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# Behaviour of Zeolite A, Faujasites X and Y Molecular Sieves in Nitrogen Gas Adsorption

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**Abstract:** In this study, we were interested to study the behaviour of some molecular sieves in nitrogen gas adsorption. We were interested in first stage in the synthesis of the molecular sieves as zeolite A and faujasites X and Y. These solid crystals were the subject of a characterization by various analytical techniques such X-ray diffraction, X-ray analysis by energy dispersion and electronic scanning microscopy. The adsorption isotherms of nitrogen on these samples are obtained of type I and therefore, the Langmuir equation was verified. X and Y zeolite samples could adsorb a big amount of gas nitrogen not like that A sample witch has a small adsorption capacity. It has been also shown that the desorption speed of nitrogen is higher than that adsorption on study zeolites.

**Key words:** Zeolite A, faujasites X and Y, Langmuir isotherm, adsorption, nitrogen

## INTRODUCTION

The current industrial needs require adsorbent materials, commonly called zeolites, having a well defined microporous texture (Breck, 1974). From a general point of view, the use of a zeolite in a process given by these characteristics of adsorption, which are primarily according to its microporous structure, its specific surface and its chemical and thermal stability, which confers molecular properties of sieve and the capacity to them to adsorb organic and inorganic molecules (Lovat *et al.*, 2001). Zeolites have much applicability in particular drying, separation of gases (Bachmann and Tissot, 2001), the purification of petrochemical gases and the catalysis (Nibou and Amkrane, 2005), as well as other no conventional branches.

Generally, the basic principles of physical adsorption on solids are known. The adsorption of gases and vapors by zeolite solids is not as complex as adsorption phenomena on amorphous, less defined, solids. This is due to the regularity in the internal pore system which provides for the type I isotherm of the Brunauer-Emett-Teller classification or the Langmuir type isotherm. Different adsorbates were used in evaluation to the porous volume of zeolites as $H_2O$, $CO_2$, $CH_4$, Ar, $N_2$ and others. It was found that they obey to the Gurvisch rule (Barrer, 1982), but some notable deviations are observed. In this context, we have studied the $N_2$ adsorption behaviour on A, X and Y synthesized zeolites using standard BET model and Dubinin-Polanyi equation.

## MATERIALS AND METHODS

**Characterization of materials:** In this study, we were interested to prepare 3 structures as zeolite A, faujasite X and Y according to the hydrothermal crystallization method (Khemaissia *et al.*, 2007). The composition of the initial gels are respectively $0.4Na_2O$ $0.5Al_2O_3$ $1.2SiO_2$ $92H_2O$, $1.1Na_2O$ $0.2Al_2O_3$ $1.0SiO_2$ $92H_2O$ and $1.4Na_2O$ $0.3Al_2O_3$ $2.5SiO_2$ $84H_2O$.

The synthesis was carried out in Teflon autoclave under various operating conditions. After crystallization, all the products were characterized by several techniques. The structure identification of synthesized materials was determined by X-ray diffraction. The Samples were identified as pure faujasites X and Y and zeolite A. Their X-ray diffraction spectra were compared and found in good agreement with those of simulated X-ray diffraction data (Traecy *et al.*, 2001).

The composition of solid samples was determined by X-ray analysis by energy dispersion. Si/Al ratios of A, X and Y zeolites have the values of 1.1, 2.2 and 4.1, respectively. The morphology of the grains of zeolite A and faujasite Y is uniform having a cubic geometry (Fig. 1).

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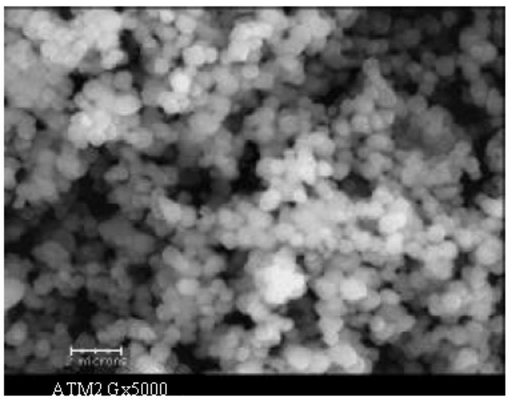
(a) Zeolite A




(b) Faujasite Y

Fig. 1: Scanning electron micrographs of samples (a) and (b)

**Adsorption of nitrogen on zeolites A, X and Y by BET method:** The adsorption technique by BET method was investigated using an apparatus of type Micrometrics 2100E. The last includes a system of manifold with valves which inter-connects supports of samples, two valves of control of adsorbed, a valve of helium control, a system of measurement of pressure, a system of evacuation which east consists of a cold nitrogen trap door liquidates, a mechanical pump and an oil diffusion pump. The system is able to evacuate well a sample with the lower part of 0.001 mmHg and able to go down to pressure even lower.

Heating sleeves are connected to degas the samples exposed to a high temperature. These sleeves contain a built-in thermocouple which allows the reading of the temperature for each sample. The maximum of temperature of degasification is of 400°C. A thermistor thermometer is built-in for the temperature measurement of the liquid nitrogen which is employed to cool the sample during the analysis. The $CO_2$ or water molecules are evacuated by a degasification of the vacuum sample and at the suitable temperature (by taking care to respect the physical properties of the sample).

## RESULTS AND DISCUSSION

With an aim of evaluating the sizes characteristic of a microporous solid adsorbing, a study was carried out for three types of zeolites: Zeolite A, faujasite X and faujasite Y. The study makes it possible to understand the phenomenon of adsorption. The studied parameters are: Specific surface, volume of the micropores and adsorption isotherm.

**Measurement of specific surface:** The determination of specific surface is an immediate application of BET equation. The chart of the transform of BET equation for the three cases of adsorption (Zeolite A, faujasites X and Y) while carrying $P_2/V_a(P_s\text{-}P_2)$ according to the relative pressure ($P_2/P_s$) is shown on Fig. 2 and 3.

The adsorption isotherm takes into account only relative pressures ranging between 0.05 and 0.35 (field of validity of BET equation). This range is sufficient for the determination of specific surface. But, it is significant to note that the adsorption process can be prolonged until a relative pressure close to 1. The calculation of the specific area is based on the analytical treatment of the adsorption isotherm determined in experiments; it is thus possible to define the quantity of gas adsorbed in full-course supplements then to calculate the surface of this layer.

**Determination of microporous volume:** Adsorption also makes it possible to know an additional characteristic which is the porous total volume of the sample using the equation of Dubinin-Polanyi (Eq. 1):

$$\log V_a = \log V_0 - D\left[\log\left(\frac{P}{P_0}\right)\right]^2 \qquad (1)$$

Where $V_0$ represents the volume of the micropores.

The layouts of Log(Va) according to $[\text{Log}(P_2/P_S)]^2$ are lines of which ordered in the beginning allows to calculate volumes of the micropores (Fig. 4).

According to the results, one notices that the volume of the micropores of the faujasite X (0.089 $cm^3\ g^{-1}$) is low compared to that of faujasite Y (0.126 $cm^3\ g^{-1}$). On the other hand, the effective surface of faujasite X is equal to 501 $m^2\ g^{-1}$ (surface accessible to the molecules from gases) and its capacity of adsorption is equal to 0.115 $cm^3$ (expressed in volume adsorbed on monolayer).

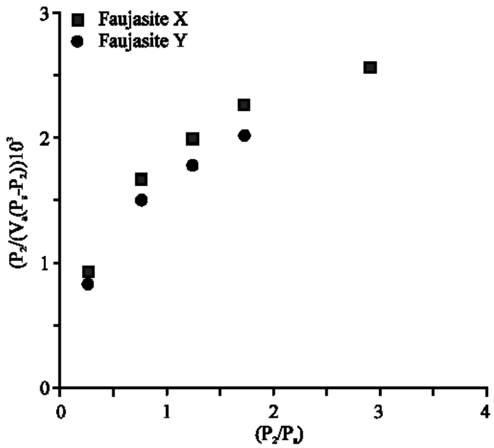



Fig. 2: Faujasite X and Y Fourier's transform

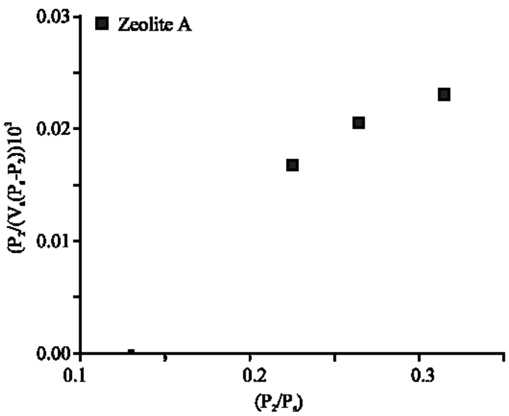



Fig. 3: Zeolite A Fourier's transform

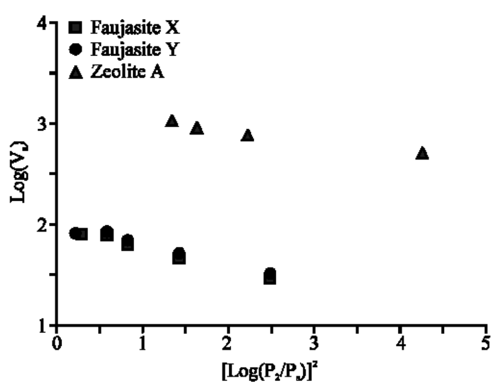



Fig. 4: Log ($V_a$) according to [Log ($P_2/P_S$)]$^2$ of faujasites X and Y and zeolite A

These values are larger than that of faujasite Y (450 $m^2$ $g^{-1}$; 0.111 $cm^3$). For zeolite A, one noticed that effective surface is about 61.64 $m^2$ $g^{-1}$ and the capacity of adsorption is roughly equal to 0.015 $cm^3$.

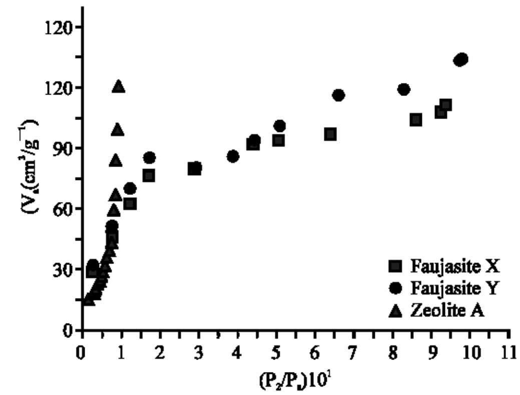



Fig. 5: Isotherms of adsorptions of nitrogen on faujasites X and Y and zeolite A

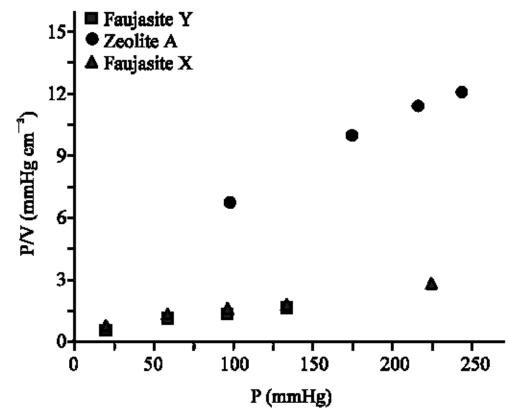



Fig. 6: Transform of Langmuir of faujasites X and Y and zeolite A

**Determination of the type of the isotherm of adsorption:** According to the classification of BET method, one notices that the adsorption isotherm of nitrogen on faujasites X and Y is probably of type I as shown in Fig. 5. In this case, the dimension of the pores varied approximately from 15 to 1000 Å and generally, the inflection reaches near of the end of the first layer. On the other hand, the adsorption isotherm of nitrogen on zeolite A can be classified either of type I or of type II. But the pace of the latter is not clear thus one must check the type of isotherm one using the approach of Langmuir.

When the adsorption isotherm is of type I, one must check the validity of the Langmuir relation sheep according to the Eq. 2 (Fremaux, 1989).

$$\frac{P}{V} = \frac{1}{b \cdot Vm} + \frac{P}{Vm} \quad (2)$$

where $V_m$: Being volume in ($cm^3$ $g^{-1}$) of adsorbed gas when all the sites are occupied (full-course),

b : The speed ratio of adsorption and desorption of gas in the studied system,
P : Pressure and
V : The total volume of gas.

The values of (P/V) versus the pressure (P) are indicated in Fig. 6. The drawings are right with ($V_m^{-1}$) as slope and ($b.V_m^{-1}$) as origin ordinate. The theory of Langmuir is verified for used zeolites, therefore the adsorption isotherm of nitrogen on faujasites X and Y and zeolite A is of type I and the solid is composed of the micropores.

The adsorbed volume ($V_m$) for faujasite Y, faujasite X and zeolite A is equal to 0.112, 0.108 and 0.027 $cm^3\ g^{-1}$, respectively.

The speed ratios of adsorption and desorption (B) are respectively 0.017, 0.014 and 0.012. Thus, the desorption speed is higher than that adsorption for the three used zeolites.

## CONCLUSION

The interest of this study is to compare the behaviour of three zeolites which have different molecular structures and kinetic diameters. The BET method was allowed to show that zeolites having higher specific areas could adsorb a big amount of nitrogen. Results of calculated specific areas have shown that the structure is microporous. The adsorption isotherms of nitrogen on studied zeolites are of type I and the Langmuir equation is verified.

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