# Conversion of Coal Fly Ash into Zeolite and Heavy Metal Removal Characteristics of the Products

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**Abstract**−Fly ash obtained from a power generation plant was used for synthesizing zeolite. Zeolites could be readily synthesized from the glassy combustion residues and showed potential for the removal of heavy metal ions. By the use of different temperatures and NaOH concentration, five different zeolites were obtained: Na-P1, faujasite, hydroxy sodalite, analcime, and cancrinite. The synthesized zeolites had greater adsorption capabilities for heavy metals than the original fly ash and natural zeolites. Na-P1 exhibited the highest adsorption capacity with a maximum value of about 1.29 mmole Pb g$^{-1}$ and had a strong affinity for Pb$^{2+}$ ion. The metal ion selectivity of Na-P1 was determined as: Pb$^{2+}$>Cu$^{2+}$>Cd$^{2+}$>Zn$^{2+}$, consistent with the decreasing order of the radius of hydrated metal ion. The adsorption isotherm for lead by Na-P1 fitted the Freundlich rather than the Langmuir isotherm.

Key words: Fly Ash, Synthetic Zeolite, Adsorption, Heavy Metal Adsorption

## INTRODUCTION

As economic development has increased and the living standard has risen throughout the world, the demand for energy and consequently the use of coal has increased. During the burning process of coal for the generation of electricity, the waste products are mainly fly ash. According to Ferraiolo et al. [1990] and Singh et al. [1993], the weight percentage of ash to coal buried world-wide is almost 10-15 wt% of which 10% is bottom ash and the rest is fly ash.

A large amount of fly ash is generated annually from the combustion of coal in power plants. Only a small amount of fly ash is used as an additive to cement. However, most of that has been disposed of by dumping, which gives rise to serious environmental pollution [Jiang and Roy, 1992]. Consequently, the development of methods for greater utilization and production of high value compounds from waste ash has been the object of recent research.

One major use of fly ash is for the removal of substances such as phenol [Singh and Rawat, 1994], oxalic acid [Jain et al., 1978], and other organic compounds [Teuney and Echelberger, 1970; Hung, 1983] from solution by adsorption. Banerjee et al. [1995] examined the dynamic behavior of fly ash in a batch experiment and found that it exhibited good adsorption properties for dissolved organic compounds in water. Some other applications are related to the removal of heavy metals [Panday et al., 1985; Yadara et al., 1987; Kapoor and Viraraghaven, 1992; Viraraghavan and Dronamraju, 1993].

A new approach for application of fly ash is to convert it into zeolites. As the chemical composition of fly ash is similar to that of zeolites, much research has been focussed on the synthesis of various zeolites by alkaline treatment of the ash [Amrhein et al., 1996; Shigemoto et al., 1995; Lin, 1995]. The constituents of fly ash are mainly aluminosilicate glass, mullite and quartz, with a small amount of residual coal and ore minerals; the aluminosilicate glass is a readily available source of Si and Al for zeolite synthesis. However, most previous studies have been done about the conversion of fly ash to zeolites at a limited temperature (80-100 °C). They merely include the classification and adsorption rate of zeolites produced.

In this study a method for the efficient utilization of fly ash by the synthesis of various zeolites, and evaluation of their potential for the removal of heavy metals, were investigated. The conditions required for zeolite synthesis, the adsorption properties, adsorption isotherms and rate of adsorption of the synthesized zeolites for heavy metals were studied and compared with those of fly ash and natural zeolites.

## EXPERIMENTAL

The fly ash sample used in this work was obtained from a power plant located at Boryung in Chungnam, Korea. The sample used in all experiments was pretreated in a high gradient magnetic separator to remove Fe$_2$O$_3$ and TiO$_2$, which are known to be undesirable for zeolite synthesis [Tazaki et al., 1989]. The zeolites were synthesized in teflon-lined stainless steel vessels in a digital-type forced-convection electric oven at 70-200 °C over 24-48 hours without stirring as described in the previous paper [Kim et al., 1997].

Powder XRD patterns (Rigaku Model max/IIIA) and SEM (Topcon ABT-32) were employed to characterize the synthetic zeolites. The chemical compositions of the synthetic zeolites were determined by using XRF (Multi-Channel X-Ray Fluo-

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rescence Spectrometer, Shimadzu model MXF-2100). The synthetic product was further characterized by thermogravimetry (Cahn TG121) with a heating rate of 8 K/min from room temperature to 500 °C.

Batch experiments were carried out to investigate the adsorption properties of the fly ash conversion products for heavy metals. Analytical grades of $Pb(NO_3)_2$, $Cd(NO_3)_2 \cdot 4H_2O$, $Cu(NO_3)_2 \cdot H_2O$ and $Zn(NO_3)_2 \cdot 6H_2O$ were used to prepare stock solutions of 20 mM which were diluted for use. One liter of metal ion solution was placed in a 2 L Erlenmeyer flask. Various amounts of adsorbent (fly ash conversion product, fly ash or natural zeolite) were added and the solutions stirred with a magnetic stirrer at greater than 300 rpm. At given time intervals, 2 mL of sample was taken, centrifuged at 6,000 rpm for 10 min, and the supernatant used for the determination of residual metal ion concentrations. The total volume of samples amounted to 16 mL. A water bath was used to maintain a constant temperature of 25 °C.

All experimental vessels were pyrex glass and all glassware was leached in 14% nitric acid and washed with distilled water prior to use. The metal ion concentrations were obtained by using atomic absorption spectroscopy (UNICAM 939 AA Spectrophotometer).

## RESULTS AND DISCUSSION

### 1. Characteristics of Fly Ash and Conversion Products of Fly Ash

The untreated coal fly ash consists mainly of hollow spheres of varying size, as described in the previous paper [Kim et al., 1997], although irregularly shaped particles with concave surfaces were also observed. The particles were distributed over a wide size range of up to 180 µm, but were mainly between 10 and 50 µm in diameter. The aluminosilicate glass comprised more than 30-40% of the fly ash and was a readily available source material for zeolite synthesis. Quartz (Q, $SiO_2$) and mullite (M, $Al_6Si_2O_{13}$) were the two major crystalline phases; a small amount of hematite (H, $Fe_2O_3$) was also identified.

By using a caustic solution, the fly ash can be easily converted to the zeolite phase via hydrothermal reaction. The chemical compositions of the untreated fly ash and synthesized products are compared in Table 1. The synthetic product showed higher loss on ignition (>10%) and higher $Na_2O$ content than
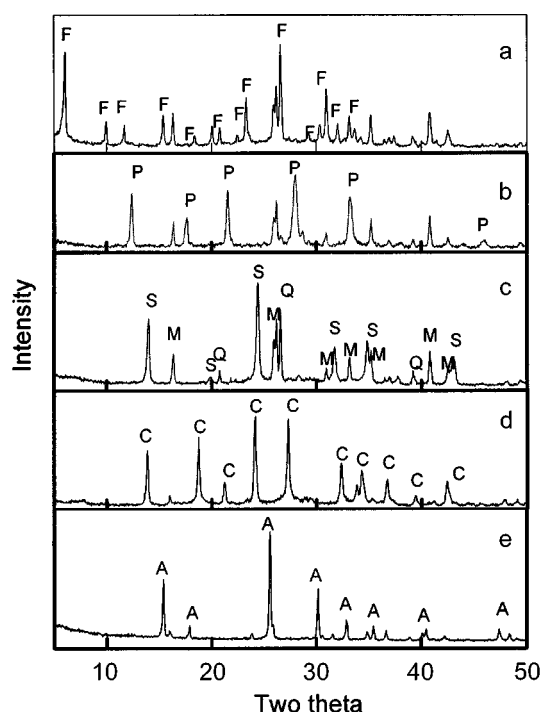


**Fig. 1. XRD patterns of the synthetic zeolites.**
(A: analcime, C: cancrinite, F: faujasite, P: Na-P1, S: hydroxy sodalite).
(a) 3 N NaOH, 70 °C, 48 h      (d) 5 N NaOH, 160 °C, 24 h
(b) 3 N NaOH, 100 °C, 48 h     (e) 1 N NaOH, 160 °C, 24 h
(c) 5 N NaOH, 100 °C, 24 h

those of fly ash. Thermogravimetric analysis also showed a high weight loss (ca. 10%) in the synthetic product. These results seem to be due to desorption of adsorbed water. After cation exchange, there was more than 90% loss of $Na_2O$ and consequent similar increase in $K_2O$. The high water adsorption and cation-exchange properties of the synthetic product are indicative of the formation of zeolite-like materials from the raw fly ash. In the case of Na-P1, the ratio of aluminium to exchangeable cation showed that ca. 37% of the total $Al_2O_3$ content was changed to the zeolite phase and the remaining aluminium content seemed to be derived from the unreacted mullite phase of the fly ash. However, almost all $Al_2O_3$, including the mullite phase, could be converted to the zeolite phase at higher temperature, such as 160

**Table 1. Chemical compositions of raw fly ash and synthesized products**

| Sample | Treatment | Compositions (wt%) | | | | | | | | |
| | | $SiO_2$ | $Al_2O_3$ | CaO | $Fe_2O_3$ | $Na_2O$ | others[a] | LOI[b] | Wt loss in TG[c] | Na/Al |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Fly ash | | 50.0 | 28.7 | 2.6 | 3.7 | 0.3 | 5.6 | 4.7 | 2.3 | 0.02 |
| FAU[e] | 3 N NaOH, 70 °C | 38.1 | 30.1 | 4.8 | 4.9 | 5.6 | 3.9 | 12.3 | 12.9 | 0.31 |
| Na-P1 | 3 N NaOH, 100 °C | 37.0 | 30.3 | 2.5 | 2.5 | 6.8 | 3.4 | 17.4 | 11.8 | 0.37 |
| SOD | 5 N NaOH, 100 °C | 34.2 | 31.7 | 4.1 | 3.1 | 10.1 | 3.1 | 13.3 | 9.6 | 0.52 |
| CAN | 5 N NaOH, 160 °C | 38.6 | 25.9 | 2.2 | 2.2 | 15.6 | 2.8 | 12.4 | 10.4 | 0.99 |
| ANA | 1 N NaOH, 160 °C | 40.3 | 25.8 | 2.2 | 2.1 | 14.2 | 2.4 | 12.5 | 11.6 | 0.91 |
| K+-exchanged | Na-P1 | 37.8 | 30.3 | 2.1 | 2.5 | 0.3 | 11.8 | 15.1 | | 0.32[d] |

[a]Sum of MgO, $K_2O$, $TiO_2$, MnO and $P_2O_5$. [b]Loss on ignition. [c]Weight loss in thermogravimetric analysis. [d]K/Al, mole ratio. [e]FAU refers to the faujasite-type zeolite obtained from fly ash.
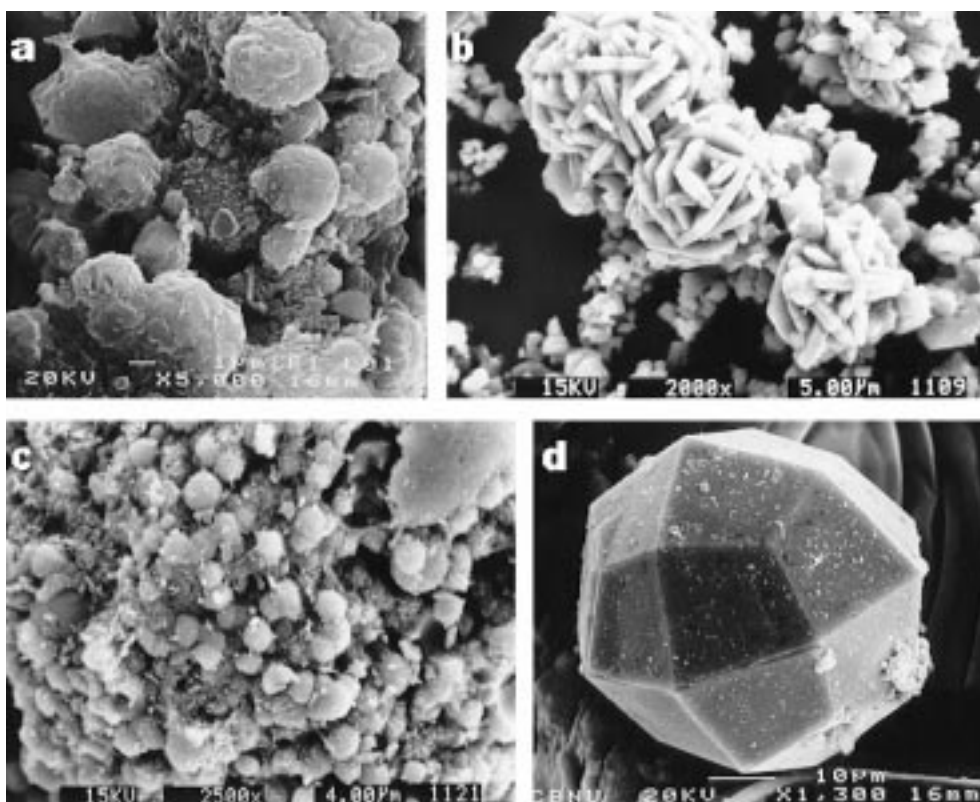
**Fig. 2. SEM micrographs of the synthesized zeolites.**
(a) 3 N NaOH, 70 °C, 48 h (b) 4 N NaOH, 100 °C, 48 h (c) 5 N NaOH, 100 °C, 24 h (d) 3 N NaOH, 160 °C, 24 h (e) 1 N NaOH, 160 °C, 24 h.

°C, as shown in CAN and ANA.

**2. Effect of NaOH Concentration and Temperature**

Fig. 1 shows the formation of zeolite, namely, faujasite (FAU), Na-P1, hydroxy sodalite (SOD), analcime (ANA) and cancrinite (CAN), according to reaction conditions. Fig. 1a shows that the faujasite phase predominantly occurred with decrease of the quartz peak in 3 N NaOH at 70 °C. At 100 °C Na-P1 was the major phase (Fig. 1b). However, the increase of molarity of NaOH (to 5 N or more) accelerated the formation of hydroxy sodalite as shown in Fig. 1c. By increasing the temperature to 160 °C, all mullite, quartz and hematite phases disappeared and only zeolite phases were present. At 160 °C, cancrinite coexisted with Na-P1 at 4 N NaOH and at higher concentrations of NaOH only cancrinite phase was formed as shown in Fig. 1d, whereas only analcime was observed at lower alkaline concentration, i.e., 1 N NaOH, as shown in Fig. 1e.

**3. Structure of the Synthesized Zeolites**

Fig. 2 shows SEM photomicrographs of zeolite phases produced in this study. As shown in Fig. 2a, the typical shape of the faujasite phase was obtained at 70 °C. The SEM micrograph of Na-P1 zeolite (Fig. 2b) clearly indicates the formation of polycrystalline aggregates of pure Na-P1 product which was crystallized from soluble species in the NaOH-fly ash-$H_2O$ system at 100 °C. Fig. 2c shows the spheroidal hydroxyl sodalite with average diameter of 2 μm. Fig. 2d shows the typical trapezohedral shape of analcime phase, and the particles of which were 60 μm in diameter.

**4. Conditions for Synthesis of Zeolite from Fly Ash**

The conditions for synthesis of zeolites are schematically depicted in Fig. 3. Five different phases of zeolites were formed in 3 N NaOH over the temperature range of 70-180 °C as shown
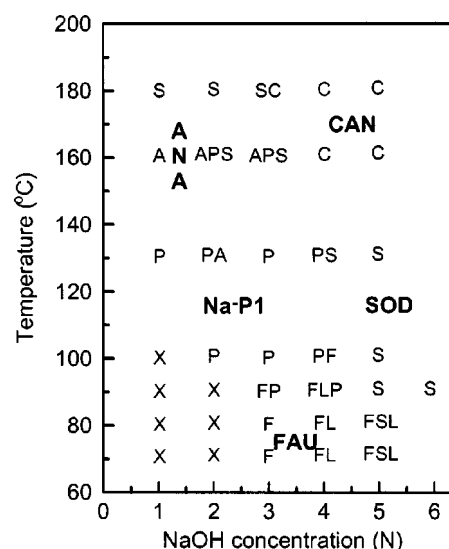


**Fig. 3. Conditions for formation of synthetic zeolites from fly ash.**
(A: analcime, C: cancrinite, F: faujasite, L: zeolite A, P: Na-P1, S: sodalite)

in the figure. Formation of faujasite was noted with a minor phase of zeolite A at lower temperature (70 °C), while Na-P1 zeolite predominated over faujasite at 100 °C. Only the Na-P1 phase was observed after 48 hours. As the temperature was increased to 160 °C, the cancrinite phase was stable. The fact that Na-P1 zeolite formed readily as equilibrium phase at elevated temperature (100-150 °C) demonstrates the fast growth rate of crystallization. Na-P1 zeolite does not crystallize at lower temperatures because of the slow formation of nuclei, while more complex and more easily nucleated faujasite dominates at 70-80 °C. These results are in accordance with the free energy-temperature relationship for the metastable phases [Breck, 1974]. The free energy curves for Na-P1 and cancrinite intersected at about 150 °C, while the curves for faujasite and Na-P1 intersected at about 90 °C.

As shown in Fig. 1b, the height of the mullite peak remained unchanged, while the decrease in the quartz peak was marked at 100 °C. Consequently, the increase in soluble silica content seemed to have accelerated the crystallization of Na-P1 zeolite. It has been reported that Na-P1 zeolite was more readily formed than zeolite A or faujasite from gels of higher silica content [Breck, 1974].

Table 1 and Fig. 1 show that the dissolution of quartz and mullite at high temperatures directly participates in zeolite synthesis. However mullite may be converted not to Na-P1, but to the analcime or cancrinite phase. Furthermore, the elemental composition of mullite, $Al_6Si_2O_{13}$, is outside the range of the Si/Al ratio for the synthesis of Na-P1 and faujasite. The types of reacting silica and alumina sources seem to be a critical factor for zeolite synthesis from fly ash.

## 5. Heavy Metal Removal Characteristics

Due to its high ion-exchange and selective adsorption properties, zeolite has many environmental applications, including removal of heavy metals; consequently, the heavy metal adsorption properties of the zeolitized fly ash were investigated.

Fig. 4 shows the removal efficiency of lead by using raw fly ash, natural zeolites and various synthetic zeolites. Raw fly ash showed a relatively small $Pb^{2+}$ removal efficiency (<8%). The natural zeolites showed better removal efficiencies, depending
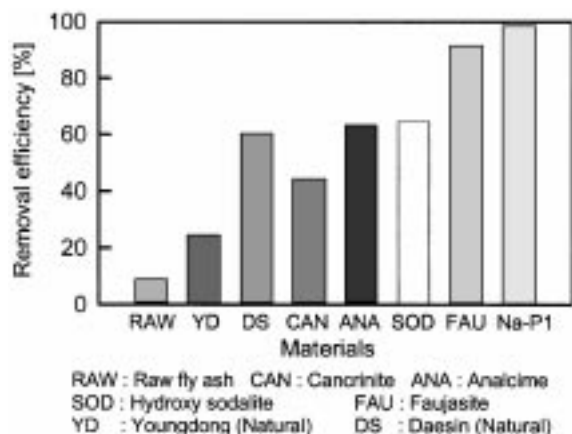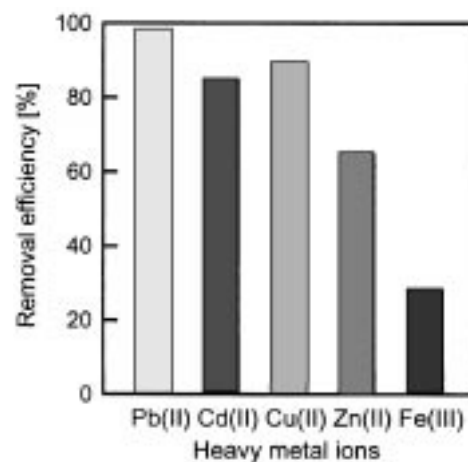


**Fig. 5. Comparison of removal efficiencies of heavy metals by Na-P1.**
(adsorbent: 0.1 g, metal ion concentration: 0.1 mM)

on their source, e.g., the natural zeolites from Yongdong and Daesin gave different removal efficiencies of 24% and 60%, respectively. The synthetic zeolite, Na-P1, showed the highest efficiency for lead removal (>98%).

Fig. 5 shows the removal efficiency of Na-P1 for various metal ions. The results show that Pb is adsorbed most by Na-P1 followed by $Cu^{2+}$, $Cd^{2+}$, and $Zn^{2+}$. Lead ion was the most efficiently removed cation, showing a value of more than 98%. Cu, Cd, and Zn were removed by 89, 85, and 65%, respectively. Therefore the cation affinity for Na-P1, in decreasing order, is as follows: $Pb^{2+}>Cu^{2+}>Cd^{2+}>Zn^{2+}$. It is likely that the removal ability differences among the various cations can be attributed to the differences in their hydration radius and energy. The order obtained is consistent with the decreasing radius of the hydrated metal ion [Moon and Jhon, 1986]. This result demonstrates the molecular sieving effect of synthetic zeolite. The smaller the radius of hydrated metal ion, and the larger the hydration energy
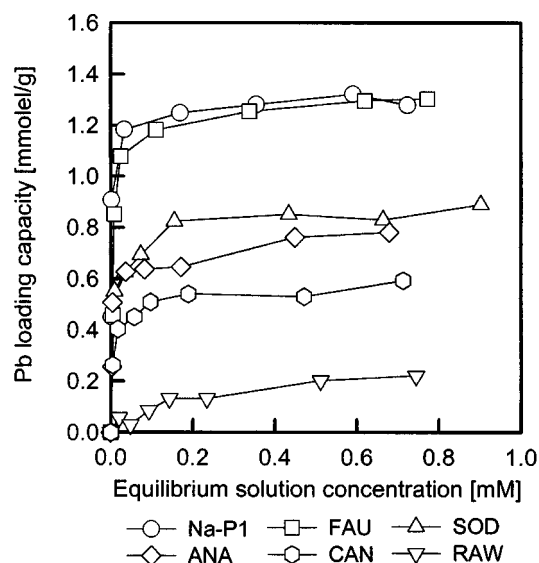


**Fig. 4. Comparison of lead removal efficiencies for each material.**
(adsorbent: 0.1 g, metal ion concentration: 0.1 mM)



**Fig. 6. The adsorption isotherms for synthesized zeolites.**
(adsorbent: 0.1 g, initial lead concentrations: 0.1-1.0 mM)

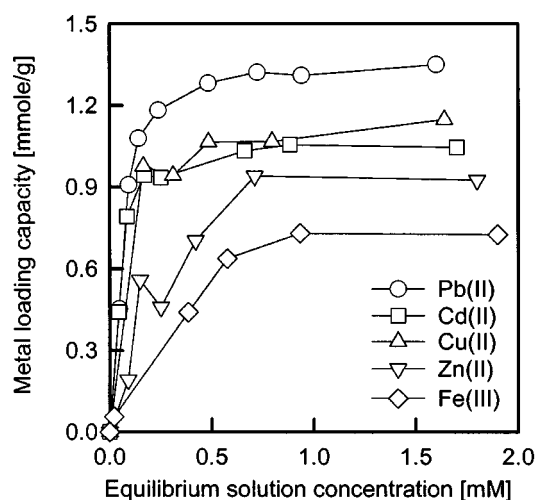**Fig. 7. The adsorption isotherms for Na-P1.**
(adsorbent: 0.1 g, initial lead concentrations: 0.1-3.0 mM)



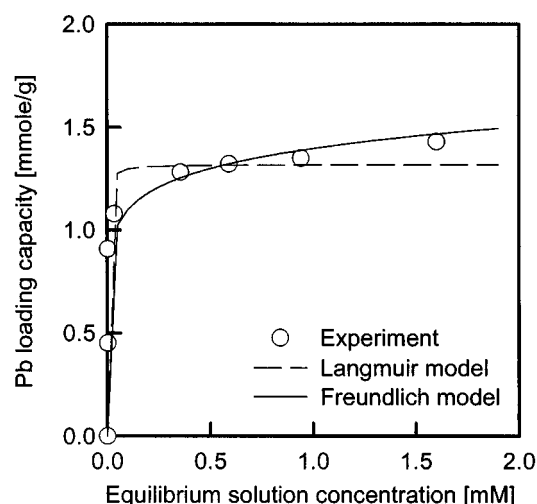**Fig. 8. Comparison of calculated values for Langmuir and Freundlich models with experimental data for Na-P1.**

of metal ion, the more difficult is it for the ion to enter the pores of the zeolite and undergo cation exchange at the available sites.

## 6. Adsorption Isotherms

Adsorption isotherm equations include constants which indicate the surface properties and affinity of the absorbent [Al-Duri et al., 1992]. Adsorption from aqueous solution at equilibrium is usually described by the Langmuir isotherm (Eq. (1)) or the Freundlich isotherm (Eq. (2)):

$$q = q_{max} \, b \, C_e / (1 + b \, C_e) \tag{1}$$

$$q = K \, C_e^{1/n} \tag{2}$$

**Table 2. Langmuir and freundlich isotherm parameters of various adsorbents for lead adsorption**

| Adsorbent | Langmuir parameters | | | Freundlich parameters | | |
|---|---|---|---|---|---|---|
| | $q_{max}$ (mmole g$^{-1}$) | B | $r^2$ | K | n | $r^2$ |
| Na-P1 | 1.286 | 457.29 | 0.869 | 1.308 | 38.46 | 0.944 |
| FAU | 1.229 | 211.34 | 0.421 | 1.383 | 8.82 | 0.673 |
| SOD | 0.798 | 214.32 | 0.812 | 0.921 | 9.31 | 0.921 |
| ANA | 0.745 | 214.74 | 0.860 | 0.891 | 6.94 | 0.743 |
| CAN | 0.537 | 205.66 | 0.972 | 0.653 | 6.97 | 0.882 |
| YD[a] | 0.219 | 8.911 | 0.769 | 0.329 | 2.67 | 0.968 |
| DS[a] | 0.339 | 19.69 | 0.685 | 0.419 | 2.74 | 0.934 |
| Fly ash | 0.186 | 24.87 | 0.714 | 0.261 | 2.34 | 0.987 |

[a]Untreated natural zeolite from Yongdong and Daesin.

**Table 3. Langmuir and freundlich isotherm parameters of various heavy metal ions for na-p1**

| Adsorbate | Langmuir parameters | | | Freundlich parameters | | |
|---|---|---|---|---|---|---|
| | $q_{max}$ (mmole g$^{-1}$) | B | $r^2$ | K | n | $r^2$ |
| Pb | 1.286 | 457.29 | 0.869 | 1.308 | 38.46 | 0.944 |
| Cd | 1.046 | 103.92 | 0.655 | 1.073 | 17.42 | 0.866 |
| Cu | 1.156 | 21.91 | 0.688 | 1.086 | 14.75 | 0.820 |
| Zn | 1.007 | 8.29 | 0.918 | 1.006 | 3.75 | 0.993 |

The adsorption isotherms of lead for the fly ash and synthetic zeolites are shown in Fig. 6; isotherms of various heavy metal ions for Na-P1 are shown in Fig. 7.

The isotherm parameters were calculated by linear regression (Tables 2 and 3). In the Langmuir equation, $q_{max}$ is theoretical maximum adsorption. Thus, the maximum lead uptake of fly ash is about 0.19 mmole g$^{-1}$ and that of natural zeolite is 0.22-0.34 mmole g$^{-1}$; however, the maximum uptake of Na-P1 is about 1.29 mmole g$^{-1}$. Therefore, it is clear that the synthetic zeolite has a much higher adsorption capacity compared with the fly ash and the natural zeolites. In the Freundlich equation, in addition, K is the capacity of adsorption and n is the intensity of adsorption. Therefore, as the value of 1/n decreases, the affinity decreases more. Fig. 8 illustrates the comparison of values calculated by using the Langmuir and Freundlich equations with experimental data for lead adsorption. The data obtained in this study fit both models, although lead adsorption by Na-P1 is better described by the Freundlich than the Langmuir adsorption isotherm in the concentration range of 0.1 to 2.0 mM. The values of K, n, and correlation coefficient ($r^2$) for biosorbent sorption systems are summarized in Tables 2 and 3. The values of n obtained were in the range of 2.34-34.46. Treybal [1980] demonstrates that the magnitude of n is an indication of system suitability, with values of n>1 representing favourable adsorption conditions. Judging from this, these n values obtained from experiment indicate that zeolites synthesized in this study are favourable adsorbent materials. The values of K giving a measure of adsorbent capacity decreased in the following sequences: Na-P1>FAU>SOD>ANA>CAN>YD>DS>Fly ash.

## 7. Adsorption Rate Model

For adsorption processes, the adsorption rate should be considered as well as the adsorption isotherms. Although the adsorption isotherm provides useful data on the maximum adsorption capacity and adsorbent affinity for the metal, it cannot provide information on the metal adsorption capacity of the adsorbent per unit time necessary for determining the contact time which directly affects the design of equipment and operating costs when an adsorption process is used for water and waste-
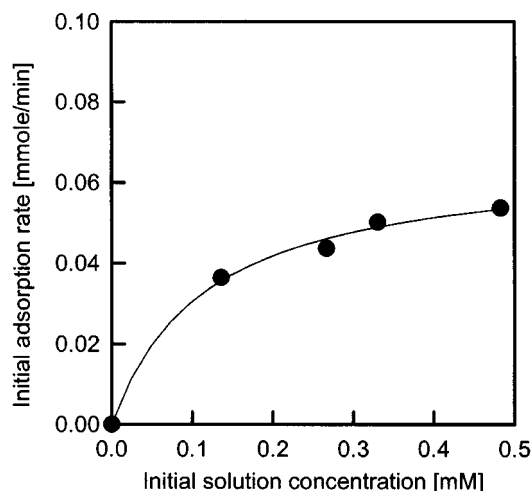
**Fig. 9. Effect of initial solution concentration on initial adsorption rate profile.**

water treatment. Not only does the information on the adsorption rate provide a useful basis for selecting a suitable adsorbent for the adsorption process, but it can also aid one in elucidating the adsorption mechanism.

Using the initial rate method [Fogler, 1999], we calculated the adsorption rate from the experimental data and a developed rate model equation, and the results are shown in Fig. 9. The relation between adsorption rate and the initial concentration of lead can be expressed as follows:

$$r_{ad} = \frac{r_{max}C_o}{\alpha + C_o} = \frac{0.066C_o}{0.117 + C_o}$$

## CONCLUSIONS

Various kinds of zeolite including Na-P1, faujasite, hydroxy sodalite, analcime and cancrinite were produced by heating fly ash in sodium hydroxide solution. Both temperature and base concentration are critical in determining the nature of the phase formed.

The synthetic zeolites showed a high affinity for metal ion and a better removal efficiency than raw fly ash and natural zeolites. Among the synthetic zeolites produced here, Na-P1 exhibited the highest affinity for metal with a maximum removal of 1.29 mmole Pb g$^{-1}$, which corresponded to 98% removal efficiency, while raw fly ash gave less than 8% removal efficiency (0.19 mmole Pb g$^{-1}$ removal), and natural zeolites from Yongdong and Daesin gave a removal efficiency of 24% and 60%, respectively (0.22-0.34 mmole Pb g$^{-1}$ removal).

Selectivity of Na-P1 for metal removal was, in decreasing order: Pb$^{2+}$>Cu$^{2+}$>Cd$^{2+}$>Zn$^{2+}$. This trend was consistent with the decreasing order of the radius of the hydrated metal ion. The adsorption isotherm of lead by Na-P1 fitted the Freundlich isotherm well rather than the Langmuir isotherm in the concentration range of 0.1 to 2.0 mM.

## ACKNOWLEDGEMENT

## NOMENCLATURE

b    : constant related to the energy of net enthalpy of adsorption [-]
$C_o$    : initial lead concentration in solution [mM]
$C_e$    : concentration of lead in solution at equilibrium state [mM]
K    : indicator of adsorption capacity [-]
$1/n$    : adsorption intensity [-]
q    : amount of heavy metal adsorbed per unit weight of adsorbent [mmole g$^{-1}$]
$q_{max}$    : amount of heavy metal adsorbed per unit weight of adsorbent [mmole g$^{-1}$]
$r_{ad}$    : initial adsorption rate of heavy metal [mmole g$^{-1}$ min$^{-1}$]
$r_{max}$    : maximum adsorption rate [-]
$\alpha$    : adsorption rate constant [-]

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