

## Adsorption and thermodynamic behavior of uranium on natural zeolite

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Adsorptive behavior of natural clinoptilolite-rich zeolite from Balıkesir deposits in Turkey was assessed for the removal of uranium from aqueous solutions. The uranium uptake and cation exchange capacities of zeolite were determined. The effect of initial uranium concentrations in solution was studied in detail at the optimum conditions determined before (pH 2.0, contact time: 60 minutes, temperature: 20 °C). The uptake equilibrium is best described by Langmuir adsorption isotherm. Some thermodynamic parameters ( $\Delta H^\circ$ ,  $\Delta S^\circ$ ,  $\Delta G^\circ$ ) of the adsorption system were also determined. Application to fixation of uranium to zeolite was performed. The uptake of uranium complex on zeolite followed Langmuir adsorption isotherm for the initial concentration (25 to 100  $\mu\text{g/ml}$ ). Thermodynamic values of  $\Delta G^\circ$ ,  $\Delta S^\circ$  and  $\Delta H^\circ$  found show the spontaneous and exothermic nature of the process of uranium ions uptake by natural zeolite.

### Introduction

Zeolites are crystalline aluminosilicates with framework structure and have excellent cation exchange properties. Zeolites  $[(\text{Na}_2, \text{K}_2, \text{Ca}, \text{Ba})(\text{Al}, \text{Si})\text{O}_2]_x \cdot n\text{H}_2\text{O}$ , (crystalline) are remarkable for their continuous and partly reversible dehydration and for their base exchange properties. Peculiar zeolite properties result from their structure being an aluminosilicate framework with water molecules and exchange cations, in most cases  $\text{Ca}^{2+}$ ,  $\text{Na}^+$  or  $\text{K}^+$ , located in cavities and channels within it. These cations that can be exchanged with other cations seem to be different with their structures, Si/Al atomic ratios and the position of ion exchange sites. Through the cavities and channels of zeolite, water and exchange cations go in and out of zeolites. Most properties of these molecular sieves are known and have been extensively studied.<sup>1–5</sup> Up to now, 34 kinds of natural zeolites are known, but several types, clinoptilolite, mordenite, philipsite are mostly often found in sedimentary rocks and are used.<sup>1</sup>

In the industrial areas of our country, use zeolite minerals which have different technological properties that has not been studied yet.<sup>2</sup> A 2-million ton reserve of natural zeolite clinoptilolite is spread over a 300-km<sup>2</sup> area in Bigadic, Turkey. This reserve has recently been evaluated as an ion exchanger and as a gas and vapour adsorbent. Although it has a very high adsorption capacity, its mechanical strength is low for use as a column packing.<sup>3</sup> Balıkesir-Bigadic region zeolites have been produced at a good level and exported every year.<sup>2</sup> Zeolite samples from Balıkesir-Bigadic region have higher ratio of alkaline earth metal cations and are thermally more stable than those from the fine grained particles.

The adsorption of radionuclides from liquid solutions by solids has been widely studied in recent years and various procedures have been adopted by different

workers. Various types of sorbents and coprecipitants were studied to develop an industrial method for removal and recovery of uranium that may be present in uranium refining waste water and other water sources and also for extraction of uranium from natural waters. Adsorption of soluble metallic species by various adsorbents appears to be an important means of controlling the soluble metal ion concentrations in heterogeneous systems. Zeolites, kinds of inorganic ion exchangers with high resistance to radiation have been widely employed for adsorption processes because of their specific ion exchange properties. The structures of some zeolites are the rigid frame works containing narrow channels and they have ion sieving properties for larger size of hydrated cations.<sup>1,4–15</sup>

One of the authors has used this material to remove uranium from aqueous solution and has studied some adsorption parameters such as pH, contact time and temperature.<sup>16</sup> In order to continue the study of adsorption processes on the zeolite, the uptake and cation exchange capacities of zeolite were determined and the effect of initial uranium concentrations in solution was studied in detail. The uptake equilibrium is best described by Langmuir adsorption isotherm. Some thermodynamic parameters ( $\Delta H^\circ$ ,  $\Delta S^\circ$ ,  $\Delta G^\circ$ ) of the adsorption system were also determined. Application to fixation of uranium to zeolite was performed.

### Experimental

All chemical reagents used in the study were of analytical reagent grade (AR Grade). All solutions were prepared in distilled water. Solutions of uranium were prepared by dissolving known quantities of uranyl nitrate hexahydrate  $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  in distilled water. The buffers of pH 4 and 8 were used for the calibration of pH meter. Solutions of  $\text{HNO}_3$  and  $\text{Na}_2\text{CO}_3\text{--NaHCO}_3$  were added for pH adjustment. Solutions were filtered through a Whatman No. 42 filter paper.

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### Adsorbent

Clinoptilolite-rich zeolite samples from Bigadic-Turkey were obtained from Dokuz Eylul University, Faculty of Engineering Department of Geology in Izmir. Chemical composition of clinoptilolite<sup>17</sup> is  $\text{Na}_6(\text{AlO}_2)_6(\text{SiO}_2)_{30} \cdot 24\text{H}_2\text{O}$  (Si/Al:5). The chemical analysis of clinoptilolite-rich zeolite samples by X-ray fluorescence technique (XRF) realized by Ata and Girgin<sup>17</sup> was shown in Table 1.

The sample was crushed in jaw and roll crushers and homogenized. It was pulverized in a ball mill and sieved through a -120 mesh screen. Clinoptilolite-rich zeolite was used for the adsorption studies after treatment with 0.1M HCl at room temperature. The sample was filtered and washed with distilled water to remove  $\text{Cl}^-$  ions. Filtered sample was dried overnight at 50 °C in a laboratory oven.

### Adsorption experiments

The adsorption of uranium on zeolite was studied by a batch technique. A known weight of the zeolite was equilibrated with definite volume of the uranium solution of known concentration in a flask at a fixed temperature in a thermostated shaker water bath for a known period of time. After equilibration the suspension was filtered. The equilibrium adsorption was calculated from the residual concentration of the sorbate in equilibrated solution. The uranium concentration in solution was determined by DBM (dibenzoylmethane) method<sup>18</sup> using Shimadzu 260 Model UV-Vis spectrophotometer.

Adsorption of uranium on zeolite was determined in terms of distribution coefficient,  $K_d$ , percentage adsorption or amount sorbed per unit weight of the sorbent,  $x/m$ . The distribution coefficient,  $K_d$ , is defined as the concentration of a species sorbed per gram of the sorbent divided by its concentration per ml in liquid phase:

$$K_d = C_{ads}/C_e (V/m) \text{ ml/g}$$

where  $C_{ads}$  and  $C_e$  are the masses of uranium sorbed and in solution, respectively,  $m$  is the mass of the sorbent in g, and  $V$  is the volume of the solution used for equilibration in ml. The percentage adsorption of zeolite was calculated from the initial ( $m_i$ ) and final ( $m_f$ ) amount of uranium of the solutions:

$$\text{Percentage adsorption (\%)} = m_i - m_f / m_i \times 100$$

The results are the average of three independent measurements and precision in most cases was  $\pm 5\%$ .

### Determination of uranium uptake capacity of the zeolite

To determine uranium uptake capacity, about 0.1 g zeolite sample (heated at 105 °C) was taken and placed in a polypropylene bottle and then 25 ml of 0.01M uranium solution prepared from  $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was added to the sample. Bottle was shaken in a thermostated bath shaker for 24 hours at 30 °C. The solid and liquid phases were separated. The filtrate was analyzed spectrophotometrically for uranium. Salicylic acid method<sup>19</sup> was used for the determination of uranium uptake capacity of zeolite.<sup>19</sup>

### Determination of cation exchange capacity (CEC) of zeolite

The zeolite sample (0.1 g) and 50 ml of 0.1M  $\text{HNO}_3$  were shaken at 30 °C for 15 hours. 5 ml portion of supernatant was titrated with 0.1M NaOH solution using methyl orange indicator. Cation exchange capacity was calculated as mmol  $\text{H}^+$  per g zeolite.<sup>20</sup>

### Leachability of uranium from calcined zeolite

In order to realize the ceramic solidification with lower leachability, zeolite sorbed uranium was calcined in the temperature range from 600 to 1100 °C in an electric furnace for 1.5 hours. The leachability of uranium from this calcined zeolite was investigated. The leaching tests have been carried out at room temperature for 24 hours using distilled water.

## Result and discussion

### Adsorption capacities of zeolite

The natural zeolite sample was prepared as granular, powder and pelletized forms. The uranium adsorption capacities of granular (32–48 mesh) and powdered (below 200 mesh) zeolite were found to be 0.214 and 0.300 mmol  $\text{UO}_2^{2+}$ /g, respectively. The pelletized form of zeolite (11 mm  $\varnothing$ , 1.0 mm height) decomposed during the contact with the solution. It was found that the adsorption capacity of powdered zeolite was greater than that of the granular form. Adsorption experiments

Table 1. Chemical analysis of Bigadic clinoptilolite<sup>17</sup>

| Metal oxides            | Chemical composition, % |
|-------------------------|-------------------------|
| $\text{SiO}_2$          | 77.96                   |
| $\text{Al}_2\text{O}_3$ | 13.17                   |
| CaO                     | 3.80                    |
| $\text{K}_2\text{O}$    | 2.20                    |
| MgO                     | 1.53                    |
| $\text{Fe}_2\text{O}_3$ | 1.06                    |
| $\text{Na}_2\text{O}$   | 0.16                    |
| $\text{TiO}_2$          | 0.08                    |

were carried out on powdered zeolite samples for further studies. Cation exchange capacity was also found 1.70 mmol of  $H^+$  per 1g of the natural zeolite.

### Effect of pH

The most important parameter influencing the adsorption is the pH of the adsorption medium. The effect of adsorption of uranium (25  $\mu\text{g/ml}$ ) on zeolite was studied by varying the pH of the uranium aqueous solution–zeolite suspension from pH 2.0 to 9.0 at 30 °C for 60 minutes. Maximum adsorption yields were observed at pH 2.0 and 8.0.

The results obtained from previous study<sup>16</sup> are shown in Fig. 1. The percentage adsorption and  $K_d$  values for uranium adsorption decrease sharply when the pH increases from 2.0 to 5.0. However, beyond pH 5 a sudden increase in  $K_d$  and percentage adsorption occurs. The high  $K_d$  and percentage adsorption of uranium on zeolite at lower pH is due to the fact that hydrolysis of uranyl ion takes place as the pH varies from 1 to 4. When pH increases around 4, precipitation starts due to the formation of complexes in aqueous solution and adsorption decreases. The oxides of aluminum, calcium, magnesium, iron and silicon are present in varying amounts in zeolite (Table 1). The hydroxylated surfaces of oxides develop a charge on the surface in aqueous solution through amphoteric dissociation. The surface will be positively charged at lower pH. The competition between metal and hydrogen ions for active sites is further evident from Fig. 1. The system was attained an

equilibrium at pH 2.0. On the basis of these results, pH 2.0 was selected for uranium adsorption on zeolite. These data suggest that the binding of metal ions involves displacement of hydrogen ions from the active sites. Hydrogen ions are displaced by uranyl ions.<sup>21</sup>

The positively charged  $\text{UO}_2^{2+}$  ions exist at pH 2.0. However, negatively charged complex hydrolyzed uranium ions have been suggested to exist at pH 4.0. Hydrolysis products are in simultaneous dynamic equilibria. The uranium hydrolysis products distribution curves available in literature suggest that for low total  $\text{U}^{6+}$  concentrations and for pH below 5.<sup>22</sup>

The percentage adsorption increases up to 92% at pH 8.0. Near pH 7.0, the uranyl ions forms stable complexes with carbonate. In pH 7.0–10.0, the soluble carbonate complexes of  $\text{UO}_2^{2+}$  are the predominant anion species:  $\text{UO}_2(\text{CO}_3)_3^{2-}$  and  $\text{UO}_2(\text{CO}_3)_3^{4-}$ . These two complexes exist in various ratios depending on the pH of the water.<sup>23</sup>

### Effect of uranium concentration

The adsorption of uranium on zeolite as a function of the uranium concentration was studied at room temperature by varying uranium concentration from 10 to 100  $\mu\text{g/ml}$ , while keeping all other parameters constant. The specific uranium uptake and the corresponding equilibrium concentration of uranium was investigated as a function of the initial uranium concentration at pH 2 at room temperature for 60 minutes (Fig. 2).

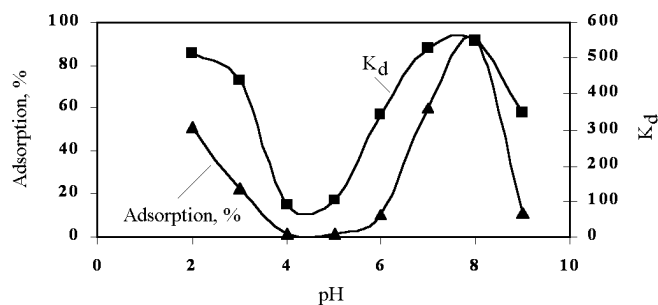


Fig. 1. Effect of pH on adsorption of uranium on zeolite

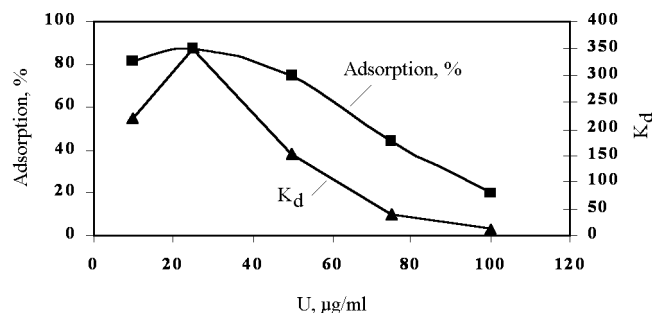


Fig. 2. Equilibrium adsorption of uranium at different initial uranium concentrations

Zeolite gave the best uranium adsorption with a  $K_d$  of 351 and uranium removal with 88% uptake at 25  $\mu\text{g/ml}$  initial uranium concentration. Percentage adsorption for uranium decreases with increasing bulk metal concentration in aqueous solution. These results indicate that energetically less favorable sites become involved with increasing metal concentrations in the aqueous solution. Uranium concentration of 25  $\mu\text{g/ml}$  in solution was selected for further studies.

#### Effect of contact time

The effect of contact time was studied at constant concentration of uranium solution (25  $\mu\text{g/ml}$ ) and solution–solid ratio (50 ml/g) at room temperature. The results obtained from previous study<sup>16</sup> are shown in Fig. 3. It is seen that there was only a small difference between 120 minutes and 360 minutes which assumes that the zeolite has reached equilibrium with the solution. Maximum  $K_d$  and U removal values were observed at 60-minute contact time.

#### Effect of temperature

Effect of temperature was also studied for uranium in the range of 10–80 °C. Room temperature was found to be the most suitable for maximum uranium adsorption

on zeolite. The results obtained from previous study<sup>16</sup> are shown in Fig. 4, which illustrates the effect of temperature for uranium uptake on zeolite at pH 2.0. Adsorption of uranium exhibited higher uptake at 20 °C as compared to that at 10 and 40 °C. Figure 4 also shows that the distribution coefficient values decreased with the increase of temperature. The results show that the adsorption of uranium ions on zeolite is an exothermic process.

#### Thermodynamic parameters

The values of  $\Delta H^\circ$  and  $\Delta S^\circ$  were calculated from the slopes and intercepts of the linear variation of  $\ln K_d$  with the reciprocal of the temperature,  $1/T$  (Fig. 5), by using the relation:<sup>24</sup>

$$\ln K_d = -(\Delta H^\circ/RT) + (\Delta S^\circ/R)$$

The thermodynamic values are given in Table 2. The free energy of the specific adsorption,  $\Delta G^\circ$  is calculated from:

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ$$

Both negative value of  $\Delta H^\circ$  and the increase in the value of  $\Delta G^\circ$  with an increase in temperature show that the reaction is more favorable at low temperature.

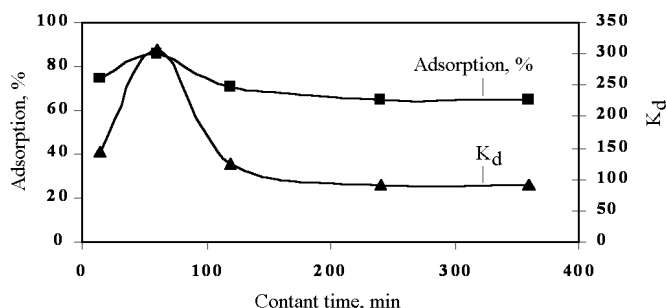


Fig. 3. Effect of contact time on adsorption of uranium on zeolite

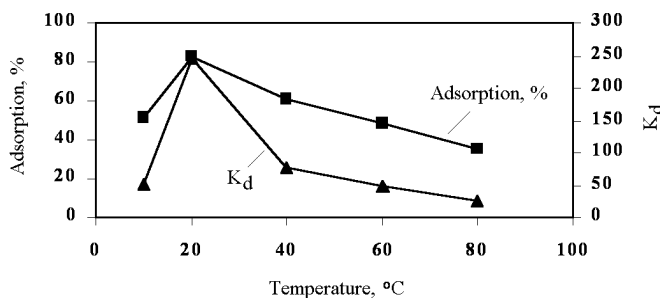


Fig. 4. Effect of temperature on adsorption of uranium on zeolite

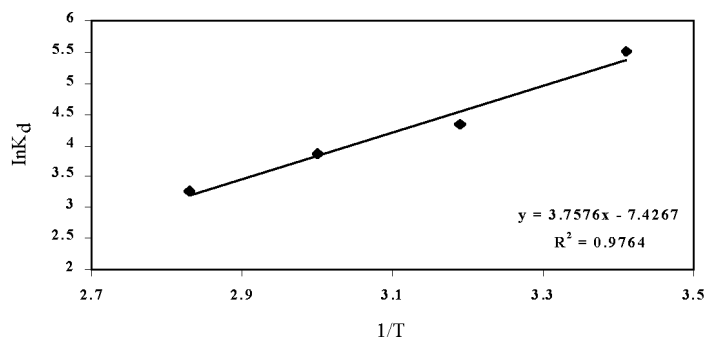


Fig. 5. Plot of  $\ln K_d$  versus  $1/T$  for uranium adsorption on zeolite

Table 2. Thermodynamic parameters for uranium adsorption on zeolite

| $\Delta H^\circ$ ,<br>kJ/mol | $\Delta S^\circ$ ,<br>J/mol | $\Delta G^\circ$ ,<br>kJ/mol |        |        |        |       |
|------------------------------|-----------------------------|------------------------------|--------|--------|--------|-------|
|                              |                             | 283 K                        | 293 K  | 313 K  | 333 K  | 353 K |
| -29.99                       | -57.62                      | -13.69                       | -13.11 | -11.96 | -10.81 | -9.66 |

### Adsorption isotherms

The adsorption isotherms for uranium adsorption on zeolite were obtained at constant temperature at various metal concentrations while keeping all parameters constant. Several common adsorption models including Langmuir, Freundlich and Dubinin-Radushkevich (D-R) were considered to fit the data for adsorption of uranium on zeolite. These isotherms which were originally derived mainly for gas-solid systems have also been used to represent the adsorption of solutes in liquid-solid system.<sup>25</sup> Among the models tested, Langmuir Model was found to give a relatively better fit to the experimental equilibrium data in a narrow range of concentrations for uranium (from 25 to 100  $\mu\text{g/ml}$ ) (Fig. 6). Langmuir isotherm was tested in the following linearized form for solid-liquid systems:

$$C_e/y = (1/a) + (b/a) \cdot C_e$$

where  $C_e$  is the concentration of solute in the liquid phase at equilibrium (g/l),  $y$  is the amount of solute sorbed per unit of sorbent (mg/g),  $b$  and  $a$  are the Langmuir constant:  $b$  is represented as the ratio of adsorption and desorption rate coefficients (l/g), and

$$a = b \cdot Q^0$$

where  $Q^0$  is the maximum adsorption capacity (mg/g). At low concentrations the Langmuir equation reduces to a linear relationship, while the maximum adsorption capacity,  $Q^0$ , is attained at concentrations corresponding to monolayer coverage.

Maximum adsorption is observed when the sorbent surface is covered with a monolayer of sorbate. The Langmuir model is developed to represent chemisorption on a set of well-defined localized adsorption sites having same adsorption energies independent of surface coverage and no interaction between adsorbed molecules.<sup>25</sup>

MISAEELIDES et al.<sup>26</sup> have proved an inhomogeneous depth distribution of sorbed metals on the mineral surface and particularly an intense metal accumulation in the near-surface layers of the material, which can be attributed to an anomalous diffusion of metal cations into the micropores due to ion-exchange reactions. This phenomenon strongly depends on the size of the zeolite crystals, the treatment time and the type of exchangeable cations. They have also showed that the surface morphology and microtopography seem to play an important role to the chemical processes taking place at the zeolite surface.<sup>26</sup>

### Fixation of uranium onto zeolite and leaching

The leachability of radionuclides from solidified product is one of the most important parameters in the safety assessment for the management of wastes because the leaching of radionuclides from solid waste is the first step of the nuclide migration with ground water in the repository.<sup>10</sup> Therefore, the leachability of uranium from calcined zeolites was investigated.

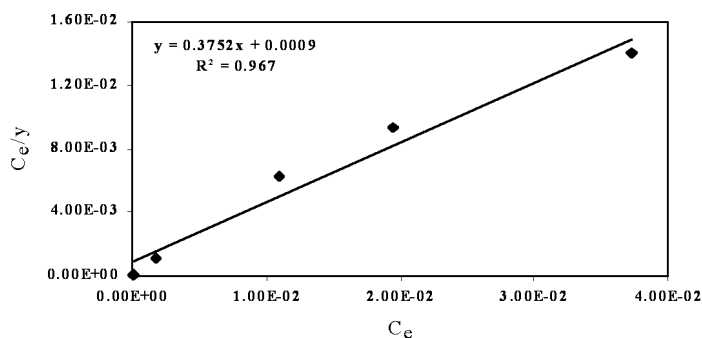


Fig. 6. Langmuir isotherm for uranium adsorption on zeolite

The variation of the leachability of uranium from calcined product into distilled water was shown in Table 3. It was observed that the leachability of uranium over 800 °C was decreased with increasing calcination temperature of zeolite. It can be seen that calcined zeolite had low leachability at high temperatures. These results are advantages of possible application for storage of uranium in radioactive wastes.

### Conclusions

In order to continue the study of the adsorption processes of uranium on zeolite, this study indicates that the removal of uranium from dilute aqueous solution also depends on uranium concentration in addition to the study<sup>16</sup> of some adsorption parameters such as pH, contact time and temperature using zeolite. Amounts of removed uranium were changed in different forms of sorbent. At the optimum conditions, zeolite exhibited highest removal capacity (around 85%) for uranium ions. The uptake and cation exchange capacities of zeolite were determined and the effect of initial uranium concentrations in solution was studied in detail.

The temperature variation has been used to evaluate the values of  $\Delta G^\circ$ ,  $\Delta H^\circ$  and  $\Delta S^\circ$ . Clinoptilolite pretreated with HCl indicated that it can be used to separate uranium ions from aqueous solution in given conditions. The uptake equilibrium is best described by Langmuir adsorption isotherm. Values of  $\Delta G^\circ$  and  $\Delta H^\circ$  found show the spontaneous and exothermic nature of the process of uranium ions by natural zeolite. The great

Table 3. Variation of leachability of uranium into distilled water with calcination temperature

| Calcination temperature,<br>°C | Leachability of uranium,<br>% |
|--------------------------------|-------------------------------|
| 600                            | 1.13                          |
| 800                            | 1.66                          |
| 1000                           | 0.25                          |
| 1100                           | 0.26                          |

Calcination time: 1.5 h, leaching time: 24 h.

advantage of natural clinoptilolite-rich zeolite in Bigadic-Turkey is economical and effective sorbent for uranium. These results show that clinoptilolite-rich zeolite can effectively be used for adsorption and removal of uranium ions from dilute aqueous solution and for storage of uranium in radioactive solutions after a calcination process.

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### References

- I. J. LIN, I. SPEKOVA, Utilization of Natural Minerals in Water Treatment, Progress in Mineral Processing Technology, DEMIREL and ERSAYIN (Eds), Balkema-Rotterdam 1994.
- U. KOKTURK, H. YILMAZ, A. BAYKAL, Fibrous Formation in Bigadic Zeolite Formation by X-Ray Microprobe Instrument, Industrial Raw Material Symposium, Izmir, Turkey 1995.
- D. BALKOSE, S. ULUTAN, F. OZKAN, S. ULKU, U. KOKTURK, Separ Sci. Technol., 31 (1996) 1279.
- H. MIMURA, K. AKIBA, Separation of Radionuclides by L Zeolite, Bulletin of the Research Institute of Mineral Dressing and Metallurgy, Tohoku Univ., 43 (1987) 23 (in Japanese).
- H. MIMURA, I. ISEKI, K. KUDO, T. KANNO, Concentration of Radioactive-Cobalt by Zeolites from Low-Level Aqueous Solutions, Bulletin of the Research Institute of Mineral Dressing and Metallurgy, Tohoku Univ. 40 (1984) 27 (in Japanese).
- H. MIMURA, T. KANNO, J. Nucl. Sci. Technol., 22 (1985) 284.
- C. N. HSU, H. MIMURA, M. TENMAN, T. KANNO, Removal of Cesium and Strontium from High-Level Radioactive Waste and Solidification of Denitrated Precipitates by Use of Zeolites, Bulletin of the Research Institute of Mineral Dressing and Metallurgy, Tohoku Univ., 40 (1984) 38 (in Japanese).
- S. WINGEFORS, G. PERSSON, J. O. LILJENZIN, Radioact. Waste Managem. Nucl. Fuel Cycle, 5 (1884) 327.
- S. FORBERG, T. WESTERMARK, R. ARNEK, I. GRENTHE, L. FALTH, S. ANDERSSON, Fixation of Medium-Level Wastes in Titanates and Zeolite, Scientific Basis for Nuclear Waste Management, Vol. 2, Clyde and Northrup, New York, 1980, p. 867.
- T. KANNO, H. MIMURA, Ion Exchange Properties of Zeolites and Their Applications to Processing of High Level Liquid Waste, IAEA-TECDOC-337, IAEA, Vienna, 1985, p. 237.
- K. KAYABALLI, H. KEZER, Environ. Geol., 34 (1998) 95.

12. H. FAGHIHIAN, M. GHANNADI MARAGHEH, A. MALEKPOUR, *J. Radioanal. Nucl. Chem.*, 254 (2002) 545.
13. M. A. HERNANDEZ, L. CORONA, F. ROJAS, *Adsorption*, 6 (2000) 33.
14. A. E. SENTALAR, A. SIRKECIOGLU, I. GURAY, F. ESENLI, I. KUMBASAR, Characterization of the Clinoptilolite Rich Tuffs of Bigadic: Variation of the Ion Exchange Capacity with Pretreatments and Zeolite Content, 9th Intern. Zeolite Conf. 1992, Montreal, Canada.
15. Y. BURKUT, A. CELENLI, F. SUNER, Investigation of Some Ion-Exchange Properties of Bigadic-Balikesir Zeolite, Proc. 6th National Clay Symp., 1993, p. 37.
16. A. GUMUS, G. BAKSI, M. S. TANER, S. OLMEZ, Investigation of Adsorption of Uranium on Zeolite from Aqueous Solution, Turkish VIIIth National Symp. on Chemistry and Chemical Engineering, 1992, p. 79 (in Turkish).
17. S. ATA, I. GIRGIN, *Progr. Mineral Technol.*, (1994) 397.
18. J. KORKISCH, *Modern Methods for the Separation of Rare Metal Ions*, Pergamon Press, Oxford, 1974.
19. N. KABAY, H. EGAWA, *Separ. Sci. Technol.*, 28 (1993) 1985.
20. N. KABAY, Investigation of Utilization of Modified Polymeric Amidoxime-resins for Recovery of Uranium from Sea Water, Ege Univ., Research Foundation Project No. 93 NBE 003, Izmir, 1994 (in Turkish).
21. N. FRIIS, P. MYERS-KEITH, *Biotechnol. Bioeng.*, 28 (1986) 21.
22. M. TSEZOS, B. VOLESKY, *Biotechnol. Bioeng.*, 23 (1981) 583.
23. T. J. SORG, Removal of Uranium from Drinking Water by Conventional Treatment Methods, in: Radon, Radium and Uranium in Drinking Water, COTHERN and REBERS (Eds), Lewis Publishers, Michigan, 1991, p. 173.
24. M. SALEEM, M. AFZAL, R. QUADER, J. HANIF, *Separ. Sci. Technol.*, 27 (1992) 239.
25. S. M. HASANY, M. M. SAEED, M. AHMED, *J. Radioanal. Nucl. Chem.*, 252 (2002) 477.
26. P. MISAELIDES, A. GODELITSAS, S. KOSSIONIDIS, G. MANOS, *Nucl. Instr. Meth. Phys. Res.*, 113 (1999) 296.