APPLICATION OF NATURAL ZEOLITE IN WASTEWATER TREATMENT – A REVIEW

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Abstract

The environmental pollution is a major problem over the world. The large amount of pollutants formed during various industrial processes reaches the ecosystem. Thus, heavy metals, oils and other organic compounds are responsible for water contamination. It is known that heavy metals have toxic effect on the environment. Additionally, they are non-degradable and have ability to reach the living organisms through the food chain. Among different methods, the adsorption is widely used in wastewater treatment due to simplicity of the process and efficiency in the removal of pollutants. Natural zeolites from different deposits have shown good selectivity for heavy metal ions. Additionally, they are environmentally friendly, low-cost and have high ion exchange capacity.

Key words: Wastewater treatment; Pollutants; Natural zeolite; Adsorption.

1. Introduction

Due to the fast development of industry, a large amount of wastewater has been produced. The wastewater contains various pollutants including heavy metals, oils and organics that have toxic effect on the environment [1]. Beside industrial processes, such as melting, galvanizing, electrolysis, wastewater originates from domestic sewage and agriculture, too [2-5]. The removal of pollutants from water is the subject of many studies [6-8]. Researchers are constantly working to determine a suitable and efficient method for wastewater treatment.

It is known that heavy metals are toxic and are non-degradable. Thus, they have ability to reach the living organisms through the food chain [9, 10]. Having this in mind, they must be eliminated from water or reduced [11].

Different efficient techniques including solvent extraction, membrane filtration, chemical precipitation, ion exchange and adsorption can be used to remove contaminants from water and they have drawn significant interest of researchers [12,

13]. Due to the simplicity of the process and the high efficiency of pollutant removal, adsorption is an increasingly examined method for wastewater treatment. In addition, various materials such as zeolites, clays and by-products from different industries can be used as adsorbents. Except these natural materials, synthetic polymer adsorbents are commercially available. Activated carbon is also a significant adsorbent derived from natural organic materials. Characteristics of activated carbon that make it effective in removing pollutants from aqueous solutions are the large specific surface area, porous structure and thermostability [14]. However, due to the high cost of this adsorbent, researchers are still trying to find new, cheaper and more efficient adsorbents. Based on researches [15-17], it has been observed that various agricultural and industrial by-products can be successfully used to remove pollutants from aquatic environments. In addition, the cost and availability of these materials have an impact on the selection of the appropriate adsorbent. Among them, zeolites are the mostly used adsorbents because they are environmentally friendly, low

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cost, possess good selectivity for different cations and have high ion exchange capacity which is accompanied with a release of non-toxic exchangeable cations (K^+ , Na^+ , Ca^{2+} and Mg^{2+}) to the environment [18].

2. The structure of zeolites and their characteristics

Considering physical properties, crystal structure and morphology of natural zeolites as well as types of exchangeable cations in zeolite structure, they are divided into seven main groups [19]. In the 1970s, natural zeolites have attracted the attention among numerous researchers because of their ability to remove heavy metals such as strontium and cesium [20]. Analcime, chabazite, clinoptilolite, erionite, mordenite and phillipsite are the most common natural zeolites. One of the most investigated zeolite in wastewater treatment is clinoptilolite [21-23]. In general, chemical formula of zeolites is given as in the following equation [1]:

where $M = Na, K, Li$ and $/$ or Ca, Mg, Ba, Sr , n is the charge of the cation $Y/X = 1-6$, $P/X = 1-4$.

Zeolites are microporous materials with cavities less than 2 nm. Their structure is based on a three-dimensional framework of tetrahedral units (SiO_4^{4-} and AlO_4^{5-}) connected by sharing one oxygen atom [24]. Such a well-defined three-dimensional structure contains channels, cavities and pores of molecular dimensions within which the catalytically active centers of are located [25]. Active centers are available only to molecules whose dimensions are smaller or approximately equal to the dimensions of the pores themselves (molecular sieve effect) [4, 26]. The aluminum ion is small enough to occupy a position in the center of the tetrahedron composed of 4 oxygen atoms, while the isomorphic replacement of Si^{4+} by Al^{3+} ions produces a negative charge on the lattice. The negative charge of the lattice is balanced by exchangeable cations, Na^+ , K^+ or Ca^{2+} , which are

relatively harmless [27, 28]. Furthermore, a characteristic of these cations is their mobility due to weak electrostatic interactions with the aluminosilicate lattice. Therefore, there is a possibility of ion exchange between zeolites and ions present in aqueous solutions.

Due to structural properties such as large specific surface, well-defined micropores in molecular dimensions, as well as high chemical and physical stability (e.g., thermal and mechanical) [29], and sorption characteristics [30], natural zeolites are interesting materials for application in wastewater treatment [31], in petroleum refining [32], in agriculture [33], in biotechnology and medicine [34].

The reaction of ion exchange in aqueous solution occurs according to the following equation:

where r is the charge of the cation R contained in the solution, while z is the charge of the moving cation Z in the zeolite.

The Si/Al ratio, specific surface area and pore size are also important features that affect the adsorption capacity of zeolites. This is the main reason that different tests on clinoptilolite obtained from different deposits may show different results. Although there may be some variations in the chemical structure of clinoptilolites from different deposits, as well as different mineral impurities in different samples, the main reason for the difference in the obtained results is the different physical and chemical conditions used in the experiments.

Table 1. Cation exchange capacity for different zeolites [35, 36]

Zeolite	Si/Al ratio	CEC, meq/g	Main cation
Chabazite	1.4–4.0	2.5–4.7	Na, K, Ca
Clinoptilolite	4.0–5.7	2.0–2.6	Na, K, Ca
Mordenite	4.0–5.7	2.0–2.4	Na, K, Ca
Erionite	2.6–3.8	2.7–3.4	K, Na, Ca, Mg
Ferrierite	4.9–5.7	2.1–2.3	Ca
Laumontite	1.9–2.4	3.8–4.3	Na, K, Mg

Additionally, in Table 1, it is shown the Si/Al ratio and cation exchange capacity (CEC) for some natural zeolites [35, 36].

Also, the chemical composition of zeolites from different deposits is shown in Table 2.

Table 2. Chemical composition of zeolites from different deposits

Zeolite	Components (wt%)								Reference
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Na ₂ O	K ₂ O	CaO	MgO	IL	
Clinoptilolite ^a	65.63	12.97	1.48	1.20	1.33	3.21	1.41	12.96	[37]
Clinoptilolite ^b	64.20	11.67	1.03	2.36	3.84	7.42	0.35	8.66	[38]
Clinoptilolite ^c	64.93	13.39	2.07	2.40	1.30	2.00	1.09	9.63	[39]
Clinoptilolite ^d	70.08	11.72	0.67	0.55	3.50	3.18	0.71	9.45	[27]
Clinoptilolite ^e	64.51	11.25	0.97	3.60	1.21	4.38	0.60	13.14	[40]
Clinoptilolite ^f	67.16	12.30	2.30	0.66	2.28	2.91	1.10	/	[41]
Clinoptilolite ^g	65.52	9.89	1.04	2.31	0.88	3.17	0.61	/	[42]
Clinoptilolite ^h	68.26	12.99	1.37	0.64	4.11	2.09	0.83	/	[43]
Clinoptilolite ⁱ	65.0-71.3	11.5-13.1	0.7-1.9	0.2-1.3	2.2-3.4	2.7-5.2	0.6-1.2	/	[22]
Clinoptilolite ^j	63.60	11.81	1.74	0.40	4.40	7.35	0.69	9.81	[74]
Clinoptilolite ^k	66.57	13.13	2.30	1.27	1.17	3.85	0.56	11.05	
Clinoptilolite ^l	65.89	12.86	2.06	0.97		4.90	0.95	11.26	

a – from Serbia, b – from Bulgaria, c – from Croatia, d – from Turkey, e – from Argentina, f – from Slovakia, g – from China, h – from Australia, i – from Slovakia, j – from Serbia (Vranjska Banja), k – from Serbia (Brus), l – from Romania (Baia Mare), / – no data, IL – ignition loss.

3. Wastewater treatment using natural zeolites

Adsorption in wastewater treatment is considered as an effective technique in comparison with other methods. The experimental conditions for removal of contaminants by adsorption process are very important. The ability of zeolite to be regenerated and its reuse is very important for the environment because this prevents the generation of new waste material [44]. Many researchers are dealing with the application of natural zeolites in removing metal ions from wastewater [22, 23, 44, 45, 46]. Additionally, clinoptilolite has been investigated as adsorbent for the removal of dyes, phenols and phenol derivatives from aqueous solutions, as summarized in Table 3.

The clinoptilolite was found to be effective in removing metal cations such as Al³⁺, Cd²⁺, Cu²⁺, Fe³⁺, Ni²⁺, Pb²⁺ and Zn²⁺ from copper mine wastewater [54]. It was observed that clinoptilolite has a high selectivity for heavy metal ions following the order: Pb²⁺ > Cd²⁺ > Cu²⁺ > Co²⁺ > Cr³⁺ > Zn²⁺ > Ni²⁺ > Hg²⁺ [20].

The ability of clinoptilolite to uptake Cu²⁺ from aqueous solution at different temperatures (25, 45 and 60 °C), at different pH values (1–4) and at different agitation speed (0, 100, 200, 400 rpm) was studied by Stylianou et al. [28]. The best efficiency of zeolite in removing copper ions was achieved at pH 4. Also, it has been observed that the agitation speed influences on the degree of removal. Specifically, the highest removal efficiency (37.3%) was achieved at an agitation rate of 400 rpm at 25 °C.

In addition to the above parameters, the value of Si/Al molar ratio as well as zeolite porosity also affects the cation exchange capacity. As Si/Al ratio is higher, the cation exchange capacity is lower [55, 56]. The ion exchange capacity can range from 0.6 to 2.3 meq/g [1]. As the specific surface area of the adsorbent is larger, the particles are smaller and the pore volume is larger. Most researchers have found that adsorbents with a smaller particle size cause a higher adsorption capacity due to larger specific surface area of the adsorbents [27, 30, 57-59], while others have noted that particle size does not affect the

adsorption process [60, 61]. The influence of adsorbent particle size on the removing of metal cations from aqueous solutions is shown in Table 4.

Table 3. Adsorption ability of natural zeolite

Adsorbent	Material	Adsorption (mg/g)	Reference
Clinoptilolite	Methylene blue	19.9	[43]
Clinoptilolite	Rhodamine B	12.4	
Clinoptilolite	Malachite green	19.7	[47]
Clinoptilolite	Toluidine blue O	33.0–58.7	[48]
Turkish clinoptilolite	Reactive Black 5	61.0	[49]
	Red 239	111.0	
	Yellow 176	89.0	
Turkish clinoptilolite modified with quaternary amine surfactant (HTAB)	Everzol Black	2.9	[50]
	Everzol Red	3.7	
	Everzol Yellow	7.6	
Australian natural zeolite	Humic acid	68.0	[51]
Jordanian zeolite	phenol	5.00–32.60	[52]
Mexican clinoptilolite	Phenol	6.50–12.76	[53]
	p-nitrophenol	0.58–01.44	

Table 4. Particle size effects of natural zeolite on adsorption of heavy metals

Particle size of clinoptilolite, (μm)	Metal adsorbed				Reference
	Pb ²⁺	Cd ²⁺	Zn ²⁺	Cu ²⁺	
160–600	1.29 mg/g	1.02 mg/g	/	/	[20]
600–1000	1.28 mg/g	1.01 mg/g	/	/	
1000–2000	1.27 mg/g	0.98 mg/g	/	/	
53–106	/	/	2.110 ^a mg/g	/	[58]
			4.406 ^b mg/g		
106–212	/	/	1.973 ^a mg/g	/	
			4.141 ^b mg/g		
212–300	/	/	1.715 ^a mg/g	/	[62]
			4.056 ^b mg/g		
< 90	/	/	/	89.34%	
90–325	/	/	/	82.64%	
325–400	/	/	/	78.33%	

/ – no data; a – Bigadic zeolite; b – Gordes zeolite

The pH value of the test solutions is an important parameter affecting the adsorption process. The activity of the adsorbent functional groups, then the type of complex formed between the metal ions and the adsorbent, depends on the environment in which the adsorption process takes place. Namely, in very acidic solutions, the protonation of the functional groups occurs, whereby they become positively charged and thus weak the attractive forces between the metal and

the adsorbent. Deprotonation of adsorbent functional groups occurs at higher pH values, and such groups more strongly attract metal ions forming a complex [63, 64]. As mentioned, the pH value of the solution is a significant parameter that affects the degree of metal ions removal. Motsi et al. [26], in their work, examined the effect of solution pH on the removal of Cu²⁺. According to the obtained results, the adsorption of copper ions from the solution increases with increasing pH of

the solution. In acidic solutions, H^+ ions are more efficiently adsorbed on zeolite active sites than metal ions. As the pH of the solution increases, the concentration of H^+ ions decreases and therefore more metal ions are adsorbed on to zeolite [30].

In the paper of Ding et al. [65], the possibility of using bentonite as adsorbent to remove copper ions was investigated. The study was conducted in the pH range from 1.0 to 9.0. It was observed that the pH of the solution affected the degree of Cu^{2+} adsorption. In the acidic environment, the lowest degree of adsorption is achieved, which can be

explained by the competition of Cu^{2+} and H^+ ions that tend to adsorb at the same zeolite active sites. A further increase in the pH value showed an increase in the degree of adsorption. At a pH range of 3.0 to 7.0, Na^+ , K^+ , Ca^{2+} and Mg^{2+} ions are exchanged with Cu^{2+} ions from solution, while adsorption of Cu^{2+} ions in the form of $Cu(OH)_2$ is observed at $pH > 8.0$.

Table 5 lists the different types of adsorbents that can be used in the heavy metal removal process and their adsorption capacity.

Table 5. Adsorption of heavy metal ions on various zeolites

Material	Metal	Adsorption	Reference	Material	Metal	Adsorption	Reference
Sardinian clinoptilolite	Cu^{2+}	0.340 meq/g	[66]	Clinoptilolite	Cu^{2+}	0.44 mg/g (pH 2.5)	[26]
	Cd^{2+}	0.05–0.190 meq/g				0.54 mg/g (pH 3.5)	
	Pb^{2+}	0.27–1.200 meq/g				0.54 mg/g (pH 4.5)	
	Zn^{2+}	0.100 meq/g			Zn^{2+}	1.60 mg/g (pH 2.5)	
Clinoptilolite	Cd^{2+}	0.12–0.180 meq/g	[67]			2.21 mg/g (pH 3.5)	
Turkish clinoptilolite	Co^{2+}	0.448 meq/g	[68]			2.14 mg/g (pH 4.5)	
	Cu^{2+}	0.282 meq/g			Mn^{2+}	0.37 mg/g (pH 2.5)	
	Zn^{2+}	0.268 meq/g				0.52 mg/g (pH 3.5)	
	Mn^{2+}	0.153 meq/g				0.52 mg/g (pH 2.5)	
Brazilian scolecite	Cr^{3+}	5.810 meq/g	[69]	Turkish natural clinoptilolite	Zn^{2+}	21.200 mg/g	[59]
	Ni^{2+}	2.080 meq/g		CC1		20.800 mg/g	
	Cd^{2+}	1.780 meq/g		CC2		22.200 mg/g	
	Mn^{2+}	4.000 meq/g		CC3		17.900 mg/g	
Mexican clinoptilolite	Cd^{2+}	0.15–0.42 mg/g	[70]	Natural zeolite	Zn^{2+}	13.060 mg/g	[72]
	Ni^{2+}	0.10–0.31 mg/g			Cu^{2+}	25.040 mg/g	
	Zn^{2+}	0.03–0.18 mg/g					
Sodium modified	Cd^{2+}	0.15–0.42 mg/g		Clinoptilolite	Pb^{2+}	101.000 mg/g	[73]
	Ni^{2+}	0.10–0.31 mg/g			Cu^{2+}	11.700 mg/g	
Mexican clinoptilolite	Zn^{2+}	0.03–0.18 mg/g			Cr^{3+}	6.800 mg/g	
Acid modified	Cd^{2+}	0.15–0.36 mg/g		Clinoptilolite	Cu^{2+}	9.544 mg/g	[44]
Mexican clinoptilolite	Ni^{2+}	0.10–0.28 mg/g					
	Zn^{2+}	0.03–0.18 mg/g					
Clinoptilolite	Al^{3+}	0.22 mg/g	[54]	Clinoptilolite VB	Cu^{2+}	7.750 mg/g	[74]
	Fe^{3+}	0.02 mg/g		Clinoptilolite I		8.510 mg/g	
	Cu^{2+}	0.13 mg/g		Clinoptilolite BM		11.180 mg/g	
	Zn^{2+}	0.16 mg/g					
Serbian clinoptilolite	Cu^{2+}	5.25 mg/g	[71]				

where: VB – clinoptilolite from the deposit Vranjska banja (Serbia), I – clinoptilolite from the deposit Igros, Kopaonik (Serbia), BM – clinoptilolite from the deposit Baia Mare (Romania)

Zdenska et al. [44] examined the possibility of using clinoptilolite to remove copper ions from synthetic solutions, at pH 3.5. It has been observed that, as the initial copper concentration in aqueous solution increases, adsorption capacity also increases, while the degree of adsorption decreases. In addition, the effect of the used adsorbent dosage on the degree of copper ion removal was investigated. As the dosage of adsorbent increases, the removal efficiency increases too. The maximum removal efficiency of Cu^{2+} was 61.8% at adsorbent dosage 10 g. This can be explained by an increase in the number of active sites where copper ion can be adsorbed. Similar conclusions were obtained by Courh [59], who investigated the adsorption of Zn^{2+} from $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ solution on a natural zeolite. Studies have shown that the initial concentration of heavy metal ions and the temperature of the solution have an important role in the adsorption process. As the temperature of the solution increases, the degree of adsorption of heavy metal ions rises too. At a temperature of 90 °C, the removal efficiency of Zn^{2+} was 77.9% [59].

Also, it has been observed that adsorption capacity and selectivity depend primarily on the type of zeolite used in various studies. In the work of Okia and Kavanage [75], two natural zeolites, clinoptilolite and chabasite, were used to remove Pb^{2+} , Cd^{2+} , Cu^{2+} , Zn^{2+} , Cr^{2+} , Ni^{2+} and Co^{2+} ions. Different selectivity of clinoptilolite and chabasite towards heavy metal ions was observed, namely $\text{Pb} > \text{Cu} > \text{Cd} > \text{Zn} > \text{Cr} > \text{Co} > \text{Ni}$ for clinoptilolite and $\text{Pb} > \text{Cd} > \text{Zn} > \text{Co} > \text{Cu} > \text{Ni} > \text{Cr}$ for chabasite.

In many studies, the adsorption process of single heavy metal ions has been analyzed, although wastewater most often contains multiple different ions between which there may be competition regarding zeolite binding. The natural zeolite, clinoptilolite, has been investigated for the removal of Pb^{2+} , Ni^{2+} , Cu^{2+} , and Zn^{2+} ions from one-component and multi-component solutions. The maximum adsorption capacities of clinoptilolite in one-component solutions were 76, 8, 7, and 5 mg/g for Pb^{2+} , Zn^{2+} , Cu^{2+} , and Ni^{2+} , respectively,

while these values were significantly smaller in the multi-component solution and were, respectively, 31, 3.5, 0.7 and 0.5 mg/g [76]. Additionally, Li et al. [54], in their study analyzed the ability of clinoptilolite as adsorbent to remove Al^{3+} , Cu^{2+} , Fe^{3+} and Zn^{2+} ions from natural mine waters. The duration of the experiments was 24h at pH 3.3.

In the test solution, the concentrations of heavy metal ions were 24 mg/dm³, 20.5 mg/dm³, 2.76 mg/dm³ and 40 mg/dm³ for Al^{3+} , Cu^{2+} , Fe^{3+} and Zn^{2+} respectively. Based on the obtained results, 90% Al^{3+} , 64% Cu^{2+} , 82% Fe^{3+} and 40% Zn^{2+} were removed from the test solution. The initial pH of the solution may affect the selectivity of the zeolite under test to certain metal ions in solution. Panajotova [27] concluded that, for the removal of copper from aqueous solutions on clinoptilolite from Bulgaria, the optimum pH value is 5. The natural zeolite from Jordan was also examined as adsorbent for the removal of Cu^{2+} and Cd^{2+} ions from synthetic two-component solutions, at pH = 6 [77]. Higher adsorption capacity for Cd^{2+} (25.5 mg/g) was achieved compared to Cu^{2+} ions (14.3 mg/g). Concerning the selectivity of natural zeolite tuff from Croatia for the removal of Zn^{2+} , Pb^{2+} and Cu^{2+} ions, Perić et al. [78] determined the following order: Pb^{2+} , $\text{Cu}^{2+} > \text{Zn}^{2+}$. Also, Sprynskyy et al. [30] investigated the removal of heavy metals (Ni^{2+} , Cu^{2+} , Pb^{2+} and Cd^{2+}) under static conditions from single- and multi-component aqueous solutions by raw and pretreated clinoptilolites. The obtained results revealed that there are no differences in removing ions as Cu^{2+} , Pb^{2+} and Cd^{2+} in single- and multi-component solutions. However, the adsorption of Ni^{2+} from multi-component solution is decreased in comparison to the single-component due to the competition with other cations in the solution. The maximum sorption capacity of Cd^{2+} was 4.22 mg/g at initial concentration of 80 mg/L and 27.7, 25.76 and 13.03 mg/g at 800 mg/L for Pb^{2+} , Cu^{2+} and Ni^{2+} .

Turkman et al. [79] studied the removal of heavy metals (Pb^{2+} , Cd^{2+} , Ni^{2+} and Zn^{2+}) in synthetic and real wastewater using activated and non-activated natural clinoptilolite. The researchers obtained better results with activated zeolite than

with non-activated clinoptilolite. Furthermore, the adsorption ability of Bigadic and Gordes zeolite in order to uptake Cd^{2+} and Ni^{2+} ions was compared. The same was done in the investigation by Oren and Kaya [80]. By using Gordes zeolite the removal efficiency for Cd^{2+} was 46% while Cd^{2+} was not removed effectively by Bigadic zeolite after 90 min contact time. For Ni^{2+} removal, the obtained efficiency was 40% for both Bigadic and Gordes zeolite. Additionally, Oren and Kaya [80] in their study showed that the removal efficiency of Gordes zeolite is twice higher than that of Bigadic zeolite. In this paper, it has been examined the removal of Cu^{2+} and Cd^{2+} ions. The initial metal concentration was in the range of 100–600 mg/L at pH 6, the adsorbent dosage was 5.0 g. The capability of adsorption was found to be stable and reached the maximum value at concentration 100 mg/L. The percentage of Cu^{2+} and Cd^{2+} removal decreases by increasing initial metal concentration. For Cd^{2+} and Cu^{2+} the adsorption capacities were 25.9 and 14.3 mg/g [77].

The sorption behavior of natural zeolite (clinoptilolite) for the removal of metal ions Cd, Cu, Ni, Pb and Zn has been studied. Metal concentration in solution ranging from 50–300 mg/L. Clinoptilolite sorbed approximately 32, 75, 28, 99 and 59% of the added Cd, Cu, Ni, Pb and Zn metal, respectively. The selectivity of clinoptilolite can be given as: $\text{Pb} > \text{Cu} > \text{Zn} > \text{Cd} > \text{Ni}$ [81].

Baker et al. [82] investigated the uptake capacity of heavy metals ions Cr^{3+} , Cd^{2+} , Cu^{2+} and Pb^{2+} using Jordanian zeolite at solution pH 3.0 (adsorbent dosage 5.0g). The Jordanian zeolite presented high selectivity for the discharge of Pb^{2+} as approximately 98% achieved within 90 minutes, while 96% of Cr^{3+} , Cu^{2+} and Cd^{2+} removed within 5h.

Alvarez-Ayuso et al. [56] compared the sorption behavior of natural clinoptilolite and synthetic NaP1 zeolites for Cr^{3+} , Ni^{2+} , Zn^{2+} , Cu^{2+} and Cd^{2+} in regard to the purification of wastewater. The obtained results revealed that the synthetic zeolite has 10 times greater sorption capacity than natural zeolite. The adsorption capacity of synthetic zeolite was

43.6 mg/g, 20.1 mg/g, 32.6 mg/g, 50.5 mg/g and 50.8 mg/g for Cr^{3+} , Ni^{2+} , Zn^{2+} , Cu^{2+} and Cd^{2+} , respectively.

In order to improve sorption abilities of raw natural zeolites, they can be modified by chemical treatments. Therefore, Cerjan-Stefanovic et al. [37] investigated the effect of pre-treatment of clinoptilolite from Serbia on the efficiency of Zn^{2+} removal. For the pre-treatment, solutions of NaCl or CaCl_2 were used. It was observed that the pre-treatment of zeolite enhanced the adsorption capacity of Zn^{2+} .

The ability of acid-modified clinoptilolite to remove Pb^{2+} from the aqueous solutions was the goal of this paper. The clinoptilolite was modified using different concentrations of H_2SO_4 solution. The maximum uptake for Pb^{2+} was higher than 95% at pH solution around 5. The lower removal capacity of Pb^{2+} at pH 2 is due to the lower selectivity of clinoptilolite in acidic solutions and the strong competition between Pb^{2+} and H^+ for the exchangeable sites on zeolite framework. The decrease in Pb^{2+} sorption at higher pH values can be explained by the formation of hydrolyzed Pb^{2+} species which could precipitate [18].

In this paper, the removal of Cd^{2+} , Ni^{2+} and Zn^{2+} using natural (clinoptilolite rich tuff from Mexico, ZPCli), sodium modified (ZPCliNa) and acid modified zeolites (ZPCliH) has been investigated. It was observed that after the treatment with NaCl, sodium content increased 6 times whereas the content of other exchangeable cations in clinoptilolite decreased, especially for K^+ and Ca^{2+} . Further, the treatment of zeolite with acid solution (H_2SO_4), sodium was totally eliminated while the content of other cations decreased due to the introduction of H^+ in zeolite. According to the obtained results for Ni^{2+} and Zn^{2+} , it is assumed that sorption occurs under the following mechanism: at initial pH 4, the precipitation on the surface of ZPCli and ZPCliNa and ion exchange with native ions from zeolite with Ni^{2+} and Zn^{2+} from the solution. The selectivity of natural and modified zeolite follows the order: $\text{Cd}^{2+} > \text{Ni}^{2+} > \text{Zn}^{2+}$ [70].

In accordance with investigations (Table 6), it is concluded that the adsorption of metal cations

by using zeolite obeys pseudo-first-order or pseudo-second-order model.

Table 6. Kinetic parameters for metal cations removal

Zeolite	Cation	Isotherm model	Reaction model	k_1 (1/min)	k_2 (g/mg·h)	Reference
Jordanian zeolite	Cd ²⁺	Langmuir model	Pseudo-second		2.321	[77]
	Cu ²⁺				1.716	
ZPCli	Cd ²⁺	Langmuir model	Pseudo-second		pH 2	[70]
ZPCliNa					pH 10	
ZPCliH					18.188	
					845.922	
					29.430	
					346.057	
					16.420	
					16.699	
ZPCli	Ni ²⁺	Langmuir model	Pseudo-second		27.102	[70]
ZPCliNa					284.240	
ZPCliH					97.612	
					195.280	
					29.782	
					16.962	
ZPCli	Zn ²⁺	Langmuir model	Pseudo-second		30.280	[70]
ZPCliNa					11.083	
ZPCliH					28.323	
					8.534	
					62.876	
					4.040	
Zeolite	Cu ²⁺		Pseudo-second		0.0016 g/mg·min	[45]
Clinoptilolite	Cu ²⁺	Langmuir model	Pseudo-second		0.00113	[30]
	Pb ²⁺				0.00077	
	Ni ²⁺				0.00410	
	Cd ²⁺				0.02260	
Clinoptilolite	Cd ²⁺		First-order	0.01370		[83]
	Pb ²⁺			0.05320		
	Ni ²⁺			0.01580		
	Cu ²⁺			0.02000		
	Zn ²⁺			0.02100		
Clinoptilolite	Pb ²⁺		First-order	0.00035		[47]

4. Conclusion

Alcaline, chabazite, clinoptilolite, erionite, mordenite and phillipsite are the most common natural zeolites. Among them, clinoptilolite was the mostly investigated as adsorbent for wastewater purification. Due to significant adsorptive properties they can be used for the removal of different pollutants such as heavy metals, oil and organic contaminants. Natural zeolites are low cost adsorbents with excellent selectivity for various cations and good ion exchange capacity. The presence of non-toxic exchangeable cations in zeolite structure such as: Na⁺, K⁺, Mg²⁺ and Ca²⁺ makes the zeolites suitable for wastewater treatment. In order to improve the sorption

properties, natural zeolites are modified. In accordance with that, researchers obtained a few times greater sorption capacities in comparison to the raw zeolite.

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PRIMENA PRIRODNIH ZEOLITA U TRETMANU OTPADNIH VODA – PREGLEDNI RAD

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Izvod

Zagađenje životne sredine je značajan svetski problem. Velike količine zagađujućih supstanci, koje nastaju tokom različitih industrijskih procesa doposevaju do ekosistema. Na taj način teški metali, ulja i druga organska jedinjenja zagađuju vodene sisteme. Poznato je da su teški metali toksični po životnu sredinu. Osim toga, teški metali nisu razgradivi i mogu dospeti do živih organizama preko lanca ishrane. Među različitim metodama koje se koriste u tretmanu otpadnih voda, adsorpcija zauzima značajno mesto zbog jednostavnosti izvođenja procesa i efikasnosti u uklanjanju zagađujućih supstanci. Prirodni zeoliti iz različitih ležišta imaju dobru selektivnost prema jonima teških metala. Pored toga, prirodni zeoliti su ekološki prihvatljivi materijali, jeftini si i poseduju veliki kapacitet jonske izmene.

Ključne reči: Tretman otpadnih voda; Zagađujuće supstance; Prirodni zeolit; Adsorpcija.