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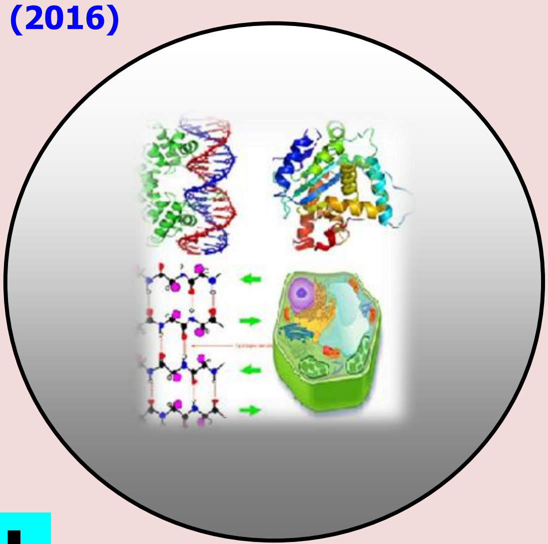

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N.O. Burkiashvili
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RESEARCH PAPER

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Combined Filters on the Bases of Natural Adsorbents for Sewage Water Treatment

Osipova N.A, Kvernadze T.K. and Burkiashvili N.O.
Peter Melikishvili Institute of Physical and organic Chemistry, Iv Javakhishvili Tbilisi State University, Georgia

ABSTRACT

Possibility of application of Georgian natural sorbents - zeolites (clinoptilolite and lomontite) has been studied in order to reduce mineralization in waste and drainage waters and remove toxic impurities by improving adsorption activity of the sorbents. For this purpose, combined filter columns filled with the hydrogen forms of the above zeolites were prepared. The sample sewage water was taken from the vicinity of Bolnisi Madneuli Ore-Dressing and Processing Enterprise (Georgia). The results of the experiment have shown the perspectives of application of the above zeolutes in their H^+ form as filtering materials for wastewater treatment. The obtained results are relevant for both practical and economical point of view and the prepared filtering columns might be used in the field of environmental protection.
Key words: Natural Zeolites, Sewage Water, Adsorbents, Ion Exchangers and Combined Filters.

INTRODUCTION

Environmental problems have direct impacts on both immune system and genetic code structure of a living organism. Forty percent of the world's population get sick and die because of the environmental pollution, namely due to the pollution of water, soil or air, as the harmful substances are taken by humans along with food and water. Waste waters are one of the most active environmental pollutants especially in our industrial age when the amount of industrial waters increases rapidly and the risk of their entering into drinking water supplies rises. Against this background, it is difficult to overestimate the relevance of water purification, especially in the regions where ore-dressing and chemical enterprises are located and the risk of contamination of waste waters by harmful heavy metals is higher due to the technological processes in those enterprises. Over the years, this issue has been very acute for Bolnisi Region (Georgia), where Madneuli ore-dressing and processing enterprise has been functioning.

One of the steps in the various technological schemes for sewage water treatment is its filtration. In some works, application of sorbents on the bases of activated charcoal, quarts sand, synthetic and natural zeolites are suggested for treatment of sewage and drainage waters (Vatin *et al.* 2013; Tsitsishvili *et al.* 1985).

The vast majority of filtering materials used in water treatment constructions are synthetic while widespread, easily available, and effective natural filtering materials such as zeolites have been absolutely vainly left beyond the attention (Getmantsev *et al.* 2012).

Nano-dimensional structure of natural zeolites conditions certain specific properties for their practical application. They are leading minerals for waste or drainage water treatment due to their sorptive, selective ion exchange, molecular sieve and catalytic properties. Well developed specific surface, good adhesion, unique adsorption and ion exchange properties of zeolites enable them to extract certain amount of colloidal and dissolved organic or inorganic substances including ammonium ions, heavy metal cations and radionuclides from contaminated waters. Zeolites, as cationic exchangers, extract heavy metal cations from water and compared to synthetic resins they have higher selectivity to cesium and strontium ions (Pimneva *et al.* 2011; Alikov *et al*. 2009; Compos 2009).

Natural zeolites outperform conventional granular media, are cost-effective, abrasion resistant, non-toxic and environmentally friendly, especially for treatment of such special wastewater streams as acid mine drainage, landfill leachate, nuclear fallout, and urban runoff (Delkash *et al.* 2015).

Zeolites are aluminosilicates with the general formula $M_{x/n}[Al_xSi_yO_{2(x+y)}]\cdot mH_2O$, where M stands for metal (usually Na, K, Ca, Mg) in its ionic form M^{+n} that compensates the negative charge of the aluminum in crystal lattice. Each silicon and aluminum atom is covalently bonded with four oxygen atoms (SiO_4 and AlO_4) forming open, "framework" structure with cages and channels. For example, the most commonly used zeolite clinoptilolite (HEU) forms a monoclinic crystal structure with channels shown in Figure 1. "wide" channels formed by 10-membered rings have sizes 0.31 x 0.75 nm; "narrow" channels formed by 8-membered rings have sizes 0.28 x 0.47 and 0.36 x 0.47 nm. Presence of channels and cavities conditions molecular-sieve, sorption and other properties of zeolites (Tsitsishvili *et al.* 2018).

Among many natural zeolites identified in the world, clinoptilolite, mordenite, phillipsite, analcime and laumontite are very common.

a. b.

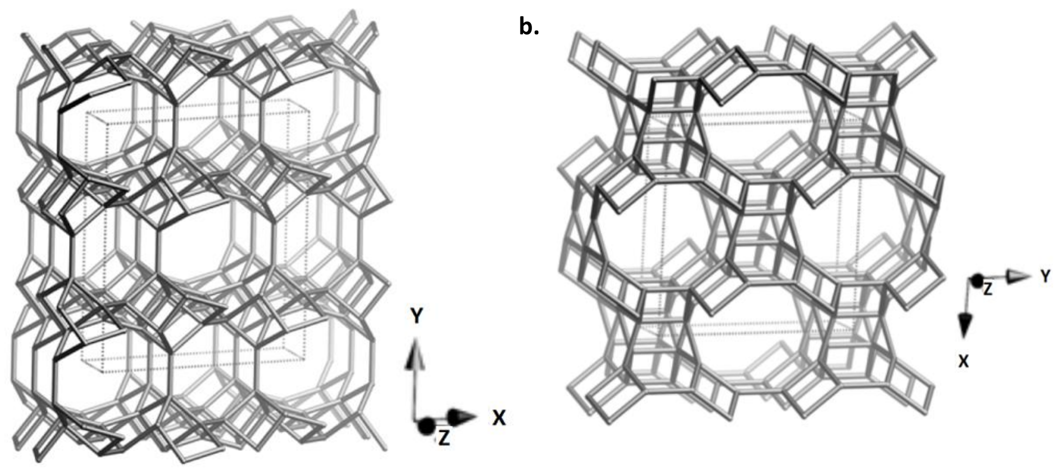

Figure 1. Zeolite framework viewed along [001]: a. HEU b. LAU

Adsorption selectivity of Na-clinoptilolite for heavy metal ions (Mihaly-Cozmuta *et al.* 2014) is strongly influenced by the pH of the contact solution, dehydration energy and diffusion coefficient of ions. At pH>4 selectivity is expressed by the series: $Pb^{2+}>Cd^{2+}>Cu^{2+}>Zn^{2+}>Mn^{2+}>Co^{2+}>Ni$. The ion exchange capacity of Ca,Na-laumontite of Georgia decreases in the following series: $Cs^{+}>Rb^{+}>NH_4^{+}>Li^{+}$, and $Ba^{2+}>Sr^{2+}>Ca^{2+}>Mg^{2+}$ (Urotadze *et al.* 2016).

The adsorption characteristics of any zeolite are defined by its chemical and structural properties (the Si/Al ratio, nature of compensating metal ions, their charge and location, etc.) that can be changed by chemical treatment to improve separation efficiency of raw natural zeolite. Acid washing of natural zeolite removes impurities that block the pores/channels, and progressively eliminate metal ions to change into H-form characterized by lower ion exchange capacity, but does not introduces extraneous metals into water under purification.

MATERIALS AND METHODS

The purpose of this work has been investigation of feasibility of application of Georgian natural zeolites, clinoptilolite and lomontite (Hekordzula and Botanikuri regions) for decreasing mineralization of sewage waters and extraction undesired toxic impurities by improving adsorption activity of sorbent.

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In order to achieve the goal, combined filtration column (5cm x 20cm) filled with the following adsorbents was prepared: 1st layer (5cm) - quartz sand washed with 1N HCl and heated at 200^{0}C for removing suspended materials and turbidity; 2nd layer (5 cm)- clinoptilolite (granules of 2-2.5 mm) washed with 1N HCl and heated at 200-250^{0}C; 3rd layer (5 cm) – lomontite (granules – 1.5-2 mm) prepared the same way; 4th layer (5cm)- natural clinoptilolite (0.5-1mm granules). To prepare granules of hydrogen forms of clinoptilolite and lomontite containing about 90% crystalline phases were placed in a round-bottom flask and added 1N HCl to it in amount of 10 volume of the solution per volume of the sorbent. The flask was connected to a backflow condenser, placed in a sand bath and boiled the content of the flask for 4 hours. According to this procedure, 200 cm^3 of sorbent (granules of 0.5-2.5 mm) was treated. The prepared sorbent was loaded into a glass column. The wastewater sample in amount of 4 liter flew through the column at a speed of 5 ml/min. Determination of cations and anions in both wastewater and filtrate was carried out on a chromatograph SHIMADZU CDD-10Avp/10ASP.

Tables 1 and 2 show the chemical composition of the studied waters before and after treatment.

Table 1. Chemical composition of sewage water.

Before treatment by the sorbent					
Cations			Anions		
	mg/dm^3	mg-eq/dm^3		mg/dm^3	mg-eq/dm^3
Ca^{2+}	215.0000	10.7285	Cl^-	1611.37	45.4509
Mg^{2+}	208.0000	17.1193	Br^-	1.8798	0.0235
Na^+	655.0000	28..4909	I^-	2.9277	0.0232
K^+	8.2100	0.2099	HCO_3^-	462.988	7.5888
Co^{2+}	0.0500	0,0017	CO_3^{2-}	2.6517	0,0442
Ni^{2+}	0.0600	0,0020	HSO_4^-	0.0000	0,0000
Sr^{2+}	3.4000	0.0785	SO_4^{2-}	684.1150	2.9576
Cs^+	0.0000	0,0000	HS^-	0.0000	0,0000
Li^+	0.1000	0.0145	NO_2^-	0.0329.	0.0011
Fe (total)	2.2300	0.0796	NO_3^-	0.1500	0.0024
Cu^{2+}	0.7030	0.0221	$H_2PO_4^-$	0.0042	0.0000
Pb^{2+}	0.4900	0.0047	HPO_4^{2-}	0.0478	0.0005
Mn^{2+}	0.3100	0.1130	PO_4^{3-}	0.0478	0.0005
Zn^{2+}	0.0600	0/0180			
total	1093.6130	56.8480	total	2766.2131	56.1020

Table 2. Chemical composition of sewage water.

After treatment by hydrogen form of clinoptilolite					
Cations			Anions		
	mg/dm^3	mg-eq/dm^3		mg/dm^3	mg-eq/dm^3
Ca^{2+}	129.0000	6.4371	Cl^-	975.2689	27.5088
Mg^{2+}	125..0000	10.2881	Br^-	1.1279	0.0141
Na^+	357.0000	15.5286	I^-	1.6102	0.0
K^+	2.9556	0.0756	HCO_3^-	0.0000	0.0000
Co^{2+}	0.0200	0.0007	CO_3^{2-}	0.0000	0.0000
Ni^{2+}	0.0300	0.0010	HSO_4^-	0.0000	0.0000
Sr^{2+}	0.3862	0.0089	SO_4^{2-}	410.469	4.2730
Cs^+	0.0000	0.0000	HS^-	0.0000	0.0000
Li^+	0.0500	0.0071	NO_2^-	0.0329	0.0007
Fe (total)	0.1800	0.0064	NO_3^-	0.1100	0.0018
Cu^{2+}	0.2530	0.0090	$H_2PO_4^-$	0.0032	0.0025
Pb^{2+}	0.0000	0.0000	HPO_4^{2-}	0.0000	0.000
Mn^{2+}	0.0500	0.0111	PO_4^{3-}	0.0000	0.000
Zn^{2+}	0.0120	0.0015			
total	615.0842	32.37	total	1388.6221	31.8136

RESULTS

It has been established that the use of hydrogen forms of lomontite and clinoptilolite significantly improves cationic composition of the filtrate. The obtained data revealed that content of Mg^{2+} and Ca^{2+} cations decreases by 40% and that for SO_4^{2-} and Cl^- ions by 40% and 39% correspondingly. Significantly decreases the content of such undesirable ions as Pb^{2+} Fe^{2+} (total), Sr^{2+}, Mn^{2+}, Zn^{2+}, Cu^{2+}, Co^{2+}, and Ni^{2+} (about 100-50 % respectively).

Based on the results, it can be concluded that the application of hydrogen forms of local natural zeolites (lomontite and clinoptilolite) can be used as a relatively inexpensive and effective filtering material for wastewater treatment.

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Corresponding author: Burkiashvili N.O., Peter Melikishvili Institute of Physical and organic Chemistry, Iv Javakhishvili Tbilisi State University Georgia
Email: nINOBURKI60@gmail.com

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