WATER VAPOR ADSORPTION ISOTHERMS IN NaX, CaA, AND NaCaA ZEOLITES

Ergashev Oybek Karimovich,

Nuridinov Olimjon Kutbidinovich
Namangan Institute of Engineering and Technology, Uzbekistan.
E-mail: nolimjon@mail.ru

Annotation

Adsorption and thermodynamic studies of water vapor adsorption in NaX zeolites, as well as in CaA and NaCaA with different degrees of exchange of Na cations for Ca at temperatures of 273, 298, 333, and 373K, have been carried out. A number of characteristic features of water adsorption in NaX, CaA, and NaCaA zeolites have been revealed.

Keywords: Isotherm, heat of adsorption, entropy, zeolites, NaX, CaA, NaCaA, adsorption calorimetry.

One large cavity of the NaX zeolite studied by us accounts for an average of 10.8 sodium cations, including 2 cations in the S_I and $S_{I'}$ positions (inside hexagonal prisms and at six-membered oxygen rings connecting cuboctahedrons and hexagonal prisms), 4 cations in the position S_{II} (in six-membered rings linking cuboctahedrons and large cavities) and an average of 4.8 cations in position S_{III} (in a large cavity). In type X zeolite, there are two types of four-membered rings - rings of cuboctahedrons, the number of which is 48 per e.i. and rings of prisms, the number of which is 96 per e.i. Joints between four-membered oxygen rings are preferred, on which, apparently, S_{III} cations are located.

Zeolites CaA and NaCaA are obtained by ion exchange from zeolite NaA. Thus, zeolite CaA was obtained by replacing most of the Na cations with Ca. All these cations are located in the SI position, in the center of the six-membered oxygen rings of the large cavity. Zeolite CaNaA is characterized by a high content of Na cations. Most of them are located in the S_I position, but a small part is also in the S_{II}.

Water adsorption on zeolites has a number of characteristic features. Figures 1, 2 and 3 present water vapor adsorption isotherms on various types of NaX, CaA, and CaNaA zeolites and at temperatures of 273, 298, 333, and 373 K.

The samples were preliminarily subjected to thermal vacuum treatment at a temperature of 350°C. All studied zeolites are characterized by a very steep rise in isotherms in the region of low water vapor concentrations. The initial equilibrium pressure in the system is below 10^{-4} mm Hg. pillar. The adsorption capacity of zeolites at room temperature already at a pressure of several mm Hg. column is close to the adsorption capacity at maximum saturation. This indicates the promise of using zeolites for drying gases with a low content of water vapor.

The second distinguishing feature of vapor adsorption in NaX, CaA, and NaCaA zeolites is the relatively low dependence of the adsorption capacity on temperature. Even at 100^0C, zeolites are able to absorb large amounts of water. At these temperatures, the adsorption capacity of silica gel and alumina is negligible. From the foregoing, we can conclude that the use of zeolites is promising for the combined method of gas purification from carbon dioxide and its deep drying.

It is known [107] that adsorption on microporous zeolites proceeds according to the volume filling mechanism. Using the provisions of Polyani's theory and generalizing a large experimental material, Dubinin M.M. and colleagues came to the conclusion that it is possible to use the Weibull distribution function as a distribution function of the adsorption volume by the value of the potential to describe adsorption on microporous adsorbents. As applied to the distribution of the degree of filling over the adsorption potential, the Weibull distribution function is represented by the relation:

$$\theta = \exp[-(A/E)^n] \qquad (1)$$

where E and n are temperature-independent parameters. The value of E is called the characteristic energy of adsorption. The exponent n is expressed as integers from 1 to 6 depending on the structure of the adsorbent. A is the work of adsorption, i.e. the work of transferring 1 mol of gas from the surface of a liquid adsorbate (pressure Ps) to the equilibrium gas phase (pressure P):

$$A = RT\ln(P_s/P) \qquad (2)$$

The degree of filling of the adsorbent can be represented as the ratio of the adsorption value a to the maximum adsorption a_o. Then from equation (1) we get:

$$a = a_0\exp[-(A/E)^n] \qquad (3)$$

Equation (3) is the general equation of the theory of volumetric saturation of micropores (VOM). In logarithmic form, it has a linear form:

$$\ln a = \ln a_0 - \frac{A^n}{E^n} \qquad (4)$$

At one time, the case of a wide distribution of micropores was considered, which is observed, for example, for activated carbons with high degrees of activation. The expediency of using the two-term equation of Izotova T.I. in this case was shown. and Dubinina M.M.:

$$a = (W_{01}/V^*)\exp[-(A/E_1)^2]+(W_{02}/V^*)\exp[-(A/E_2)] \qquad (5)$$

with different characteristic adsorption energies E_1 and E_2, moreover, E_1 and W_{01} express adsorption in micropores, and E_2 and W_{02} ($E_1>E_2$) in the modern interpretation - in supermicropores. In this equation, V* is the molar volume of the adsorbed substance. Parameters W_{01}, E_1, W_{02} and E_2 are easily determined from one experimental adsorption isotherm for a wide range of equilibrium relative pressures, for example, $1\cdot10^{-6} \div 5\cdot10^{-1}$. Parameters with index 1 are the characteristics of the microporous structure, and those with index 2 are the characteristics of the supermicroporous structure of the carbon adsorbent. The latter is an important characteristic of the ability of active carbon to desorb relatively well adsorbed substances, for example, amyl acetate. Adsorption isotherms of substances on zeolites in many cases are practically not completely described by the two-term equation (5). The authors of showed that the adsorption isotherm can be completely described from zero filling to saturation by the three-term VOM equation.

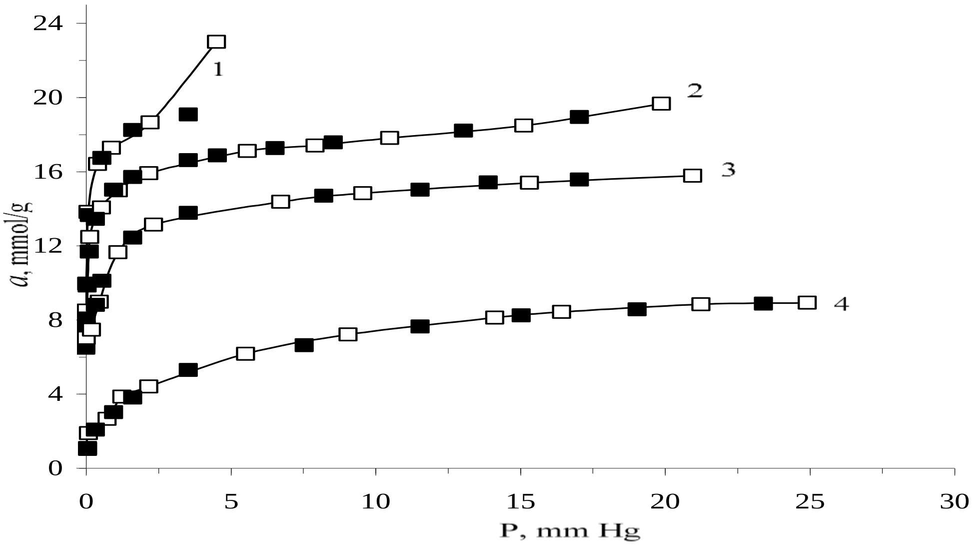

Fig 1. Water vapor adsorption isotherms in NaX zeolite:
1- 273 K; 2- 298 K; 3- 333 K; 4- 373 K. ■— calculated using VOM.

All studied zeolites are characterized by a very steep rise in isotherms in the region of low water vapor concentrations. The initial equilibrium pressure in the system is below 10^{-4} mm Hg. The adsorption cap acity of zeolites at room temperature already at a pressure of several mm Hg. column is close to the adsorption capacity at maximum saturation. This indicates the promise of using zeolites for drying gases with a low content of water vapor.

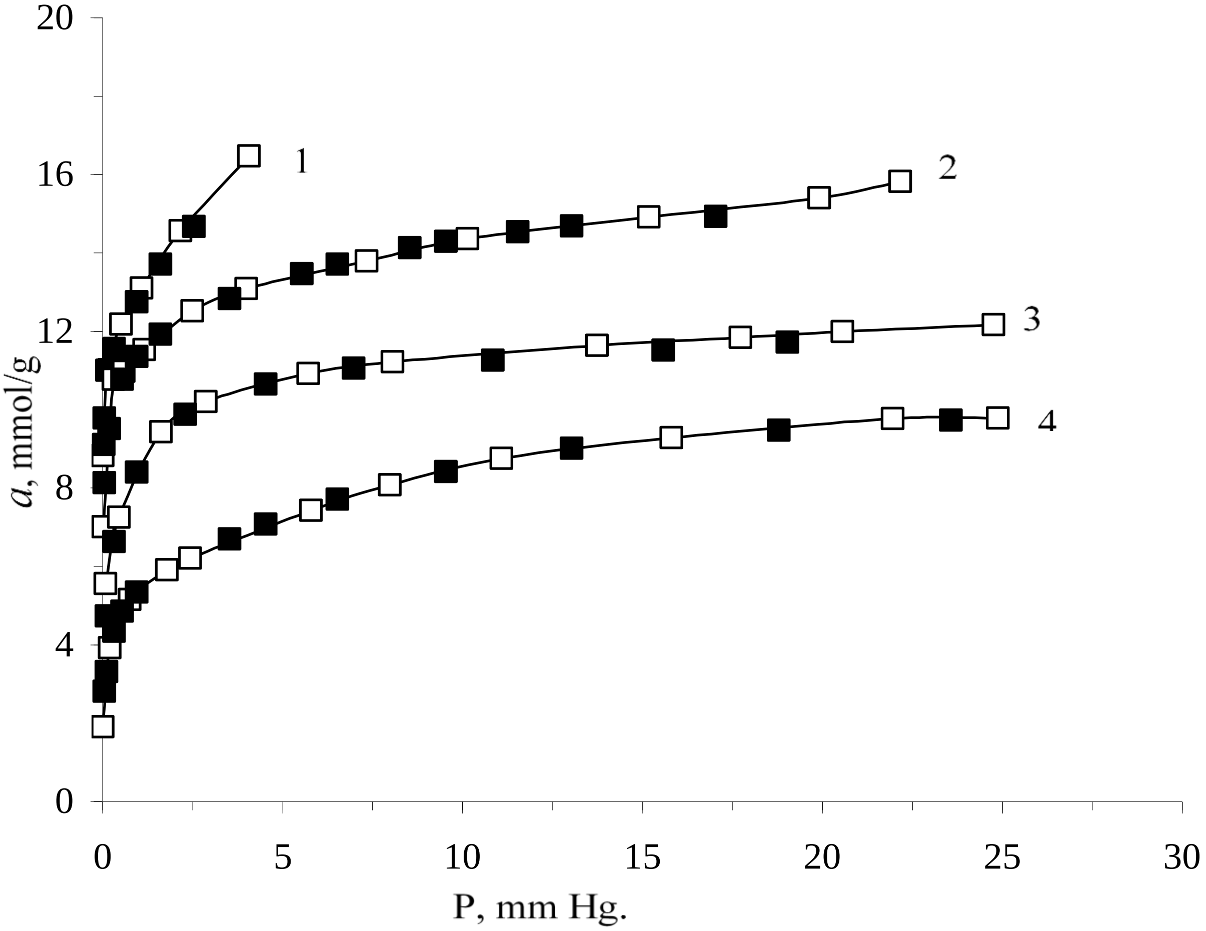

Fig 2. Water vapor adsorption isotherms in CaA zeolite:

1-273 K; 2- 298 K; 3-333 K; 4-373 K. ■ – calculated using VOM.

The second distinguishing feature of vapor adsorption in NaX, CaA, and NaCaA zeolites is the relatively low dependence of the adsorption capacity on temperature. Even at 373 K, zeolites are able to absorb large amounts of water. At these temperatures, the adsorption capacity of silica gel and alumina is negligible. From the foregoing, we can conclude that the use of zeolites is promising for the combined method of gas purification from carbon dioxide and its deep drying.

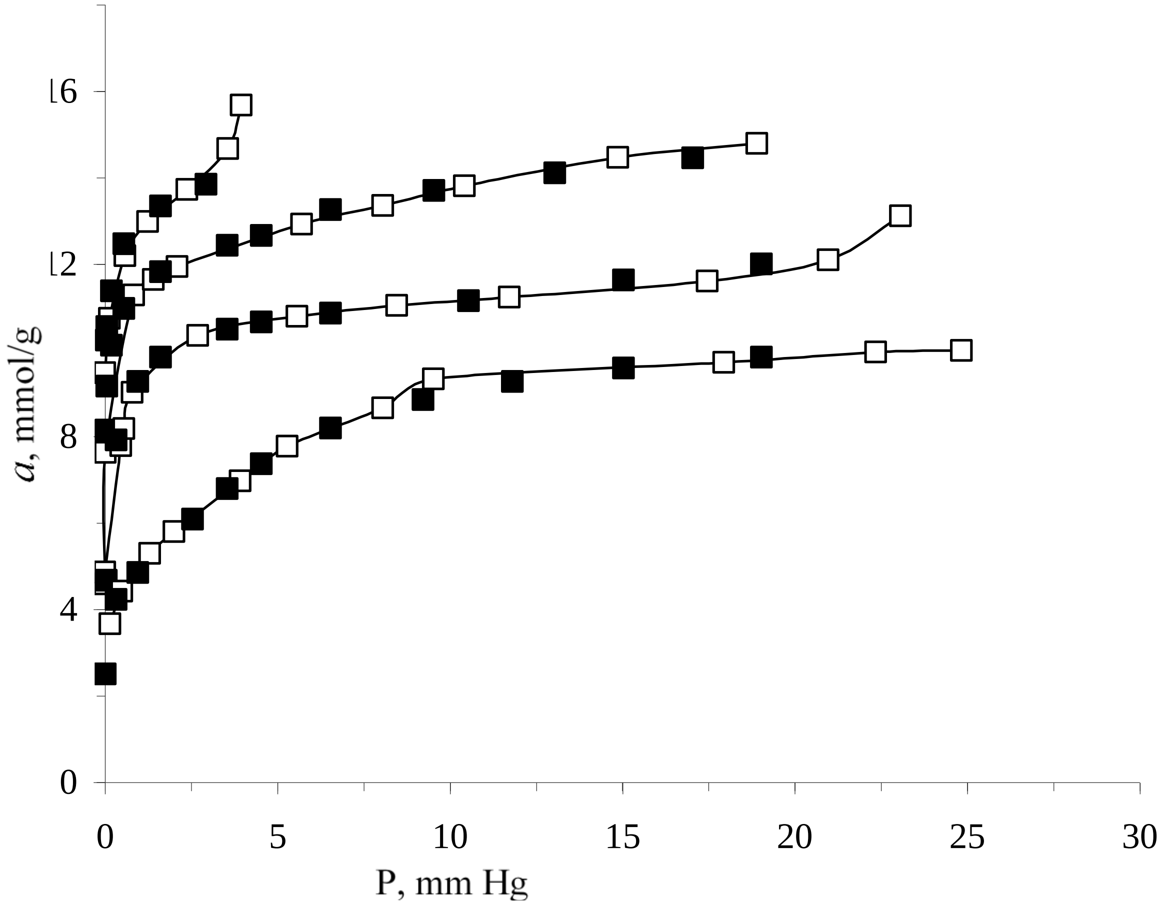

Figure 3. Water vapor adsorption isotherms in CaNaA zeolite:

1-273 K; 2- 298 K; 3-333 K; 4-373 K. ■ - calculated using VOM.

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