Dealumination Treatment of Zeolite for Krypton Gas Adsorption

Jin-Myeong Shin, Seol-Woo Shin, Jang-Jin Park, Ho-Hee Lee, Myung-Seung Yang
Dept. of DUPIC Facility and Process Waste Management, KAERI

1. Introduction

The concept of the direct use of spent fuel in CANDU reactors (DUPIC) is a dry processing technology to manufacture CANDU fuel from spent PWR fuel material. The heart of the DUPIC fuel cycle is the OREOX (Oxidation and Reduction of Oxide fuels) process. During this dry process, uranium from spent PWR fuel is oxidized and reduced to a fine powder, which forms the starting material for fabrication of DUPIC fuel pellets. During the OREOX process of DUPIC fuel fabrication, krypton is released as a noble fission gas. As fuel burnup proceeds, more fission products are formed. About 15 % of the fission products are inert gases such as xenon and krypton. The dealumination of zeolite is examined in this study in order to know that the structural changes in zeolite and the effect of krypton adsorption on zeolite.

2. Experimental

2.1 Dealumination

Zeolite (Molecular Sieve 5A, 8-12 mesh) spheres were obtained from the Aldrich Company. The materials were washed with boiling water and dried for 12hours at 105℃, then contacted with 105℃ steam for 12hours. That spheres were mixed using 0.1 to 10 N HCl solutions (0.5N, 1N, 2N, 5N) (10mL/g zeolite) and stirred at 50-60 rpm for 12 hours. Samples were carefully washed with water till the water above the zeolite was clear. Samples were dried for 12hours at 105℃ and then stored silica-gel desiccator.

2.2 Krypton gas adsorption

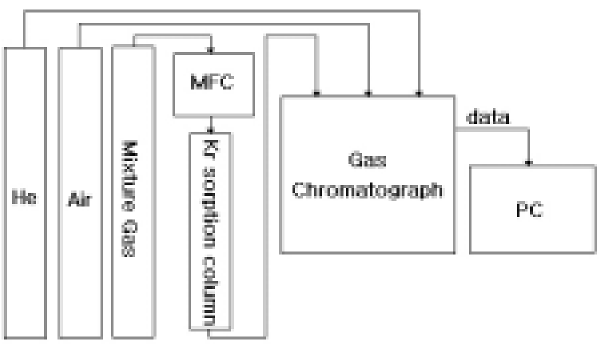


Figure 1. The Kr adsorption system and GC analyzer.

The mixture gas (Kr 300pm, H_2 4%, Ar balance) is injected into the krypton adsorption system (see Fig. 1); in the first stage it passes through the Mass Flow Controller. The gas flow rate is set 30mL/min and then the gas is pushed into the Kr adsorption column. The adsorption column packed with zeolite treated with HCl solution and the mixtutre gas passed through the column and goes to the GC analyzer.

The chromatogram outputs of GC are automatically recorded and analyzer by PC.

2.3 Analytical Methods

The krypton adsorption amount on zeolite was measured by Gas Chromatograph analysis equipped with TCD and GS-Molsieve column was used. Pure Helium gas was uesd as carrier gas (4.6mL/min).. X-ray diffraction (XRD) techniques were used to determine the crystal structure of a zeolite. The Surface area and pore volume were measured by BET analyzer. The surface morphology was measured by SEM

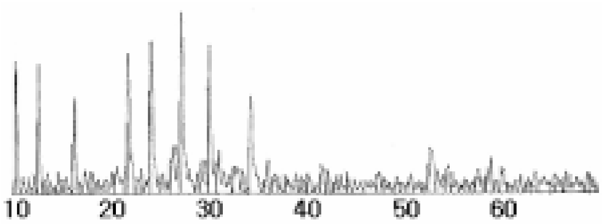
2. Results

Table 1 shows the effect of HCl solution concentration on krypton sorption amount. The krypton sorption amount on zeolite treated with 2N HCl solution is superior to the zeolites treated with 0.5N, 1N, and 5N HCl solution and not treated zeolite.

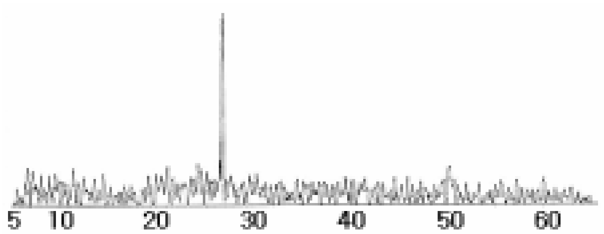
Table 1. Effect of HCl solution concentration (on krypton adsorption)

HCl solution conc.	Krypton sorption amount (g Kr/ g zeolite)
No HCl	0.55
0.5N	0.17
1N	0.35
2N	1.52
5N	0.001

Figure 2 shows the change of crystal structure of zeolite.



(a) untreated with HCl solution



(b) treated with 2N HCl solution

Figure 2. X-ray diffraction of zeolites.

Figure 3 and Figure 4 display the changes of pore size distribution and pore volume in zeolite. After treatment zeolite with 2N HCl solution, the pore diameter under 100A pore volume increases.

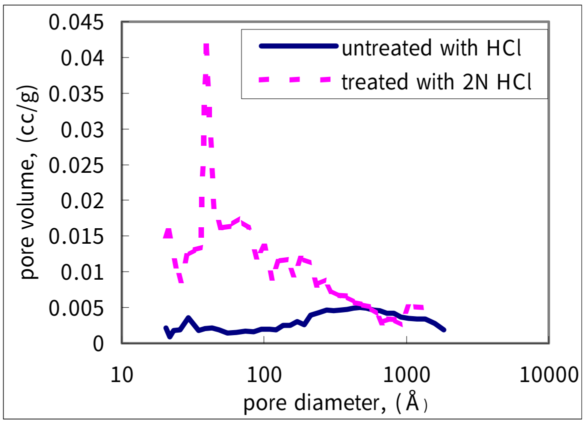


Figure 3. Pore size distribution.

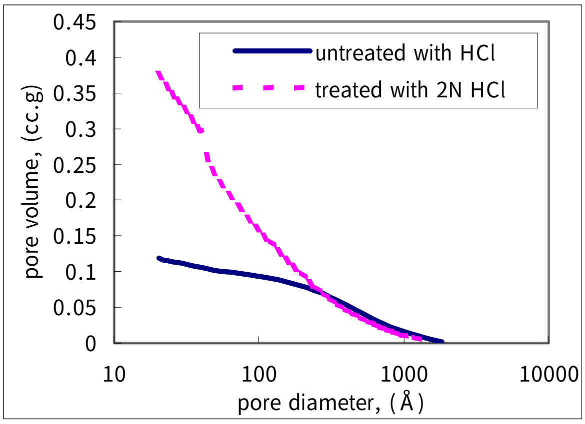
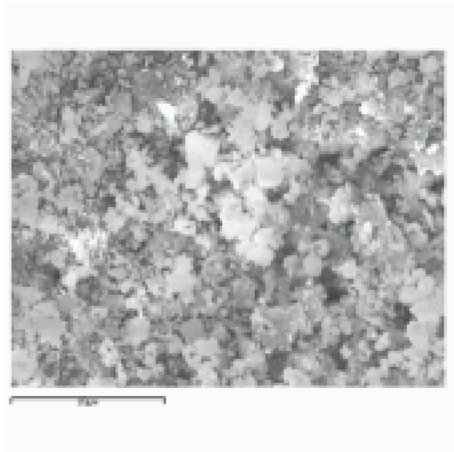


Figure 4. Cumulative pore volume.

Figure 5 shows the SEM photographs of the surfaces of zeolite before and after treatment with 2N HCl solution. The SEM photographs show the change in the surface structure of the zeolite.


(a)

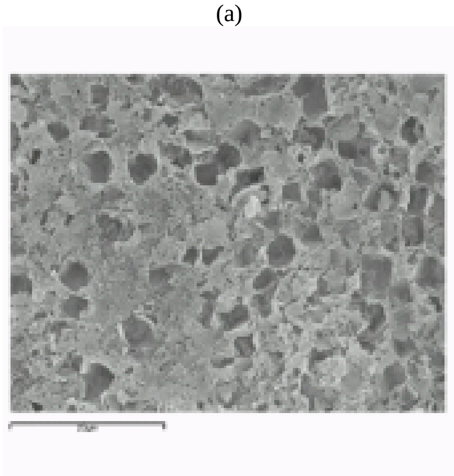

(b)

Figure 5. SEM photographs of the surfaces (a) no treated zeolite, (b) zeolite treated with 2N HCl solution.

3. Conclusion

Kr adsorption capacity of dealuminated zeolite 5A with HCl solution was executed. The krypton sorption amount on zeolite treated with 2N HCl solution was increased about 3 times on no treated zeolite. The crystal structure of zeolite was changed by HCl solution treatment. The micropore volume of zeolite treated with 2N HCl solution was increased. The surface of zeolite was changed by HCl solution.

REFERENCES

[1] H. S. Park et al., "The DUPIC Fuel Cycle Alternative: Status & Perspective ", Proceedings of the 10th PBNC, 1996, Kobe, Japan.

[2] Green, R., and Boczar, P., Advanced Fuel cycles in CANDU Reactors : Reconfirming the Miid, AECL-10156, 1990.

[3] Liu, Z., Semeniuk, D., et al., Report on the HOX2 Experiment : Fission-Product Release and Trapping during the OREOX and Sintering Processes, AECL-EACL, DUPIC-AE-022, Fuel Safety Branch Chalk River Lab., 1998, Canada.