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A Review on Role of Zeolites in Next-Generation Wastewater Treatment and Resource Recovery

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Abstract
The world-wide need and desire to treat and recycle wastewater resources and ensure sustainability provide the increased demand and search of innovative materials. Crystalline microporous aluminosilicates known as zeolites are currently evaluated as promising materials due to their high surface area, pore structure control, ion-exchange capability and chemical stability. This review is a critical analysis of how zeolites are utilized in the next-generation processes of wastewater purification, including their use in the adsorption, ion exchange, photocatalysis, and membrane-based processes. Special consideration is considered the elimination of heavy metals, organic, nitrogenous and phosphoric chemicals, and dyes and valorizing wastewater streams- is investigated. The latest developments in the adaptation of natural and synthetic zeolites are also subject to discussion in the review to increase the selectivity, regeneration, and multifunctionality. Issues such as fouling, regeneration cost and scalability are critically examined in addition to the possible future hospitality of integrating the systems based on zeolite with the approaches of the circular economy. On the whole, this paper highlights the importance of zeolites as multi-purpose substances in the development of the sustainable treatment process of wastewater and its resources.

Keywords: *Zeolites, Wastewater treatment, Heavy metal removal, Microporous materials.*

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1. INTRODUCTION

One of the vital resources that help sustain life on the earth is water. The availability of fresh water on the surface of the planet takes place in a small percentage despite covering over 70 percent of the earth surface. In addition, a high portion of this freshwater is in glaciers making it unavailable in their pure form for direct use [1]. Also, the fresh waters that the world has are not distributed equally and reserves are evident in certain regions whereas some lack them. On top of the problem of relative scarcity and imbalanced distribution of this resource, the growing industrial activity has also contributed significantly to the pollution of the waterways through the dumping of the generated effluents in a fashion that is usually inadequate [2].

According to the demand in the society, food, cosmetics, mining and metallurgy industry, chemicals and pharmaceuticals industries have enhanced production at high rates. Therefore, the flow of volume and complication of the industrial effluents have attracted significant interest. The wastes of these industries are typically rich in hodgepodge of alcohols, inorganic polluters are in the form of dyes [6], bases of nitrogen [4] and phosphorus bases [5], heavy metals such as Pb (lead), As (arsenic), Cr (chromium) and Hg (mercury) [3], and numerous organic substances [7]. Upon release, these wastes have the potential to contaminate water, land, vegetation and wildlife and finally affect the human health via the food chain. The effects of these compounds are extremely toxic to living things, and the impact of the compounds depends on the concentration and duration of exposure. Excessive exposure to heavy metals and chemical pollutants in the long run or high-level has been associated with severe health challenges including cancer, genetic changes, organ dysfunction, system illness as well as neurological and mental deficits [8]. In addition, careless discharge of effluent, with high content of dyes, nitrogen, or phosphorus, may be disastrous to the ecosystems. Specifically, the introduction of these nutrients in water bodies can cause the development of eutrophication which is the process that starves oxygen water bodies causing death to aquatic organisms [9].

As a way of solving these fears, numerous procedures used in treatment of waste water have been invented with a view to minimise waste content prior to their disposal in the land fill. All have proved effective like chemical precipitation [10], ion flotation [11], electro-dialysis [12] and biological ones [13]; however, none have issues related to cost, efficiency, scalability and sustainability by the environment. As an illustration, they generate toxic waste which they have to be stored effectively, they are considerably costly in regard to chemical reagents, are costly to maintain, and are also inadequate in removing small concentration contaminant [14]. On the contrary, adsorption process seems to be extremely promising and has numerous strengths as compared to traditional processes and thus makes it an option in terms of wastewater treatment. The technique is flexible and can be used on a large variety of adsorbents, such as activated carbon and industrial waste [15].

As adsorbents to eliminate the impurities of water different kinds of materials have been utilized. In the wastewater treatment, the activated carbon, charcoal, clay minerals, and numerous new compounds are the most researched. They are effective due to the attribute of high surfaces, negative charge of surfaces (in case with clay minerals) or the structure of micro- to mesopores also noted in carbon-based materials [16]. Despite that, it has numerous barriers to its use in spite of the benefits. Examples of such electrophilic carbon include activated carbon and biochar, which are usually formed by roasting organic biomass, e.g., bamboo, maize straw, or rice husk, in limited oxygen conditions. But the nature of precursor material determines the characteristics of the final product as well as causing great performance variability in adsorption. It is a disadvantage to standardization and scale as the products are very uneven. Although advanced materials, which are the newly introduced category of materials employed in water treatment processes are not economical in terms of high costs of manufacturing, their high temperature in the calcifications is one of the main barriers particularly since the temperatures go as high as 900 °C [16].

This study has the objective of taking a closer look to the adsorption mechanisms linked to zeolites, critical findings, research gaps, and insights that may help in future research on the same. In contrast to most other reviews where the researchers declare their interest in synthetic and natural zeolites independently, the paper in question is also an attempt to provide a general representation of both brand of zeolites to the reader, helping him/her gain a wider perspective of their respective relative performance and applicability in wastewater treatment. This is done by summarizing the features, the advantages and disadvantages of the two types of the zeolites and their aptitude to treat wastewater based on differences in chemical and physical properties along with how the differences can be changed to enhance the adsorbent characteristic of the zeolites. The nature of zeolites and their properties is discussed in the section that follows, with the overview of at least five of the current works that utilised zeolites to remove five types of pollutants available in the section 3. Section 4 discusses the findings, challenges, and possibilities of using zeolite in the treatment of wastewater at last.

2. MICROPORE- STRUCTUTED ZEOLITE MATERIALS

2.1 Zeolite

Swedish mineralogist Axel Fredrik Cronstedt named the mineral in 1758 zeolite to describe a kind of mineral which swells upon heating. The phrase originates in the Greek words lithos [GB peace]: stone and zein [GB peace]: to boil; which means the effect following heating, one gets [17]. Only in 1857, it was explained, that zeolites possess a crystalline microporous structure, capable of trapping in platform and roomy water molecules within their pore and releasing them when the allot stem undergoes heating, and thus they followed reversible hydrating-dehydrating cycles. Within the same time frame, it was also found out that the zeolite structure has exchangeable compensatory cations. It has been demonstrated that ability of zeolites to exchange ions is exploitable to water purification and also remediation of polluted environments [17].

2.2 Synthetic Zeolites

Crystalline solids can be prepared in two ways; through reactions in the liquid or solid state. In liquid-phase reactions, reactions can occur more quickly at very low temperatures [18], whereas reactions in the solid state tend to occur slowly at high temperature (above 300 °C). Hydrothermal process is most favored and is considered to be one of the cheaper procedures of synthesising zeolite compared to other liquid phase process. This is a method in which the sources of water, aluminum and silicon are reacted in an aqueous solution. As well metal cations to balance the charge, a mineralizing agent (such as OH^- or F^-) or a structure-directing agent (typically an organic surfactant) to guide the formation of the desired crystalline structure, are all present in the process [19].The hydrothermal synthesis procedure is illustrated in Fig.1. The reaction occurs in the autoclave reactor at an exceptionally high temperature and at autogenous pressure of usually up to 15 bar. The resultant reaction product is filtered, dried in an oven and washed a number of times. A resultant solid is the zeolite which has many applications. Factors that are critical to the synthesis process but vary between zeolite types include the temperature and the time under which crystallization is to take place [18].

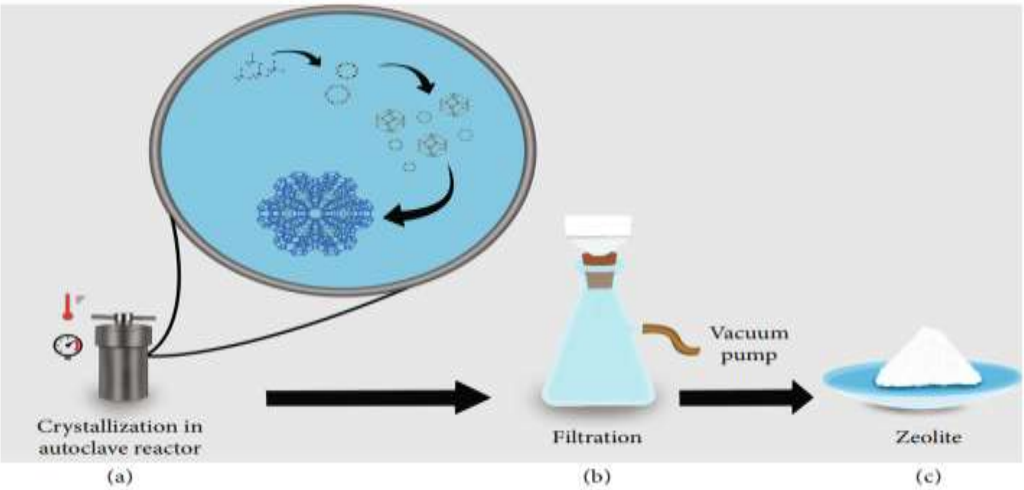

Figure.1: Diagram of the hydro-thermal synthesis. (a) Crystallization is done in an autoclave under controlled pressure and temperature. (b) The final product is dried in the oven.

Notwithstanding the large-scale application, the hydrothermal synthesis technique has some weaknesses that can act as barriers to its enhanced utilization. A major disadvantage is that it consumes a lot of water that leads to the production of alkaline wastes water that are difficult to treat and handle. Also, the process tends to take long reaction times which is inefficient when it comes to large production. In an attempt to surmount such difficulties, different alternative synthesis methods have also been established such as the microwave-assistance form of synthesis, sol-gel, ultrasound-assistance synthesis, and non-aqueous-based synthesis, the latter types replacing the conventionally used solvents of alcohols, hydrocarbons and ionic liquids respectively. All these approaches have their particular flaws and benefits. It is important to note that certain zeolites crystalline structures are only available through specific construction methods and this further limits the number of ways one can select certain methods of zeolites that can be suitable in a given application [19].

The process of synthesizing zeolites using chemical substances to obtain SiO_2 and Al_2O_3 is an expensive process. This prompted research on substitute, plentiful and cheap sources of alumina and silicon to increase in the recent decades as a result. To this, materials, including the waste ones, have been fallen into successful use. The inexpensive, readily available and produced in bulk to meet these leftovers are needed to contain silica besides alumina, and contain low levels of contaminants to qualify as being suitable precursors [20].

2.3 Natural Zeolites

Zeolites are naturally found in the fissures and pores of volcanic rocks and this limits the formation of large-scale, high concentration materials. This made naturally occurring reserves of zeolites quite rare. The earliest constituents used to form the zeolites are volcanic rocks, although the mineral composition of the rocks varies depending on conditions such as temperature, pressure as well as the supply of the mineralizing fluid used in the dissolution-precipitation system. The process of the formation of zeolitic structures can also be affected by other minerals and be facilitated by such minerals as volcanic glass, smectite, kaolinite, and feldspars, and other variants of zeolites [21]. Zeolite deposits that can be mined are current in extremely low geographical region on the earth and can be mined by the option of open-pits. Processing of zeolitic ores is relatively straightforward; the oil will need several mixture operations and grading in terms of particle diameters [22]. Amongst the sixty zeolites which occur naturally, only chaba zite, erionite and phillipsite, mordenite, ferrierite, clinoptilolite and analcime occur in sedimentary amounts, chiefly in China [22]. No estimates of the zeolitic ore reserves have been made since most of the producing countries do not disclose their anticipated production. In 2019, the United States (98.000t), New Zealand (100.000t), Slovakia (120.000t), Korea (150.000t), and China (320.000t) were the top producers of the items [23].

2.4 Zeolite Surface and Structural Modifications

The physicochemical and structural characteristics are closely related to the effectiveness of zeolites in the case of particular uses. Zeolites intrinsically have an anionized crystalline structure that is counter-balanced by exchangeable cations, and thus come in handy especially when removing cationic wastewater. This aspect, however, limits their adsorption of the organic molecules and the anionic species. Limitation

Surface modification of zeolites is often used. These changes do not only increase their ability to remove cations but allow adsorbing organic and anionic pollutants widening their relevance in wastewater treatment procedures.

Table 1: Waste materials with low cost for zeolite synthesis [14, 24, 25].

Discarded material	**Oxide source**	
	SiO_2	Al_2O_3
Kaolin/metakaolin	Yes	Yes
Glass powder	Yes	No
Aluminum waste	No	Yes
Coal fly ash	Yes	Yes
Fumed silica	Yes	No
Red mud	No	Yes
Oil shale ash	Yes	Yes
Bauxite residue	No	Yes
Electrolytic manganese residue	Yes	Yes
Diatomite	Yes	Yes

2.5 Water Treatment Using Zeolites: A Characterization Study

It is important to know the properties of a material in order to know the physical and chemical properties of the material and make projections of how the material will perform significantly in practical application. Certain characterisation strategies are of importance when applying the zeolites in the treatment of wastewater. X-ray diffraction or XRD is the most popular technique of estimating zeolitic phases. In addition to phase identification, XRD and Rietveld methodology enables one to detect and quantify crystallinity. This is especially important to the wild zeolites that often contain additional impurities minerals, as well as synthetic zeolites, wherein residual noncrystalline mining components in the manufacturing process can adversely affect the adsorption capacity [26].

In the case of organic contaminants, heat treatment is also considered a regeneration technique that produces effective zeolites for reuse with the chance of better economics of the process. A thermogravimetric analysis (TG) or differential thermal analysis (DTA) is usually used to assure the maximum temperature, at which zeolites retain their crystalline structure. These methods avoid situations where adsorption capacity is lost, or optimization of conditions where the regeneration takes place [27].

3. UTILIZATION OF ZEOLITES IN WASTEWATER TREATMENT

Improved attention on industrial effluents quality has been due to concerns regarding the nature that the industrial sector can have on the environment. Mismanipulation of water in this case represents several environmental hazards requiring the introduction of the efficient system of gathering and treatment [6, 28]. Moreover, environmental laws positioned seriously on the reusing of wastewater so as to reduce the consumption of freshwater and guard against natural resources [29].

3.1 Organic Compound Removal

3.1.1 Organic Compound

The zeolite- Based adsorption of organic compounds will depend on the Si / Al ratio of the adsorbent. Low-silica zeolites generally contain more compensatory cations exemplified by magnitude aluminum levels and hence their cation-exchange capacity grows as a result of containing an increased quantity of compensatory cations. Alternatively, high silica zeolites are more hydrophobic and organic compoundity is removed [30]. Organic materials can be modified to bring more affinity with regard to the anions as well as their adsorption ability to low-silica zeolites using surfactants. The molecule stays on the zeolite surface that is facing the solution and the chain of the hydrocarbon; value of which there are, therefore, themselves, hydrophobic. The importance of the concentration of the surfactant resides in the fact that it forms a second-layer-surfactant in case it reaches out past critical concentration (CMC) and consequently get back the hydrophilic nature of the zeolite [31]. Hosseini Hashemi et al. [32] tested zeolite Y that has been modified by hexadecyltri-methylammonium bromide (CTAB) to eliminate organic contaminant during an olefin production process. In order to evaluate the effect of dosage of surfactant in the cleaning of organic compounds, the zeolite y was modified to the different concentrations of CTAB, with the best performance of 0.025M CTAB, whereas higher concentrations led to the development of the double layer which renders a particles of zeolite not only hydrophilic but also decreased a adsorption of organic molecules. The adsorption of organic compounds takes place according to Langmuir model where adsorption occurs in monolayers with the same affinity of all sites on the zeolite Y after it is modified with CTAB. One can also assert that adsorption is favorable and that, physical processes (physisorption) are the driving force of it, all investigable isotherm models (Freundlich, Sips, as well as Dubinin-Radushkevich) have acceptable errors [32].

Table 2: The most common models used in explaining the kinetics and adsorption isotherm of zeolites in wastewater treatment system [33,34,35]

Adsorption isotherm models	**Model equation**
Langmuir	(1) $q_e = q_m (K_L C_e / 1 + K_L C_e)$
Freundlich	(2) $q_e = K_F C_e^{1/n}$
Sips	(3) $q_e = q_s a_s C_e{}^{n}{}_{s} / 1 + a_s C_e{}^{n}{}_{s}$
Koble-Corrigan	(4) $q_e = aC_e{}^{n} / 1 + bC_e{}^{n}$
Dubinin-Radushkevich	(5) $q_e = qm_{e\,(}\beta\varepsilon^2)$
Redlich-Peterson	(6) $q_e = K_{RP} C_e / 1 + BC_e{}^{g}$
Temkin	(7) $q_e = (RT/b_T) \ln (A_T C_e)$
Adsorption kinetic models	Model equation
Pseudo-first-order	(8) $q_t = q_e (1 - e^{(-k_1 t)})$
Pseudo-second-order	(9) $q_t = k_2 q^2{}_e t / 1 + k_2 q_e t$
Intraparticle diffusion	(10) $q_t = k_i{}^{t\,1/2}$

3.1.2 Dyes

Dyes are removed by ion exchange characteristics in zeolite surfaces. Due to their higher compositional stability and different physical properties such as crystal structure, pore size, compensating cations among others, synthetic zeolites often have higher adsorption capacity than the wild zeolites. Zeolites are not among the best in adsorbing ions and in multi-component systems competitive adsorption is often observed. The article by Ayar et al. [36] examined how the safranine O (SO) reacted during the presence of Cs^+ ions in zeolites prepared using natural clinoptilolite or fly ash (analcime + sodalite). When the concentration of Cs^+ was increased, the removal of SO was reduced in both substances, as the cations competed on the free active adsorption sites. Clinoptilolite (4.1 AFs) was able to absorb more SO than analcime (3.2 AFs) and sodalite (2.3 AFs), probably because it has a larger pore size.

The adsorbed particles should be measured, which determines the adsorption efficiency of dye adsorption systems to a considerable extent. Wang and Zhu were comparing extraction ability of clinoptilolite zeolite on rhodamine and methylene blue dyes [37]. They have found that the larger size of the molecule of rhodamine B increased the hindrance in the dispersion process within the zeolite pores. At room temperature, the substance had a removal capacity of 9.3 mg-g -1, of which 55 percent of capacity was confirmed by the production of methylene blue (16.7mg-g -1). Regrettably, anionic dyes were observed to adsorb much less naturally occurring and in reactionary prepared zeolites than dyes with cationic properties due to repulsive forces created by the Static electricity. It is due to this fact that pH correction is a necessary procedure that helps to eliminate the said species. This is due to the fact that the two types of zeolites are capable of obtaining a positive charge. Temperature is one more factor, which defines zeolite adsorption of color. The temperature also accelerates the adsorption process because it not only stimulates the ion mobility but also the zeolite crystalline structure development process. Wang and Zhu have explored the adsorption of MB and rhodamine B using the natural clinoptilolite [37]. They were discovered therefore to adsorb more at the temperatures of around 50 °C due to the adsorption of both the dyes implying that adsorption was an endothermic process and that the temperature rise of the high was boosting the process.

3.2 Inorganic Compound

3.2.1 Phosphoric compounds

They are especially hard to eliminate in solutions because they occur over a broad range of pH in an anionic form and zeolites are less efficient in elimination of anionic species in solutions. H_3PO_4 is the major species below the pH 2, $H_2PO_4^-$, HPO_4^{2-}, and PO_4^{3-}, are the dominant species between pH 2 and 7, pH 7 and 12, and above the pH12, respectively. These species have their negative charges thereby repelling electrostatically. At acidic pH regime, there is protonation of the zeolite surface, and this binding reaction leads to attachment of phosphate ions and impartation of a positive electrical charge. After this the molecular species react chemically with the zeolite surface. This effect was fixed by Zhang et al. [38] using granular natural zeolite to extract phosphorus in the wastewater. Their studies, over the broad pH range (1.8-12), found that remover efficiency ranged through the state of 100 percent at pH 2, virtually zero percent at pH 7 and strategically increased in the alkalinity. The authors assigned enhanced performance in alkaline media was formed through chemical precipitation but with acidic conditions, performance was more enhanced through adsorption due to expansion of zeolite pores which further increased the available surface area.

The other important factor in adsorption of phosphates on zeolites is the temperature. An increase in temperature positively affects adsorption capacity. These phosphate molecules become even more soluble in the solution thereby increasing adsorption activity of the molecules on the pollutant. According to Goscianska et al. [39], the adsorption of synthetic zeolite NaP1 (10.5 to 16.2 mg.g like) increased with temperature, focusing on the temperature, temperature increase, to 25 to 60 °C along with the synthetic zeolite LaP1 (57.7 to 63.9 mg.g like), which showed the same phenomenon (notably the endothermic nature of the process). pH control is of particular concern to anionic species such as phosphates. Zeolite adsorption is preferable at a lower pH level than the zero charge (PZC) where the adsorbent acquires positively charged sites on its surface.

3.2.2 Heavy metals

The most effective are synthetic zeolites, which work best in the treatment of the wastewater, which is preceded by the modified zeolites. Natural zeolites the least amount of metal sorption of most of the heavy metals, and are very appealing to the corporate sector, because of the high regional as well as inter-regional variations of the mineralogy [22]. Also, the ore is usually rich in various kinds of zeolites as well as other types of mineral which serve as pollutants due to their low sorption capacity of metals. Examples of the contaminated minerals are, Heulandite, quartz, feldspar, calcite, halite, biotite, illite, and montmorillonite [40].

The synthetic zeolites are very compatible with adsorbents having one compensatory cation, hydrophobicity/hydrophilicity, and pore size distribution. They are normally single-phased. Fast exchange of cations is ensured by a combination of these characteristics as compared to natural zeolites. The adsorption of the zeolites on the metal is usually an endothermic and spontaneous phenomenon and this one has a greater rate of adsorption at a greater temperature [41]. This has been demonstrated by Dal Bosco et al. [40] by carrying out adsorption experiments in zeolite scolecite at 25, 40, and 60 °C using Cr^{2+}, Ni^{2+} , Cd^{2+}, and Mn^{2+}. The authors demonstrated that adsorption of each metal were highest at experiment temperature of 60 °C, then 40 °C followed by 25°C in that order. Other research works conducted by Čurkovic et al. [42] involved investigation of Pb^{2+} and Cd^{2+} removal in different temperatures (20- 70 °C) using the natural clinoptilolite. It was also found that in all scenario that was under investigation, temperature increases positively affect the adsorption rate.

Change in pH level also significantly reduces the adsorptive property of the zeolites. This was demonstrated by the fact that in extremely alkaline conditions, the precipitated form of metal hydroxides and oxides reduce the number of accessible ions in the process of adsorption thereby lowering adsorption rate. Since in this case the metal (that is going to be adsorbed) is opposite to this chemical constituent and, in its turn, reducing the effectiveness of adsorption, upper concentration level of H^+ in a solution does not enable adsorption under extremely acidic environmental conditions, as stated by Zanin et al. [43]. These authors determined the efficiency of clinoptilolite in receiving Cr^{3+}, Cu^{2+}, and Fe^{3+} ions out of solutions at pH 3, 4 and 5. The highest rate of chromium removal was at pH 5; the lowest rate was in pH 3. Aluminum adsorption is at its maximum limit at pH 4, whereas iron was slightly lower at pH 5. It was observed by Dal Bosco et al. [40] that the density of adsorption of Cd^{2+}, Mn^{2+} and Cr^{3+} gradually scaled as the pH of the solutions changed to 3, 4 and 5 respectively: in zeolite scolecite. The speed at which Ni^{2+} was adsorbed conversely increased substantially on the shift to pH5 but quickly between pH3 and pH4.

3.2.3 Compounds of Nitrogen

Demonstrates that zeolites have a great potential in eradication of ammonia in wastewater. The man-made forms are more modified to adapt small molecules of ammonia (0.283 nm on average) because of the increased diameter of the pore that allow the entry into the crystalline structure of zeolites [44]. Temperature also determines how much ammonia is removed by zeolite whereby, the more the temperature, the lower the calculation of ammonia removal due to the Ammonia hydrolysis of the surface [45]. On the other hand evidence indicates that an increase in temperature increases the amount of ammonia adsorbed (Widis et al. [46]). This means that we are yet to have a thorough understanding in the effects of temperature on adsorption of ammonia on zeolites. Although the temperature also plays major role in the adsorption mechanisms, it has not been thoroughly researched and it will be necessary to conduct further research to understand how temperature influences the ammonia-zeolite adsorption system. pH also influences the power of sorption process. Valence Positive ions inhibit the sorption capacity at the sorption in low PH levels by competing with the molecules of ammonia. When NH^{4+} is transformed to NH_3 in a alkaline conditions, the electrostatic attraction on the adsorbent x between the adsorbent and adsorbate is overcome thus reducing the adsorption rate. Fu et al. [47] employed the natural clinoptilolite to examine the ammonia adsorption within a pH of 4-10. They discovered that NH_4^+ was removed best at a PH range of between 5 and 8. On the same note, Zhao et al. [48] undertook adsorption studies using a wider range of pH of 2 to 11.

3.3 Studies of Kinetics

The dynamic models of adsorption assist in explaining the simple law of transfer of mass besides providing an understanding on the rate at which adsorption takes place, alongside the overall efficiency of an adsorbent [43]. The ideal adsorbent would be an excellent adsorbent with a high adsorption capacity and rate of adsorption. The mass transfer entails three steps. The adsorbate is attached onto the solid by a film of liquid in the former method, the so-called external diffusion. Differences in concentrations of the adsorbate accelerate external diffusion process, which depends upon the amount of adsorbate. The second step takes place when entering into-pores, adsorbate when it is called internal. The third and last stage is the adsorbate-adsorbent active sites associated. The most employed adsorption isotherms models of sorption, as well as the other, models of adsorption kinetics, are visible, which can be deployed with zeolites as adsorbents in an adsorption system.

Adhesion has been reported by a few sources in different studies on heavy metal adsorption, including intraparticle diffusion and Jin et al. [49] established the validity of their results. This implies that the adsorption will occur in the holes of the crystalline structure of the particles after it has occurred at external surface the particles. Thanks to the fast adsorption of metallic ions which is possible based on their similarity with the pores, which concern both size, a couple of seconds suffices, regardless of the heavy metal to be removed and the zeolite employed. Zhao et al., Lin et al. shared the findings of ammonia adsorption research performed on analcime, P zeolites and chabazite zeolites, at which ammonia extraction was 56.4 and 50 percent, respectively within the framework of the first five minutes of interaction between ammonia and zeolites [50]. Originally, the intraparticle diffusion model had a poor fit on the observed sample values, which ought to have been the case, however, dispersion was believed to be the main adsorption mechanism the experiment experienced within the first 30 minutes of interaction [50]. With consideration to the dye adsorption, the data reveal that the zeolites and dyes under study adsorb onto their outer surface rather rapidly. Molecules can hardly enter through the holes of zeolites, which results in a very low rate of intraparticle diffusion as indicated by the size of the zeolite. Nowadays the capability of synthesized zeolites to adsorb dye is higher due to larger pore size, more uniform chemical structure (enhanced stability owing to Si/Al ratio). Again, cell size in a particular dye molecule and the pore diameters in the zeolae play an important role in the adsorption time. This is important information that can guide the development of the methods that will eliminate this type of contamination.

The process of removal of other impurities is not the same as the adsorption on the pure substances to zeolites since the hydrophobic interaction between the adsorbent and the latter is of more significance in this condition. Such an interaction is made possible by a hydrophobic monolayer that is created on the surface of the zeolites when surfactants are added such that the zeolites become protected. Nevertheless, I will only be covered by the surface of particle and accordingly the width of their pore cannot support the dimension of the surfactant molecules. A study by Kuleyin [51] establishes that the removal occurs within initial half an hour of contact and becomes steady which is indicative of limitation in adsorption by diffusion in zeolite pores. The fact that the mechanism does not fit very well with the intraparticle diffusion hypothesis still indicates the possible external adsorption of organic molecules. Adsorption is expected to occur on the side of particles; consequently, the particle size of a zeolite is expected to influence the effectiveness of the process. When the value of this parameter is priori information the comminution process would be optimized considering natural zeolites. One of the physical characteristics of adsorbents, however, is not granulometry, which were utilized in the considered study. Another factor that has not been examined is PH that may influence the adsorption process. This variable can have a serious procedure influence by modifying ionization of surfactant or adsorbate molecules.

3.4 Adsorption Isotherms

The training of efficient adsorption equipment to demonstrate wastewater treatment efficiency is dependent on the adsorption mistake and determining the maximum adsorption ability (mg/g) because the two variables are quantified by the isotherms of adsorption [52]. At a given concentration level of adsorbate, the counterrelation between the amount for adsorbate held on the top surface of the adsorbent and the types of liquid at a constant temperature are prescribed by isotherm curves. Table 2 contains the most commonly used adsorption model to characterize adsorption processes of the microporous materials. The models that are mostly used to explain the adsorption isotherms are the Freundlich and Langmuir models. Whereas Freundlich model describes the multilayer adsorption of the heterogeneous surface, Langmuir teaches multilayer adsorption of the homogenous adsorbent surface of a surface [52]. Temkin model describes adsorption to be characterized by adsorbate-adsorbate interactions which denotes that the most important is the hydrophilic interaction with the tail group. The Redlich-Peterson model is more appropriate with a wider range of contaminant concentrations than the Langmuir (Fontabel) and the Freundlich models, non-the-less the modes can fit the circumstances very successfully in certain cases [53].

The experimental outcomes of studies under evaluation were best associated with Langmuir model. It means that adsorption on heavy metals systems with the application of zeolites as an adsorbent is applied on the surface of a particle, creating a monolayer. In the case of adsorption, more catalogs are available at the start of the initial few minutes of contact that makes adsorption rates high. The pores of the crystalline structure become open to allow the ions move after identifying a space in the surface [49]. Sodium concentration formation at equilibrium level was found increased in the studies of Bai et al. [54] and Hui et al. [47], implying that there was also ion exchange in the solution that led to the elimination of heavy metals. Among the studies reviewed, the Langmuir isotherm model ofpinacolone adsorption ware best fitted to gaps around the observed values but this is not due to the reaction of pinacolone with ammonia to form a single-layered structure but it is due to the creation of a single-layered structure based on the reaction between pinacolone and the hydrogen competitive mispartitioning layer caused by the ammonia molecule's lone pair of electrons interacting with intrinsic Wasserkorrekturns parameter. Other than the spontaneity of the process, the Freundlich model was also able to establish the heterogeneity of the surface of the adsorbent [35, 55].

Like in case of nitrogenous compounds and heavy metals, both electrostatic interactions and cationic exchange play a significant role in the adsorption process of the dye. The action is more effective with zeolites with lower Si/Al ratio due to the higher percentage of aluminum atoms that concentrate to form a negative charge. This causes the creation of monolayer due to advancement of only one dye molecule on any given negative site. Most of the studies have supported this theory, and it has been established that most of the isotherms can be best described by the Langmuir isotherm. The phosphoric compounds can also be adsorbable through the electrostatic attraction as in the case of the other ones; however the number of molecules that can adsorb at a given site is one, which leads to a formation of a monolayer. The Langmuir isotherm is a clear expression of this phenomenon and it just coincided that it was a criterion anaerobic model appearing to be the most significant matching experimental information when it comes to researching the use of zeolites as an adsorbent. On the one hand, Freundlich isotherm is optimal if the Langmuir model is applied to the inorganic compounds, which implies an invitation to such a way of interaction to form a layer of pollutant molecules at the top of the surfactant layer.

4. CONCLUSION, CHALLENGES AND FUTURE PRESPECTIVE

The same experiments by various authors show that with synthesized zeolites, the crystal structure arrangement order is much higher than the one produced naturally, and as a result, pore sizes and strength of cation exchange capacities of synthesized forms are more coherent. This permits a greater ability to adsorb in the scenario of the numerous chemicals used. Natural zeolites are not effective enough because, the adsorption capacity of the contaminated minerals do not have high adsorption properties. Hence, it can be concluded that naturally occurring zeolites can be partially modified in respect of their nature and abilities to apply them in wastewater. The rate of adsorption of, such contaminants as ammonia, dyes, heavy metals onto zeolites is extremely quick. Nonetheless, the adsorption of anionic molecules such as phosphoric acid and a number of heavy metals (such as Cr and As) is not possible because the adsorption takes place when the adsorber and adsorbate want to remain in close contact.

This has been achieved by modifying zeolites, which bear a hydrophobic layer around their surfaces resulting in their attraction to organic molecules, which are in effect mentioned as not being very hydrophilic. This makes dosage of the surfactant to be an aspect of consideration. Despite the fact that it is clear that dessicated zeolites are effective, as pollutants in a number of bench-scale studies. Two other directions that need to be followed are adsorbents, desorption, temperatures (dyes), granulometry, pH (organic substances), adsorption kinetics (phosphoric substances), and pilot-scale studies. In addition to this, they employed some synthetic solution, which could not mimic the nature of various wastewater. The fact that there may be other chemical species and may have different effects on the adsorption of the processes in the large-scale commercial process using zeolites should always be raised.

The high surface area, ion-exchange capacity and porosity tunability of zeolites prove to be very impressive in the treatment of wastewater and recovery of resources. They can recover heavy metals, nutrients and organic contaminants, as well as allowing used water natural reuse and recovery of a useful by-product. Their real-world applications are however constrained by obstacles including pore blockage, regeneration performance, cost-effective scale production, and detrimented performance when in a complex wastewater matrix. Further studies are needed to give increased selectivity, stability, and scalability by modifying surfaces, forming composite and integrating it with advanced technologies, so that zeolites play a significant role in the next generation of circular and sustainable wastewater management.

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