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Application of Natural Zeolites on Wastewater Treatment

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ABSTRACT
The purpose of this study is to experimentally investigate the SAR (Sodium Adsorption Ratio) and TDS (Total Dissolved Solids) reduction in Reverse Osmosis (RO) concentrate using two types of natural zeolites. In order to reduce salinity of wastewater, experiments were carried out by varying the type of zeolite, concentration of zeolite and residence time. The results show that both zeolites can lower the SAR and TDS of wastewater; however, Rhyolitic tuff is more effective than clinoptilolite. It is observed that the concentration of zeolite has not significant effects on wastewater treatment, so, using the lowest level of selected concentration reduces the cost of desalination. In addition, the effect of residence time is negligible. The experimental set up shows that the SAR reduction slop is higher than TDS.

Key words: Salinity, RO concentrates, zeolite, irrigation

INTRODUCTION
Reverse Osmosis (RO) is a well-known membrane technology for treatment of wastewater and production of potable or irrigation water. The main challenge of RO is the concentrated waste produced during the process (Subramani and Jacangelo, 2014). Several materials and technologies are employed for further treatment of reverse osmosis concentrate in order to reduce the rejected volume or treat it to reuse in other applications.

Jing *et al*. (2009) evaluated the cost of the TDS removal by electricity and investigated ion treatment of wastewater produced in polymer-flooding. They revealed that electricity greatly increases the energy consumption; however, the flow rate effects on energy consumption are negligible. Mohammadesmaeili *et al*. (2010) evaluated the RO concentrate treatment using lime-soda softening process and showed that efficiency of seawater desalination using lime-soda is 80-90%. Nkwonta and Ochieng (2010) designed a pilot plant for wastewater pretreatment using charcoal and gravel. They demonstrated that filters roughness enhances efficiency of pretreatment process for mine water. They also showed that in general, charcoal outperforms gravel.

Mrayed *et al*. (2011) evaluated the effectiveness and flexibility of Nano Filtration (NF) in producing irrigation water from reverse osmosis concentrates. They showed that Poly Acrylic Acid (PAA) increased the ion adsorption capacity of KOCH membrane; however, it did not affect rejected volume of ions by the NF270 element. Al-Rawajfeh *et al*. (2011) investigated the influences of three layered porous media composed of Tripoli, Pozzolana and Feldspar on seawater pre-treatment. They showed that the composite media has a significant capacity for adsorbing the seawater ions. Ghyselbrecht *et al*. (2012) investigated the treatment of RO concentrated volume by a combination of an electro dialysis and willow field. They showed that the combination of electrodialysis and willow field is appropriate for treating RO concentrate. Tabatabaei *et al*. (2012) investigated the clinoptilolite abilities on decreasing chemical and biological index of the compost factory's leachate.

They showed that clay loam soil texture with pre-treatment is effective in treatment of waste water. Aslania *et al*. (2013) experimentally investigated the influence of Zeolite and Activated carbon on decreasing the total dissolved solids in water. Their results indicated that the efficiency of various treatment methods depend on the concentrations of chemical contaminants in water.

Most industries, specially oil and gas refineries, utilize RO plants to provide highly purified water for different plants and equipment such as reboilers. The main problem associated with RO plants is the concentrated volume, i.e., when concentrated stream of RO is discharged back into the source of water or over land surface, it can cause many environmental damage. Sodium Adsorption Ratio (SAR) is a criterion of the suitability of water for irrigation. In order to reuse RO concentrate, its salinity must be reduced to prevent long term damages to environment, soil and underground water around the plant.

The main purposes of this study are treating and reusing the RO concentrate. For this goal, application of natural zeolites in the SAR and TDS rejection is investigated. The objective of the work is threefold: Assessing the impact of two types of natural zeolites, the residence time and the zeolite concentration.

MATERIALS AND METHODS

Reverse osmosis is a water purification technology based on membranes. The RO is most commonly known for production of drinking water from seawater. The main problem of reverse osmosis is the salty by product.

In this study, natural zeolites are chosen to adsorb the salty particles in the water. Zeolites have high cation exchange capacity and ion selectivity, which make them appropriate for removal of different ions from water and wastewater.

Methodology: In order to study SAR and TDS as dependent covariates, experiments were designed in three categories:

- Variety of natural zeolite
- Concentration of zeolite
- Residence time of zeolite in RO concentrate

For this purpose, 2 types of natural zeolites in 24 samples (12 clinoptilolite samples and 12 Rhyolitic tuff samples) are tested and results statistically studied. For the second parameter, the concentration of zeolite has three levels: 5, 7 and 10 g of each zeolite per 50^{CC} of RO concentrate. The third parameter is the contact time with three levels: 24, 48 and 72 h. The experiments are designed in batch and well mixed systems. After the mentioned contact time, the mixtures are filtered out. Then the solutions are provided to measure SAR and TDS values. Table 1 summarizes the experimental design of the study.

Table 1: Variables and values used for the design of experiments

Factor	No. of level	Levels
Zeolite	2	Clinoptilolite
		Rhyolitic tuff
Concentration (gr per 50^{CC})	3	5
		7
		10
Residence time	3	24
		48
		72

Materials: The zeolites used as an adsorbent in the current study are obtained from Western Azarbaijan of Iran. The samples are characterized using X-ray diffractometer and chemical analysis. Figure 1 and 2 show X-Ray Diffraction Patterns for clinoptilolite and Rhyolitic tuff, respectively. The chemical composition of the zeolites used in the study is shown in Table 2.

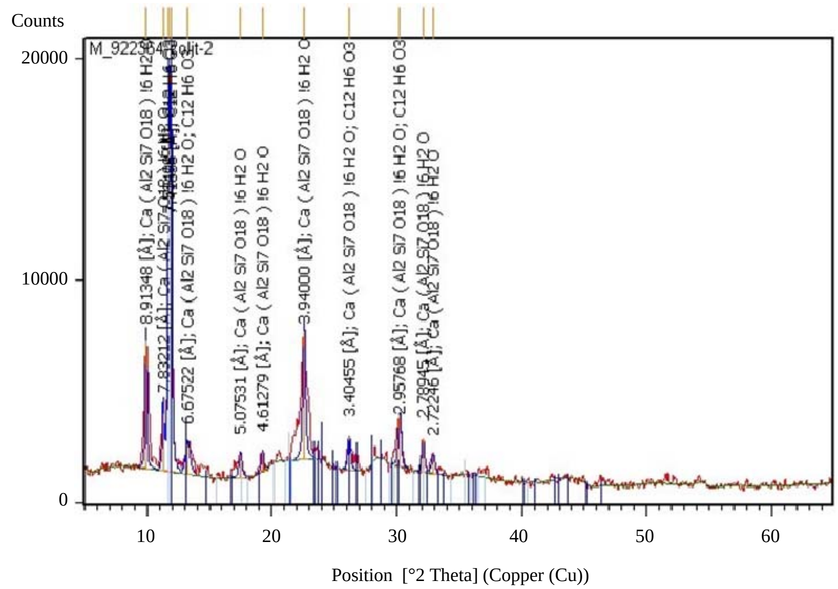

Fig. 1: Representative X-Ray diffraction patterns for Clinoptilolite

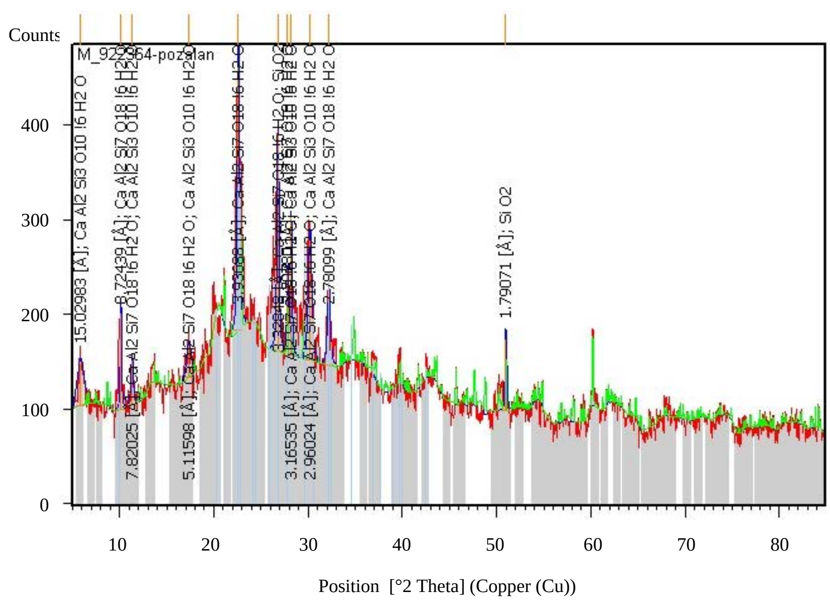

Fig. 2: Representative X-Ray diffraction patterns for rhyolitic tuff

Table 2: Chemical analysis of zeolites

Sample (%)	SiO_2	Al_2O_3	Na_2O	MgO	K_2O	TiO_2	MnO	CaO	P_2O_5	Fe_2O_3	SO_3	L.O.I
Clinoptilolite	68.45	9.65	2.59	1.06	1.28	0.21	0.02	1.45	0.03	1.25	0.00	13.77
Rhyolitic tuff	61.46	10.43	0.86	2.31	1.52	0.30	0.03	4.55	0.06	2.28	0.00	15.90

RESULTS

In order to study the SAR and TDS values, each zeolite is added to 50^{cc} of RO concentrate. The three samples of 9 clinoptilolite/Rhyolitic tuff samples are stirred for 24 h. The residence time of the second set of clinoptilolite/Rhyolitic tuff samples is 48 h and the contact time of third set is 72 h. The next sections present the results of each experiment.

Zeolite effects on SAR reduction: Figure 3a-c illustrated the effects of clinoptilolite and Rhyolitic tuff on SAR reduction after three residence time: 24, 48 and 72 h, respectively. As shown in Fig. 3. The SAR value after 24 h reduce from 137.4-72.3, 72.5 and 72.1 for 5, 7 and 10 g of clinoptilolite per 50^{cc} RO concentrate., respectively. In addition, the SAR value after 48 h contact time, decrease from 153.9-73.0 for 5 g clinoptilolite per 50^{cc} RO concentrate. The SAR values for 7 and 10 g of clinoptilolite per 50^{cc} RO concentrate are 76.0 and 73.3, respectively (Fig. 4). Comparison of Fig. 3a-c show that the effect of concentration of clinoptilolite on SAR rejection is negligible.

Figure 3a-c show the SAR values after contact with Rhyolitic tuff. The figures illustrate that the Rhyolitic tuff significantly boosts the SAR rejection. In the other word, Rhyolitic tuff performs

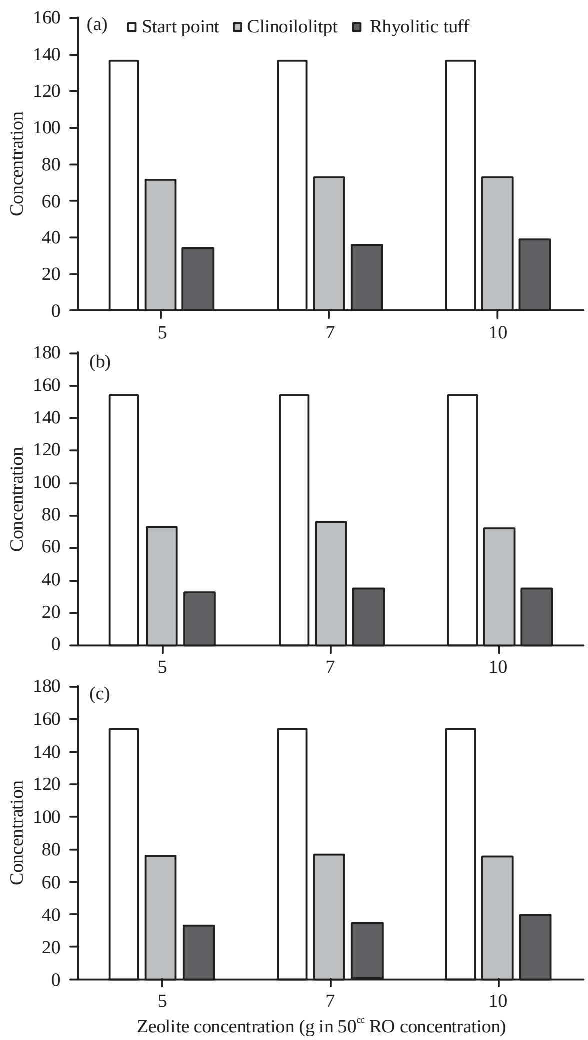

Fig. 3(a-c): Zeolite effects on SAR reduction duration (a) 24 h, (b) 48 h and (c) 72 h extremely well in removing Na^+ from RO concentrated samples, i.e., it decreases sodium adsorption ratio from 153.9-33.5, 35.5 and 39.3 for 5, 7 and 10 g zeolite per 50^{cc} RO concentrate, respectively. In addition, it is clearly seen from the figures, SAR levels are approximately independent of Rhyolitic tuff concentration.

Zeolite effects on TDS reduction: Figure 4a-c illustrate the variation of TDS for three different contact times with two types of zeolite. As it is seen from the figures, TDS levels are approximately independent of concentration of clinoptilolite, so, an increase in the clinoptilolite concentration does not significantly affect the TDS reduction. In addition, the figures show that the efficiency of Rhyolitic tuff is higher than clinoptilolite in TDS rejection; however, the TDS value is also independent of Rhyolitic tuff concentration.

DISCUSSION

It is obvious that natural zeolites are suitable for reducing the SAR from wastewater. Comparison of two types of zeolites illustrated that rhyolitic tuff is more effective than

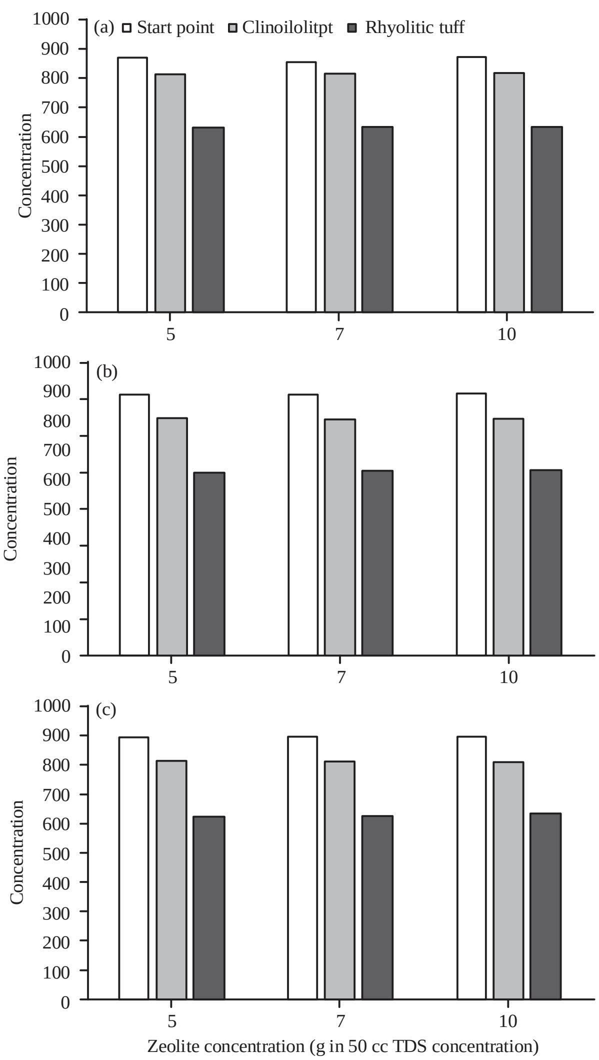

Fig. 4(a-c): Zeolite effects on TDS reduction duration (a) 24 h, (b) 48 h and (c) 72 h

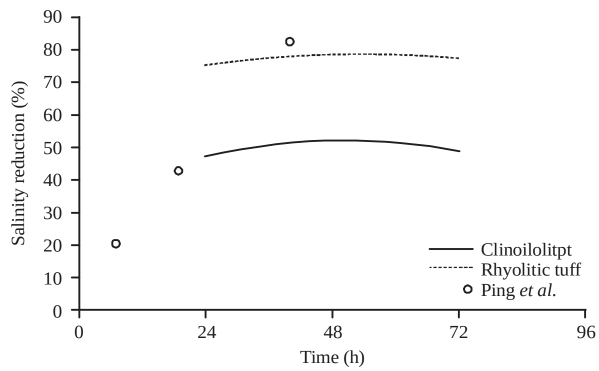

Fig. 5: Effects of residence time on SAR reduction

Table 3: Comparison of the results with those in the literature (Bunani *et al*., 2015)

Method	Start point	End point	SAR reduction (%)	SAR rejection value (meq L^{-1})	Operating pressure (bar)
Rhyolitic tuff without pretreatment(7 g/50^{cc})	154.0	40.50	73.70	113.50	1
AD-SWRO membrane (50% feed, 50% permeate)	80.1	10.00	87.12	70.10	10
AD-SWRO membrane (50% feed, 50% permeate)	80.1	9.92	87.23	70.18	20

clinoptilolite. Furthermore, pretreatment of zeolites reduces the efficiency of separation process. So, rhyolitic tuff without pretreatment is the best choice for removing the SAR value from wastewater. Table 3 depicts the comparison of the results of present study with the results of Bunani *et al*. (2015). It is clear that wastewater treatment using Rhyolitic tuff requires to significantly low operating pressure than reverse osmosis process. In addition, SAR reduction value using, Rhyolitic tuff is more than the value obtained by membrane, however, the SAR value of treated wastewater by Rhyolitic tuff is more than the limit of water salinity for irrigation. So, it is proposed that the other treatment sections such as RO section will be added to the process after treatment with zeolite.

Figure 5 depicts the effect of time on wastewater treatment. It is clear that Rhyolitic tuff is more effective than clinoptilolite; however, the effect of contact time on SAR reduction by two types of natural zeolites is negligible. Comparison of result with the results of Ping *et al*. (2015) show that microbial treatment is more sensitive to the residence time than zeolites. In the other hand increasing the contact time increases significantly SAR reduction value; however, the SAR start point for microbial treatment and the present study is respectively 13.47 and 153.9. So, despite of a moderate SAR reduction by zeolites, it is considered to be applicable in high values of SAR.

In order to TDS removal, it seems that Lee *et al*. (2008) method in using polymer-carbon composite membranes and Wang *et al*. (2014) in using sequencing batch reactor containing halophilic bacterial community are more effective than treatment by zeolites.

CONCLUSION

In this study, in order to reuse in irrigation, the application of natural zeolites is investigated for salinity reduction of RO concentrate. It is seen that clinoptilolite lowers the water salinity; however, Rhyolitic tuff is more effective than clinoptilolite in water treatment. It is observed that for both zeolites effects of concentration on SAR and TDS reduction are negligible. In addition, it can neglect from the effect of residence time on salinity reduction.

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