CEAC-R

4-89

COMISION DE ENERGIA ATOMICA DE CUBA

CEAC - R -- 4 - 89

TITULO:

# DETERMINATION OF WATER CONTENT IN NATURAL ZEOLITES BY REFLECTION METHOD.

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Clasificación temática del INIS

B 31. 20 NUCLEAR METHODS

DESCRIPTORES

MEASURING INSTRUMENTS
HYDROGEN METERS
SLOWING-DOWN
THERMALIZATION
BF3 COUNTERS
MOISTURE GAGES
ZEOLITES

Aprobado por el Consejo Científico Ramal de
la Esfera Nuclear el día 14 de noviembre de 1988.

# Determination of water content in natural zeolites by neutron reflection method.

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# DETERMINATION OF WATER CONTENT IN NATURAL ZEOLITES BY NEUTRON REFLECTION.

Abstract Water content in natural zeolites collected from different site places in Cuba has been determined by neutron reflection method. Results show that it is possible to separate the minerals abundant in zeolite from the surrounding barren rocks. Water content of about 10 w% can be determined with 2-3 % relative accuracy for different matrices, using 10 m measuring time.

Resumen El contenido de agua de zeolitas naturales de diferentes yacimientos ha sido determinado por reflexión de neutrones. Los resultados indican que es posible distinguir un mineral rico en zeolita de los otros minerales que le rodeen. Un contenido de agua de alrededor de 10% puede ser determinado con una exactitud relativa de 2-3% para diferentes matrices, usando 10 m como tiempo de medición.

## INTRODUCCION

During the last decades extensive investigations were devoted to the study of the chemical composition of zeolites because of their increasing role both in science and technology. Gottardi and Galli [1] have given a comprehensive review on the properties of zeolites which depend mainly on the topology of their frameworks.

In previous works the neutron reflection methods was used for the fast nondestructive determination of the hydrogen content of crude oil [2], chlorine content in chlorinate polyethylene [3] and bitumen content in asphalt concrete [4].

The aim of this work was to determine the water content in zeolites of different origins collected from Cuba.

## EXPERIMENTAL PROCEDURE.

The water content of zeolite has been determined by thermal neutron reflection method. In a previous experiment it was found that the relative excess in the counting rates measured with (I) and without ($I_0$) sample depend linearly on the hydrogen content [4], i.e.

$$\eta = \frac{1}{\rho} \frac{(I - I_o)}{I_o} \qquad (1)$$

where $\rho$ is the density of the sample in $g/cm^3$.

The experimental arrangement for the determination of $\eta$ value is shown in Fig.1.Samples of 10 cm diameter and 10 cm height(z) were used in this experiment. Before the measurements the samples were heated during one hour at 95 °C to eliminate the moisture.

As it can be seen in Fig.2 the $\eta(z)$ function exhibits a saturation at around z=8 cm for any kind of samples (reflector). The matrix of zeolites in such experiments can be well approximated by a mixture of $SiO_2$ and $Al_2O_3$. It was found that the $\eta$ values as a function of sample thickness are the same for $SiO_2$ and $Al_2O_3$ within the limits of errors. As an example the $\eta(z)$ function for $Al_2O_3$ and sand is indicated in Fig.2.The results show that the saturation value of $\eta$ is higher by a factor of 2 for a typical zeolite sample (from Palmarito) than for the matrix. The calibration line $\eta(S + H_2O)$ vs. $H_2O$ wt% shown in Fig.2 has been determined by adding well known amount of water to the mixture of $SiO_2$ and $Al_2O_3$ powder. At zero water content we have the $\eta_M$ value for the matrix. The relation between $\eta$ and the water content for the experimental arrangement shown in Fig.1 is the following

$$\eta = 0.02529 \times \omega\% + 0.2592 \qquad (2)$$

As it can be seen in Table 1 the average atomic number ($\bar{z}$) of different zeolite matrices are very close to each other and to the values refer to $SiO_2$ and $Al_2O_3$. For $CaCO_3$ and $Na_2CO_3$ the $\bar{z}$ data are 12.57 and 9.075, respectively and from measurements a value of

$$\Delta\eta/\Delta\bar{z} = 5\times$$

was found which result in a ratio of

$$\Delta\omega\% \ H_2O \ / \ \Delta\bar{z} = 0.2$$

In spite of this result it should be noted that the accuracy of the determination of the water content is limited by the matrix effect.

Therefore, systematic measurements would be necessary to determine the dependence of $\eta$ on the average atomic number ( $\bar{z}$ ) of different zeolite matrices. The $\bar{z}$ values of $CaCO_3$ and $Na_2CO_3$ are the two extremes which can be expected for zeolites.Zeolite samples of about 800g in powder form were placed in the measuring vessel. The physical density of the samples was around 1 g/cm$^3$.In the experimental arrangement shown in Fig.1 about $2\times10^5$ counts were detected during $10^3$ s

measuring time.

## RESULTS AND DISCUSSION

Measurements show that the thermal neutron reflection method can be successfully used for the determination of water content in zeolites.

An accuracy of about 1% can be achieved within 10 minutes measuring time at a concentration of 10 w% $H_2O$. Water contents of zeolite samples collected from different site places in Cuba are summarized in Table 1. For comparison data measured by gravimetry and transmision methods are also given.[5]

The difference between the results obtained by reflection, gravimetry and transmision can be explaned if we take in account that in the last two methods, the zeolites were heated up to 450 °C during 4 hours. In this case the zeolites can loss not only structural water but also other associated elements weak bounded. However in the reflection method the zeolites were heated only up to 95 °C during two hours to eliminate the moisture, by this way we avoided the undesirable effects. The relative low value for sample from Tasajera is related to the presence of about 20% clay mineral in addition to zeolite. The neutron reflection method renders it possible the in-situ selection of the samples during exploitation.

Water content at around 10 w% can be determined with a

relative accuracy of about 1% for the same matrix choosing 10 minutes measuring time.The spread in the concentration of zeolite in the mineral exploited depends on the lower limit of water content chosen for selection. Further measurements are needed to determine the spread in the water content for samples collected from the same site places.

In addition, there are possibility of the proton presence in zeolites structure which may affect the accuracy of the present results. For this reason are required additional researches to evaluate the proton contribution.

ACKNOWLEDGEMENT

The authors are indebted to Mr. R. Lopez Mendez, Head of the Neutron Generator Laboratory for the excellent working conditions.

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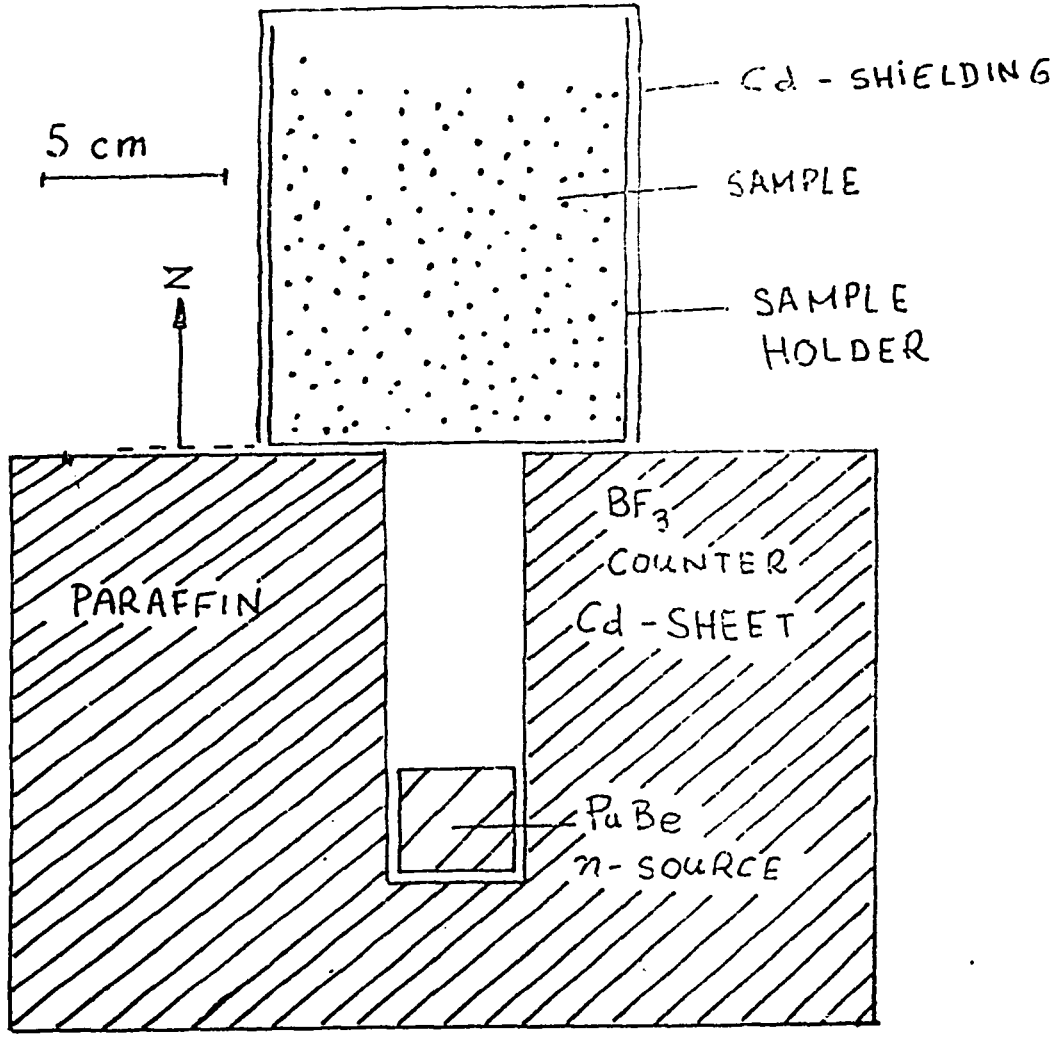
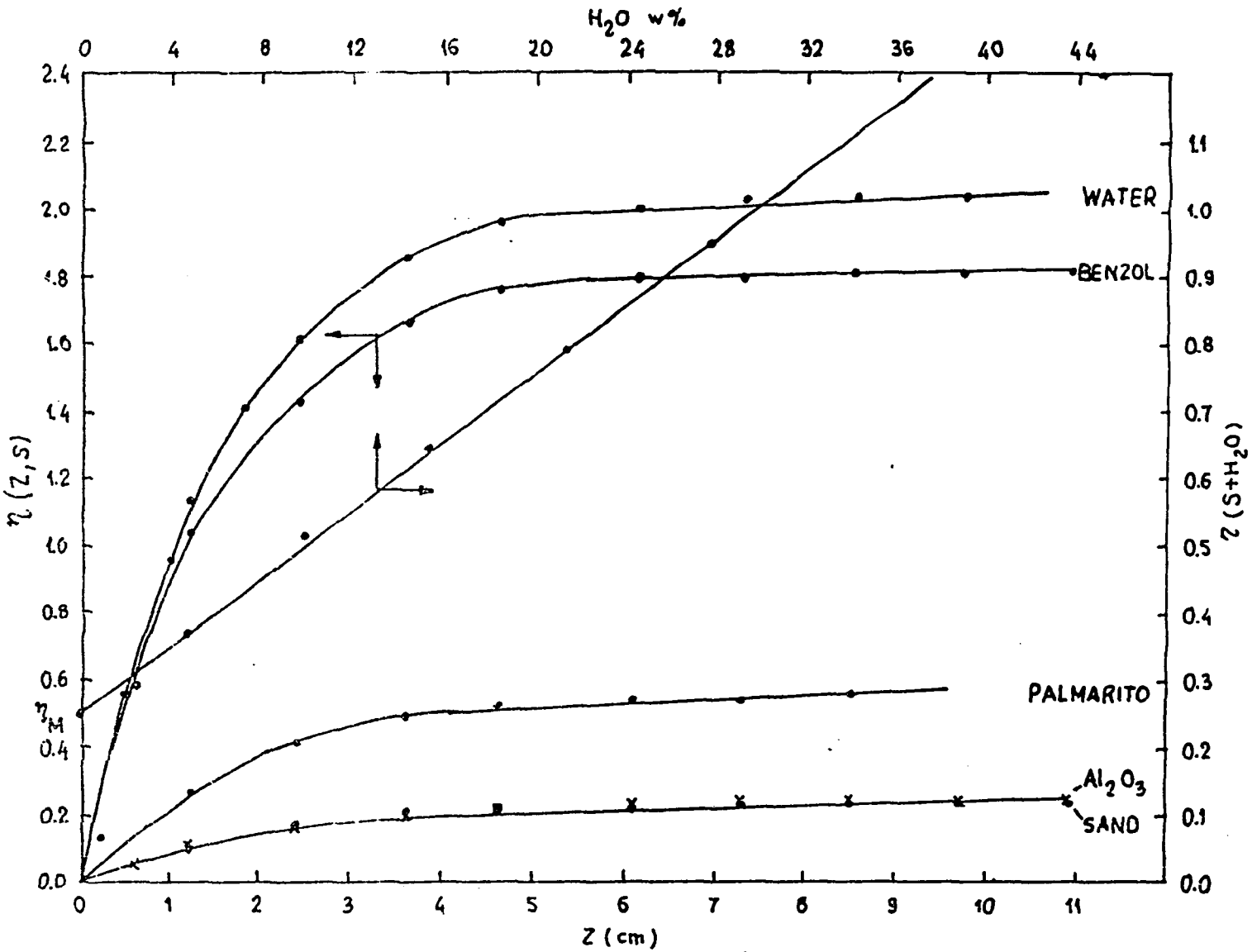
TABLA1. Some characteristic data of zeolites.

| Site place | Measured $H_2O$ in w% | Gravimetry | Trasmision |
|---|---|---|---|
| Palmarito | 12.17 | 14.12 | 13.83 |
| Orosco | 15.16 | 17.82 | 18.52 |
| Piojillo | 13.51 | 14.23 | 14.08 |
| Tasajera | 10.75 | - | - |


5 cm
Z
Cd - SHIELDING
SAMPLE
SAMPLE HOLDER
PARAFFIN
$BF_3$ COUNTER
Cd - SHEET
PuBe n-SOURCE


$H_2O$ w%
0
4
8
12
16
20
24
28
32
36
40
44
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
$\eta$ (Z,S)
$\eta_M$
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
Z (S+$H_2O$)
WATER
BENZOL
PALMARITO
$Al_2O_3$
SAND
0
1
2
3
4
5
6
7
8
9
10
11
Z (cm)

Impreso en el Centro de Información de la Energía Nuclear.
La Habana, Cuba, diciembre 1989.