific surface, the diameters of pores, and the volume of the pores have an important role in the adsorption for that in the synthesis of this material it is necessary to take into consideration these parameters(Chaouati et al., 2013).

The purpose of post-synthesis modifications of zeolites is to adjust the physical and chemical properties to the use for which the zeolite is intended. Synthetic zeolites whose compensating cation is an alkali metal (Na or K) have practically zero acidity. Therefore, ion exchange is the essential method for adjusting the acid-base properties of zeolites. The alkaline cations can be partially or exchanged either directly by protons in an acid medium (if the stability of the structure allows it) or indirectly by ammonium ions subsequently decomposed by calcination into volatile NH_3 and protons. In addition, they can be exchanged by multivalent cations (Ca^{2+}, La^{3+}, Ce°, etc.), which makes zeolites able to catalyze acid reactions. The nature of the exchanged cation and the degree of exchange determine the acidity and activity of the zeolite. The exchange of the zeolite by multivalent cations modifies its acidity and its selectivity and improves its thermal stability.

The parameters determining the speed and degree of the exchange are:

- The structural characteristics and the Si/Al ratio of the zeolite.
- The nature, the charge, and the size of the cationic species.
- The concentration of cationic species in the exchange solution.
- Temperature.
- The formation of acid sites using hydrolysis.

The chemical composition of a zeolite can be modified by reducing the number of aluminum atoms present in the network, which is done by dealumination. These operations can be carried out by various treatments, particularly acid treatment, hydrothermal treatment, and isomorphic substitution.

The number of aluminum in the crystal lattice of the zeolites, which defines both the number and the strength of the acid sites, is adjusted by dealumination; this is the main way to modify the acidity and the Si/Al ratio of a zeolite. The Si/Al ratio, therefore, conditions the strength of the acid sites.

From the results of Table 3, it is concluded that the adsorbed amount of organic and inorganic pollutants was affected by the effect of modéfication undergone by the zeolitic material. It is noticed that in the case of methylene blue, under conditions close to the level of temperature initial concentration and pH, it is found that the adsorbed amount increases from the order of 28 to 660 mg/g for a modified zeolite. In several authors' works on cadmium adsorption on zeolitic materials, it is noticed that the adsorbed quantity exceeds 190 mg/g compared with a non-modified zeolite 50 mg/g.

5. Adsorption isotherms

The adsorption isotherms are experimental curves representing at constant temperature the adsorbed quantity per mass of adsorbent (q_e in mg/g) at the concentration remaining in the fluid phase after the adsorption equilibrium (C_e in g/L). They are expressed in mathematical equations, these isotherms are intended to describe the adsorption process. Their appearance may represent certain phenomena involved: monolayer or multilayer adsorption and lateral interaction between molecules.

An in-depth search of the bibliographical data shows the modeling of the exemplary results of the adsorption of pollutants on zeolitic materials by the mathematical modules: Langmuir, Freundlich, Temkin, and Sips. The Fig. 4 shows that the Langmuir model describes 52 % of the works published, 34 % of the works are described by the Freundlich model, and 7 % by the Sips model. The rest of the work is described by other models by Tempkin, Redlich-Peterson, and others. An important remark is that the Freundlich or Langmuir model can describe the adsorption isotherm for the same adsorbate and in the same temperature range. As an example the case of the adsorption of phenol in the work of R. I. Yousef we find the model of Freundlich, and in the work of B. Ba Mohammed we find Langmuir.

6. Adsorption kinetics

The kinetics of adsorption describes the decrease in the

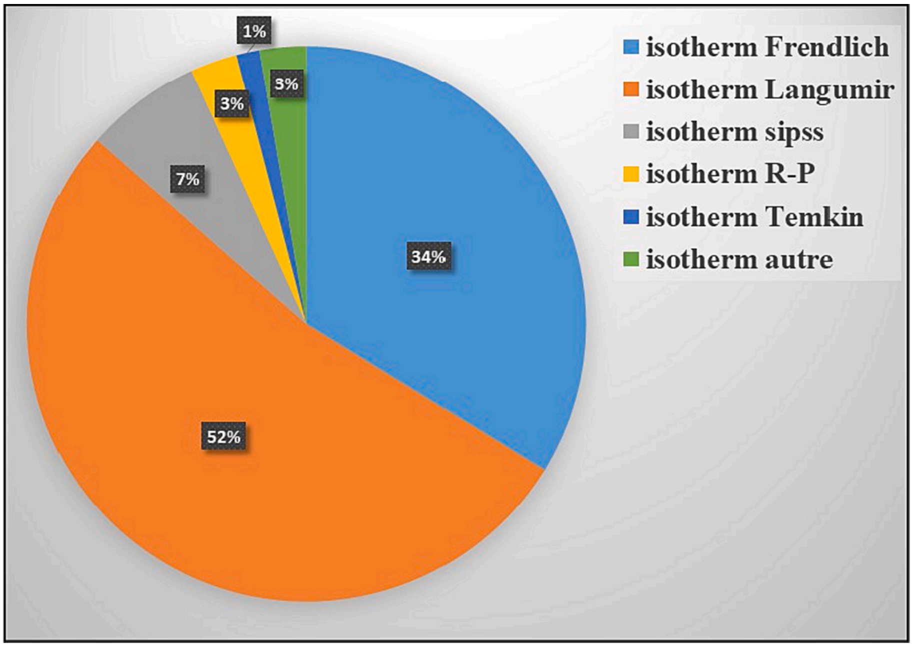

Fig. 4. Modeling statistics of the adsorption isotherm on zeolite.

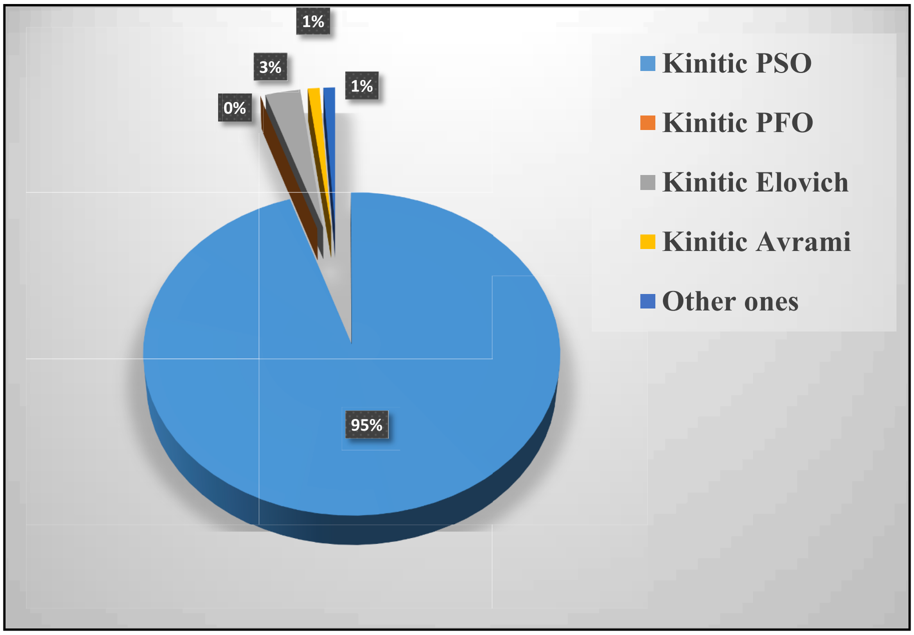

Fig. 5. Modeling statistics of adsorption kinetics on Zeolite.

concentration of the adsorbate in the solution as a function of contact time (Dehbi et al., 2019). Knowledge of the kinetics of adsorption is of considerable practical interest in the optimal use of an adsorbent and in the knowledge of the factors to be optimized to optimize to manufacture or improve an adsorbent leading to the fastest kinetics possible (Hassan et al., n.d.). The adsorption rate of a solute from a solution depends on many factors, including the adsorbent nature, the adsorbate, and the stirring rate of the medium. Therefore, it is very important to know the speed and the mechanism by which the materials eliminate the pollutant. For that, three kinetic models have been applied in order to evaluate the adsorption parameters. The models applied are:

- The pseudo-first-order model.
- The pseudo-second-order model.
- The model of intra-particle diffusion.

As shown in Fig. 5 the majority of the adsorption works of organic and inorganic pollutants follow a pseudo-second-order model (95 %); this is something that is in agreement with the nature of the zeolitic solid surface and the role played by the functional groups of zeolite and the ionic exchanges of this material.

7. Adsorption thermodynamics

Two types of adsorption exist, chemical adsorption and the other physical. Chemical adsorption forms a covalent bond between the

Table 4
Temperature and thermodynamic parameters effect on zeolite materials' adsorption.

	Temperature (K)		Thermodynamic parameter			Type of interaction	Ref
			ΔH/kJ·mol^{-1}	ΔS/J·mol^{-1}·K^{-1}	ΔG/kJ·mol^{-1}		
Ni^{+}(zeolite X)	298	318	31.039	118.581	−6.670	endothermic	(Pei et al., 2022)
Cd^{+}(zeolite X)	298	318	13.329	45.532	−1.150	endothermic	(Pei et al., 2022)
Phenol (faujasitezeolite)	298	313	18.69	14.62	−4.33	endothermic	(Ba Mohammed et al., 2019b)
Phenol (naturalzeolites)	298	318	−10.16	−0.0053	−8.58	exothermic	(Yousef et al., 2011)

adsorbent's surface and the adsorbate species. The energies of attraction put in this type of adsorption are very high, and the values of these energies are above 200 kJ/mol. In addition, physical adsorption is a process with weak interaction forces between the adsorbent and the adsorbent put in this type are of electrostatic nature (<80 kJ mol^{-1}), hydrogen bond (<60 kJ/mol), and type VanderWaals(Sabbatini et al., 2009) (<30 kJ/mol). When we put a solid (zeolite) in contact with a solution, each molecule of the latter manifests a capacity to adsorb on the surface of the solid; several conditions affect this phenomenon as the temperature, concentration, pH, the adsorbent nature, and the structural properties and textures of the adsorbent and their nature(Rajendran et al., 2022).

The temperature: Adsorption is an endothermic or exothermic phenomenon, depending on the adsorbent nature and molecules adsorbed, so the temperature has a very important role depending on the nature of the adsorption process (exothermic or endothermic. In the adsorption of organic and inorganic pollutants process, it is observed in this investigation that both processes exist even in the case of the same adsorbate. The chosen solid is noticed in several works indexed in the first table and the following table (Tables 4 and 2) that the temperature effect depends on the type of zeolite used. In work as an example, we conclude that the adsorption of phenol on natural zeolite is isothermal, a rare case in the literature. This justifies the importance of using natural zeolite in treating organic pollutants in aquatic systems. As for the inorganic pollutants, we notice in Y. Pei's work that the variation of the adsorbate does not affect the effect of temperature on the adsorption. Increasing the temperature leads to an improvement in the adsorption capacity of either nickel or cadmium ions. It is concluded in this part that the adsorption of zeolitic materials is a spontaneous process, and the other thermodynamic parameters are dependent on the type of zeolite used.

8. Conclusions and future outlooks

Aquatic systems containing organic and inorganic contaminants have been managed by using natural and engineered zeolites as adsorbent materials. The technical feasibility of zeolite adsorption techniques is in the function of the flexibility to remove organic and inorganic impurities present in these water sources, even though zeolites are acknowledged as a widely accessible and affordable alternative adsorbent. Additionally, some types of natural or altered zeolites could be appropriate for some applications but not others. So that existing information in the area can regularly be habituated to understanding each zeolite's technical performance as an adsorbent for the intended application, this review paper seeks to highlight the adsorption performance of various zeolites for diverse pollutants.

However, compared to carbon-based materials and graphene/zeolite, several investigations have found that the adsorption capabilities of 2,4-dichlorophenol and bisphenol are 10 % and 40 % higher, respectively. Zeolite-based adsorption methods also have advantages over other biological processes in cost (approximately 20 times less expensive than activated carbon) and byproduct release into the environment. Furthermore, organic and inorganic substances will not be eliminated by chemical pollutants. Although zeolites' adsorption efficiency is considerably improved by using surfactants in their preparation, further research should investigate and analyze the potential of thermally and surface-modified zeolites. Examining the possibility of organic/inorganic composites for pollutant removal is another way to enhance zeolite technology's adsorption performance potentially. In reality, the zeolite properties (nature, type,size), structure of the target pollutants, and operating conditions strongly affect the adsorption capabilities of both primitive and modified zeolites.

In order to maximize the use of particular zeolites against particular contaminants, a comprehensive approach must be used. Although the use of zeolite as an organic compound adsorbent was investigated about 70 years ago, only a few industrial uses, like oil spills, are in use nowadays. Therefore, the potential of zeolite-based adsorbents in comparison to other conventional techniques has been highlighted in this review. Nevertheless, most of the research reported in the literature is carried out on a small scale in the lab. Therefore, applying the performance statistics to large-scale facilities is impossible.

Additionally, an absence of reproducibility of performance and a variety of contradictory results are reported within the literature. In published studies, the regeneration of used clays is not examined. These are considered significant obstacles to using zeolite-based adsorbents in practical settings because organic and inorganic contaminants are invariably mixed in wastewater. Therefore, additional research is required to address these issues utilizing wastewater generated under industrial operating circumstances.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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