RESEARCH ARTICLE

JORDANIAN NATURAL ZEOLITE USE AS SORBENT FOR REMOVAL ACETIC ACID FROM AQUEOUS SOLUTIONS

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ARTICLE INFO	ABSTRACT
	Removal of acetic acid from wastewater has been investigated using adsorbents based on Jordanian natural zeolite (JNZ-1, JNZ -2, JNZ -3 JNZ-4) or (JNZ1-4) .The effects of acetic acid concentration, adsorbents dosage, contact time, temperature, were studied in batch experiment. The removal is most effective with activated (Jordanian natural zeolite). A maximum removal of around 41.82 was achieved under the conditions of the experiments.

INTRODUCTION

Jordanian natural zeolites are good potential material for wastewater treatment. It is due to the advantages of low cost, ion- exchange and a adsorption capability of the natural zeolites. It can also be modified and regenerated. The concept or removal of acetic acid from wastewater streams is gaining momentum presently due to economic and environmental reasons. Another new application is found in municipal wastewater treatment plants. Removal of acetic acid from wastewater can be undertaken through various methods, viz, distillation, solvent extraction and adsorption. With such law concentration streams, distillation would not be economically viable. The adsorption of acetic acid on activated carbon has been well documented (Kippling, 1965; Flanigen, 1991). However, the adsorption is poor compared to other larger organic compounds, such as, phenol (<10% -vs. 20% organic weight/carbon weight) Since operational costs are generally related directly to zeolites consumption, her is need for emphasis on the improvement of the adsorption characteristic of adsorbents based on Jordanian natural zeolite (JNZ).

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In the present studies, therefore, an attempt has been made to prepare adsorbents from natural materials, such as Jordanian natural zeolites and use them for the removal of acetic acid from dilute aqueous waste. Natural zeolites are able to bind significant amounts of ammonia, helping to remove nitrogen from these streams. Finally, it should be mentioned that natural zeolites may be used as additives for fertilizers and for soil conditioning. It is believed that they may retain desirable cations in the soil for a longer period of time. Their high adsorption capacity may prove to be useful for carriers of pesticides and fungicides (Flanigen, 1991). Wastewater streams DMT productions units, Wood pulping units and petrochemical industries are prime sources of acetic acid with concentration ranging 0.5 to 6.0wt%. Corrosion in water structure. (Rodhe and Herraera, 1988), odor problems (Lund 1971), water toxicity and water treatment problems are some results of the presence of acetic acid in water. Wastewater containing acetic acid cause treatment problem when biologically oxidized, as this converted into activated sludge having poor settling characteristic and difficult to separate. Presence of acetic acid in aquatic environment results in an increased oxygen demand. A number of studies have been reported with regard to the adsorption equilibrium of dye removal processes using various adsorbents.

In most adsorption systems dyestuffs-adsorbents Langmuir (El-Geundi,1993; McKay, 1982, 1998; McKay *et al.*, 1980; Meshko *et al.*,1999), isotherms have been applied to describe equilibrium between liquid and solid phases. The natural zeolites are cheap materials which are readily available in large quantities in many parts of the world. These have become especially important in the purification of water (10). Being a natural zeolite with a representative unit-cell formula of (Na3K3)(Al6Si30O72)•24H2O, a void volume of 34%, channel imensions of 3.9 × 5.4 A and a cation-exchange capacity of 2.16 mequiv/g, clinoptilolite seems to be the most attractive material for the removal of NH4 + ions from drinking water and wastewater because of its NH4 + ion selectivity and good performance towards ion sorption at low temperatures (Tsitsishvili *et al.*, 1985; Mumpton, 1999). An additional advantage is the ability to use NH4 + -saturated zeolite as anitrogen fertilizer without regeneration rather than the toxic admixtures normally present in sewage.

The most important features of zeolites can be summarized as follows: (Corma, 1995). They are well-defined inorganic crystalline aluminosilicates with a variety of structures, which differ in channel diameter, geometry and dimensionality; (Perego and Ingallina, 2002). defined inner void volume providing high surface area up to 600 m2. g21 or even higher for delaminated zeolites exceeding 700 m2. 21 g, (Čejka and Wichterlova´, 2002). Ability to absorb and transform molecules in the zeolite inner volume; (van Donk *et al.*, 2003).

Removal of acetic acid from wastewater can be undertaken through various methods, viz, distillation, solvent extraction and adsorption. With such law concentration streams, distillation would not be economically viable. The adsorption of acetic acid on activated carbon has been well documented (4.1). However, the adsorption is poor compared to other larger organic compounds, such as, phenol (<10% -vs. 20% organic weight/carbon weight) Since operational costs are generally related directly to carbon consumption, her is need for emphasis on the improvement of the adsorption characteristic of adsorbents based on Jordanian natural zeolite (JNZ). In the present studies, therefore, an attempt has been made to prepare adsorbents from natural materials, such as natural Jordanian zeolites and use them for the removal of acetic acid from dilute aqueous waste.
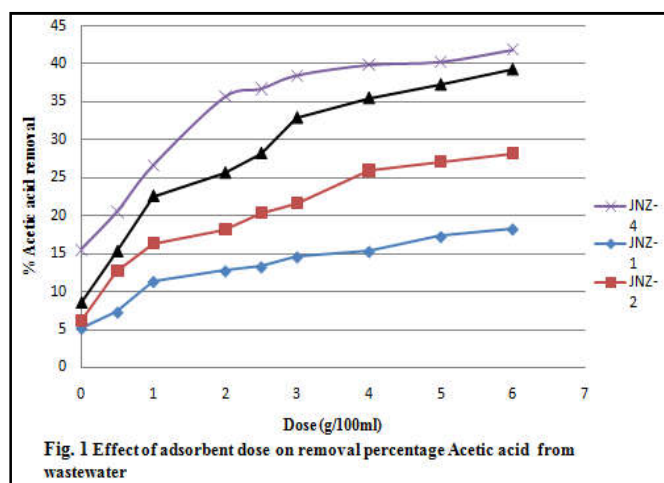
MATERIALS AND METHODS

In order to assess the performance of each adsorbent and to avoid interference by other element in the wastewater, the experiments were conducted with aqueous solution of acetic acid in redistilled water. Solutions of acetic acid were prepared by dissolving acetic acid in redistilled water. Estimation of acid concentration was done by simple titration method using standard sodium hydroxide (NaOH) solution and phenolphthalein as indicator. Activated Jordanian natural zeolite (JNZ-1, JNZ -2, JNZ -3 JNZ -4) or (JNZ1-4) prepared by treating 4 parts of raw NJZ with 3 parts by weight of concentrated sulphuric acid and kept in the air oven maintained at 110°C for the period of 24hr. The zeolites material was washed well with water to remove the free acid and dried at 105□c for 24hr.

The dried JNZ1-4 material was subjected to thermal activation a 600°c for period of 15minutes in the absence of air. The JNZ material was ground and particle size range (200-500 micron) was used throughout the studies. Activated different JNZ1-4 were also prepared in the same manner and the particle size ranges of 250-500, 200-300, and 200-500 microns respectively were used. The variable studied in each case were adsorbent dosage 0.5-6 g/100 ml of feed, initial acid concentration 0.5-6% by volume and temperature 20-40°C. An equilibrium time is around 200 minutes was observed in each case. All the runs were taken for a constant period of 250 minutes duration.

RESULTS AND DISCUSSION
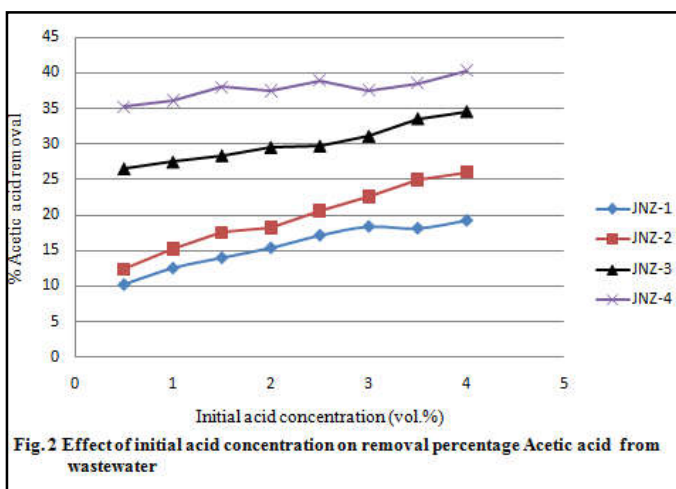
Effect of adsorbent dose

A comparatives study of all the adsorbent has been presented in the (Fig. 1) at constant initial acid concentration of 4% by volume and the temperature 20±1 °C. It may be observed that, in general, the % acetic acid removal increases with the increase in the adsorbent dose. Out of the four adsorbent, activated NJZ-4 gave the maximum removal at all the level of the adsorbent dose, followed by activated NJZ-3, NJZ-2 and NJZ-1. A maximum removal of 41.82% was achieved with 6g/100ml of NJZ-4 dose. The activated NJZ-1 was found to be the poorest of all giving a removal of on 18.25% at 6g/100ml of its dose. The rate of increase in the percent acetic acid removal has been found to be rapid in the beginning which slow down as the dose may be observed beyond which there was no appreciable increase in the adsorption. This phenomenon may be explained based on the facts that because of the increased adsorbent dose more and more adsorbent. However, the very slow increase, if any, beyond the optimum adsorbent dose may be attributed to the attainment of equilibrium between the adsorbate and adsorbent at existing operating condition.

Fig. 1 Effect of adsorbent dose on removal percentage Acetic acid from wastewater

Effect of initial acid concentration

The comparative study has been shown in (Fig 2). The results indicate that out of the four adsorbents studied, activated NJZ-4 gave the maximum removal at all the levels of initial acid concentration followed by NJZ-3, NJZ-2 and NJZ-1 in that order.

A Maximum removal 40.29% may be obtained with NJZ-4 at 4% initial concentration in comparison to just 19.2% removal, in general NJZ-1 at identical condition. The rate of removal, in general, has been found to increase with the increase in the initial acid concentration. However the rate of increase has been very slow in all the cases except the NJZ-1 which showed an almost constant removal with increase in the initial acid concentration from 0.5 to 4%. The increase in the adsorption capacity with increase in the initial feed concentration may be attributed to the fact that the adsorbent having poor affinity toward the adsorbate have lower capacity for adsorption at low acid concentration .As the concentration of acetic acid in the feed is increased, the probability of acetic acid molecules to reach to the adsorbent surfaces increase, resulting in the increased adsorption rate.

Fig. 2 Effect of initial acid concentration on removal percentage Acetic acid from wastewater

Effect of temperature

The temperature dependence of adsorption process is of a very complex nature. Thermodynamic parameter like heat of adsorption and the energy of activation play an important role in predicating the adsorption behavior and both are strongly temperature dependent. Heat of adsorption influences the equilibrium adsorption capacity and also indicate that the nature of interaction taking place between the adsorbate and adsorbents dependent on the energy of activation. Fig.3 presents the results of variation in adsorption rate with temperature. It has been generally observed that with the increase in temperature the percent removal due to the adsorption decrease, which is also evident from result. It may be observed that among of four adsorbent studied, activated NJZ-4 has the maximum removal capacity of acetic acid at all the temperature level, followed by NJZ-3, NJZ-2and NJZ-1. There has been about 17.1% drop in the percent removal value by NJZ-4 at 40°C with respect toits value at 20°C. In case of NJZ-3, NJZ-2and NJZ-1 the decrease has been 10.07%, 6.02%, 5.60% respectively for the same order temperature rise.

This phenomena can be explained on the basis of the fact that at higher temperature, due to the high intrinsic energy of adsorbing molecules, the probability of adhering to the adsorbent surfaces less. Also the overall effect of the temperature is due to its effect on the acetic acid adsorbent

bond, water-adsorbent bond and the water-acetic acid interactions. The water-adsorbent bond is important because adsorption of acetic acid probably involves displacement of water molecules from the adsorbent surface. It may conclude that at higher temperature, the decrease in the adsorption could be due to weakening of either of these bonds or due to the possible exothermic nature of the adsorption process.
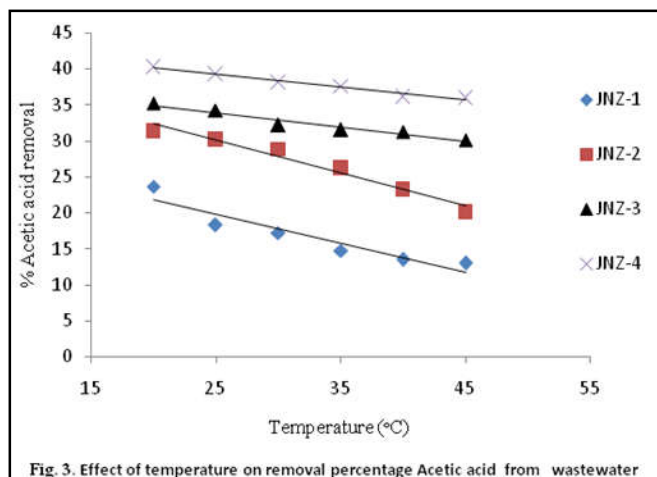

Fig. 3. Effect of temperature on removal percentage Acetic acid from wastewater

Conclusion

The present investigation reveal that activated NJZ-4 has the highest capacity of adsorbing acetic acid from diluted solution maximum removal of the order 40.29.2% was achieved with and adsorbent dose 6gm per 100ml of solution for an initial acid concentration of 4% by volume . Activated NJZ-1 was found to be the poorest of all.

REFERENCES

Čejka, J. and Wichterlová, B. 2002. Acid-catalyzed synthesis of mono- and dialkylbenzenes over zeolites: active sites, zeolite topology, and reaction mechanism. *Catal. Rev.*, 44: 375–421.

Corma, A. 1995. Inorganic solid acids and their use in acid-catalyzed hydrocarbon reactions. *Chem. Rev.*, 95: 559–614.

El-Geundi, M. S. 1993. Branched-pore kinetic model for basic dyestuff adsorption onto natural clay. *Adsor. Sci. Technol.* 9(3), 199–211.

Flanigen, E.M. 1991. Zeolites and molecular sieves – an historical perspective. In: Van Bekkum, H., Flanigen, E.M., Jansen, J.C. (eds) Introduction to zeolite science and practice. Elsevier, Amsterdam, chap 2, p 13.

Kippling, J.J. 1965. Adsorption from solution of nonelectrolyes, academic press, new. York ,pp180.

Lund, H.F. 1971. Industrial pollution control handbook, MC gram hill, pp20.

McKay, G. 1982. Adsorption of dyestuffs from aqueous solutions with activated carbon I: Equilibrium and batch contact-time studies. *J. Chem. Technol. Biotechnol.*, 32, 759–772.

McKay, G. 1998. Application of surface diffusion model to the adsorption of dyes on bagasse pith. *Adsorption,* 4(3–4), 361–372.

McKay, G., Otterburn, M. S. and Sweeney, A. G. 1980. The removal of color from effluent using various adsorbents} III. Silica: Rate processes. *Water Res*., 14, 15–20.

Meshko, V., Markovska, L. and Mincheva, M., 1999. Two resistance mass transfer model for the adsorption of basic dyes from aqueous solution on natural zeolite. *Bull. Chem. Technol. Macedonia,* 18(2), 161–169.

Misaelides, P. (ed.) Zeolite '02, 6 th International Conference on the Occurrence, Properties and Utilization of Natural Zeolites, Thessaloniki, Greece, pp. 247-248

Mumpton, F.A. 1999. *Proc. Natl. Acad. Sci. USA* 96, 3463. Press, Oxford, p. 471.

Perego, C. and Ingallina, P. 2002. Recent advances in the industrial alkylation of aromatics: new catalysts and new processes. Catal. Today, 73: 3–22.

Rodhe, R. and Herraera, R. 1988. Acidification in tropical countries ,John wilely & Sons. Briatian p.p. 188

Tsitsishvili, G.V., Andronikashvili, T.G., Kirov, G.N. and Filizova, L.D., 1985, *Prirodnyje Tseolity,* Khimia, Moscow.

van Donk, S., Janssen, A.H., Bitter, J.H.; de Jong, K.P. 2003. Generation, characterization, and impact of mesopores in zeolite catalysts. *Catal. Rev*., 45: 297–319.

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