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Elaboration And Characterization of Zeolite Membranes Type Na-A on Supports Plans with Clay: Application for The Removel of Heavy Metals.

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Abstract: *In this work, the characteristics of NaA zeolite membranes for Cr^{6+}, Zn^{2+}, Cd^{2+}, Co^{2+} and Pb^{2+} metals removal from synthesized solutions are presented. These Membranes were elaborated on clay (region of Meknas) which were prepared and characterized by XRD, SEM and N_2 permeation. Then we study the effects of synthesis parameters such as temperature T and time on the proprieties of the zeolite membranes. The best conditions for prepared membranes are obtained for T= 80°C, t= 24 h.*
The tests for the removel of heavy metals in particular: Cr^{6+}, Zn^{2+}, Cd^{2+}, Co^{2+} and Pb^{2+} on these NaA zeolite membranes, show that Cr(VI) is around 25%, while it reaches 98% for the other metals after 30 min of filtration.

Keywords: *NaA zeolite, clay, membrane zeolite, heavy metals, filtration.*

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I. Introduction

In the last few decades, widely progress has been witnessed on zeolite membranes, for their applications in catalysis and separation method, zeolite membranes, especially NaA type have been intensively utilized in wide range of applications due to their uniform pore and thermal stability. [1-5] The goal of this work is to test the performance of NaA zeolite membranes for the filtration of heavy metals such as, Cr^{6+}, Zn^{2+}, Cd^{2+}, Co^{2+} and Pb^{2+} these are considered the toxic elements. For the clay mineral used we controlled the porosity by organic additives such as carbon…….

Experimental Section

a) Elaboration of supports

Three planes supports (4 cm in diameter and 2 mm in thickness), based on clay collected from the same site (in the region of Meknes in Morocco), were elaborated by using activated carbon have named (AC) and starch (AA) as porosity agents and without additive called (AS).

b) Characterization of supports

The supports was examined using x-ray diffraction XRD for the clay powder used in the synthesis (cukα radiation: λ= 1.5406 A°) and scanning electron microscopy SEM and tested by a filtration pilot then we study their porosity.

II. Study Of The Porosity For Supports.

The principle of this study is to measure the percentage of the porosity for the supports. Indeed, This is done by measuring the mass of the supports dry and soaked in water during 24 hours with 25°C.

$$P_{water} = \left(\frac{M1 - M0}{M0}\right) * 100$$ [6]

With:

P_{water}: porosity in percentage (%);
M_0 : The dry mass of the sample in gram (g);
M_1 : The mass of the sample after 24 hours in water in grams (g).

c) Synthesis of NaA membrane.

The synthesis mixture was prepared by hydrothermal process, hydrogel solution (synthesis gel) was prepared by agitation of silica source and alumina source in a polyethylene flask containing water and sodium hydroxide. Then the support was placed horizontally and bottle was covered.

The syntheses were performed at different temperature (T=40, 60 and 80°C) for 24h. After each synthesis, the membranes were washed several times with deionized water and dried at 100 °C for 4h.

d) Characterization of membrane.

X-ray powder diffraction data were collected with diffractometer shimadzu 6100 using Cukα radiation and a monochromator . The range (5-60°) was scanned with time of 3s/step and with SEM.

e) Filtration of metals.

The filtration system used is show in figure 1. The pilot of filtration consists of a closed circuit of a circulating pump and a membrane module. The pressure applied is ensured thanks to the nitrogen bottle directly related to the water tank.

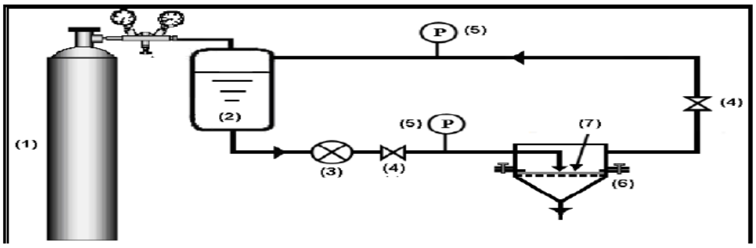


Fig 1: Diagram of the pilot of filtration: (1) nitrogenBottle (2)Water tank (3)circulating pump(4) valves(5) manometers (6)Modulates membrane (7) membrane.

The experiments were obtained by prepared different water solution of $pbcl_2$ and $zncl_2$ and cro_3………….the concentrations were determined by atomic absoption spectroscopy.

For ionic solutions, rejection factors were obtained as:

$$R = \frac{C_f - C_p}{C_f} \times 100 \quad [7]$$

Where c_f and c_p ions concentrations in the feed and permeate solutions, respectively.

III. Results And Discussion.

a. Filtration tests and porosity study for the supports.

After the development of the pilot we determined flow through the supports (AS), (AA) and (AC) by using tap water. (The pressure of circulation of the liquid is of 0.8 bar).

- **Of the support (AS) clay without additive .**

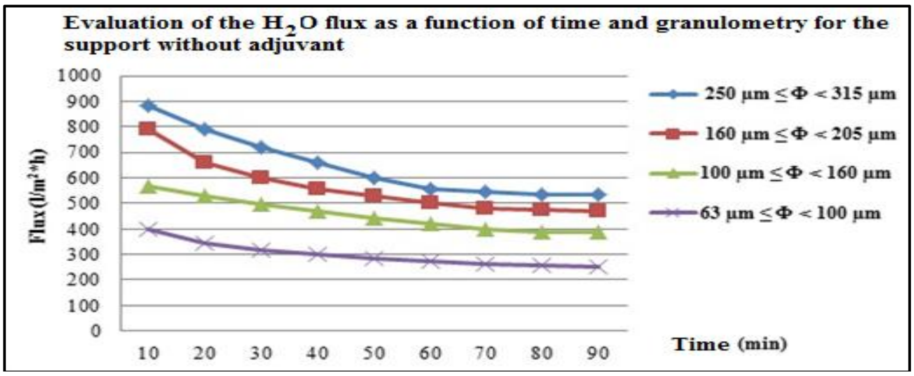


Fig 2: Flow through the support (AS) with different granulometry

Fig. 3 shows the flux through the (AS) supports of various particle sizes, it being noted that the flux decreases as a function of the size of the grains. This phenomen can be explained by the grains having small diameters sintered better than the grains having higher diameters and unlike diameter of the pores obtained during sintering of the supports (pore diameter increases with grain size).

Tab 1: % of porosity for (AS) support.

The granulometry(Φ)	Porosity %	Flux after min (l/m²*h)	Flux after 90 min (l/m²*h)
250 µm ≤ Φ < 315 µm	10.37	881,582	534,652
160 µm ≤ Φ < 250 µm	12.68	793,423	466,720
100 µm ≤ Φ < 160 µm	14.12	566,731	385,157
63 µm ≤ Φ <100 µm	16.81	396,712	252,683

- **Of the support (AC) clay with activated carbon and (AA) clay plus starch**

In this study, we opted for the powder-based support whose particle size: (63 µm ≤ Φ<100 µm) for the development of the supports (AC) and (AA).

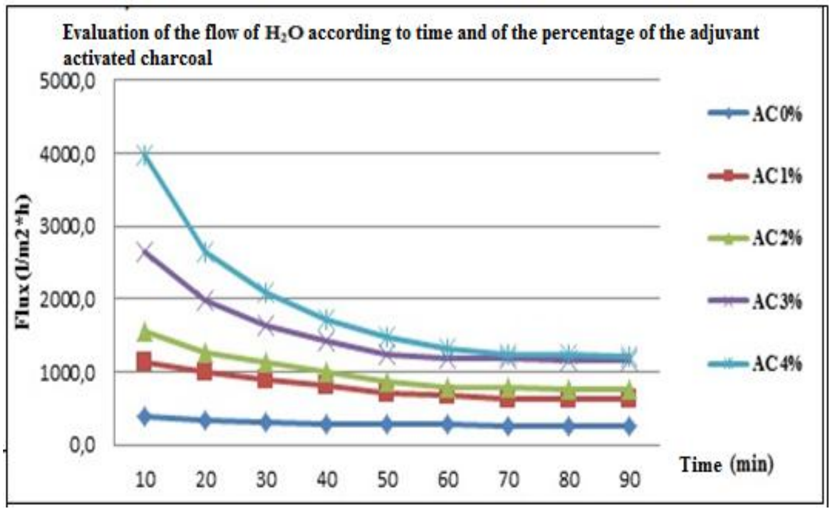


Fig3: Flow through the various supports AC.

Different (AC) supports are developed by varying % of activated carbon (2%, 3% and 4%). Figure 3 shows the flux through in the different AC supports.

We notice:

- ✓ Flow through the supports decrease according to time.
- ✓ Flow increases with the percentage of the additive.

What can be explained by the increase in porosity with the percentage of the additive during sintering .

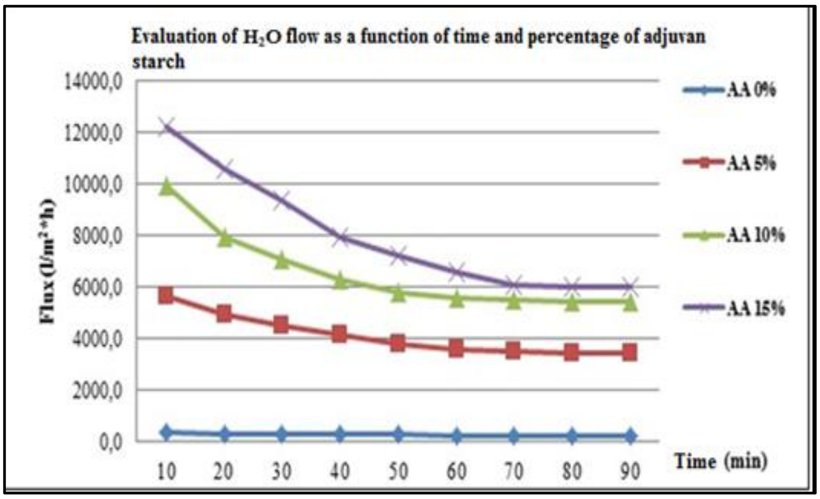


Fig4: Flow through the various supports (AA).

N.B: one note that flow through supports (AA) is more important than that of the supports (AC) that can be explained by the increase in the porosity of the supports obtained with the auxiliary starch during sintering.

Tab 2: % of porosity for the (AC) and (AA) supports.

Supports AC	Porosity %	Supports AA	Porosity %
AC 0%	16.81	AA 0%	16.81
AC 2%	19.79	AA 5%	23.28
AC 3%	23.86	AA 10%	33.60
AC 4%	26.07	AA 15%	42.45

Table 2 shows the percent porosity values for the AC and AA supports. It is noted that, for both types of support, the porosity increases with the increase in the percentage of the adjuvant added.

N.B: In the rest of the work, we will use the supports AA 10% and AC 3% for the membrane deposition. This choice is justified by the fact that with these percentages there are supports of different porosities which do not break during their uses in filtration.

a) **b)**

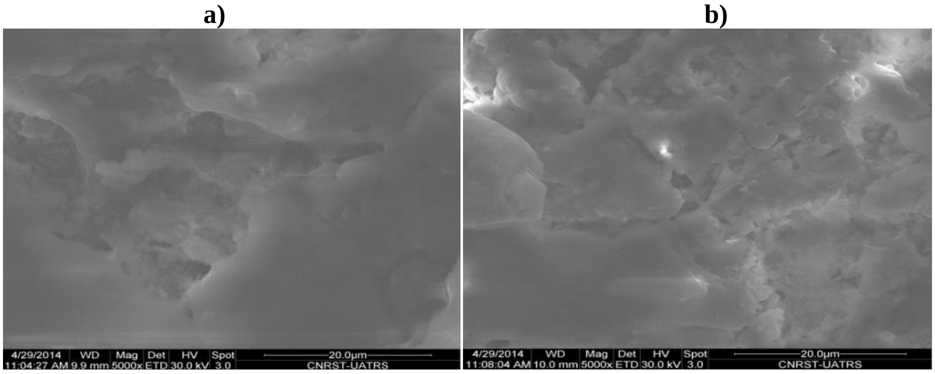
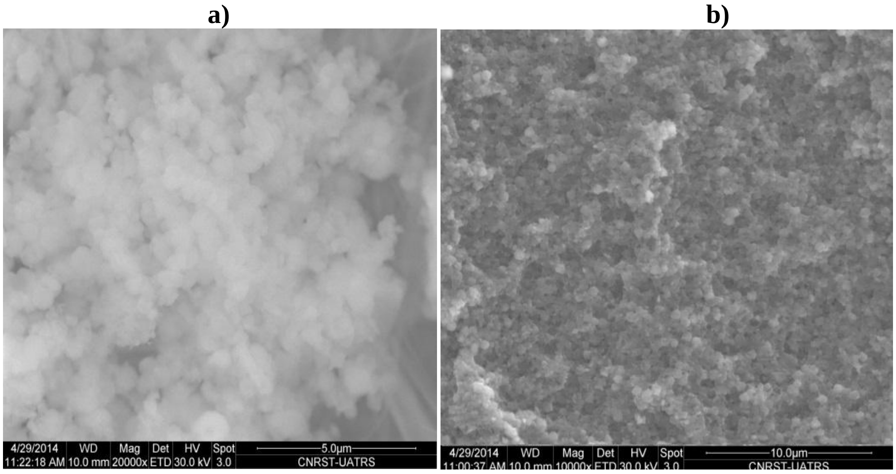


Fig 6: Micrographs of the support (a) AA 10% and (b) AC 3%

b . Scaning electronic microscopy SEM of zeolitic membrane.

a) **b)**



c)

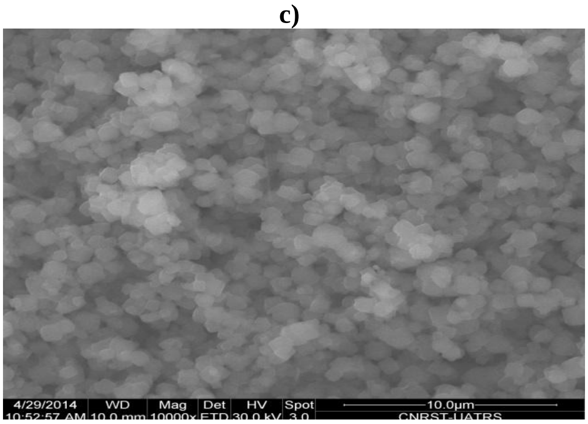


Fig 7: Images of zeolithic membrane.
(A) membrane with 40°C/24h;(B)membrane with 60°C/24h;(C)membrane with 80°C/24h.

Figure 7 shows the SEM micrographs of the prepared NaA at different temperatures. For T=40°c and T=60°c the support is covered by a discontinuous NaA zeolite with no clear crystalline but a synthesis at 80 °c , the typical cubic morphology of zeolite NaA and almost covers the support surface . So the best NaA zeolite membrane was obtained for a synthesis temperature of 80 °c.

c. X-ray diffraction (DRX).

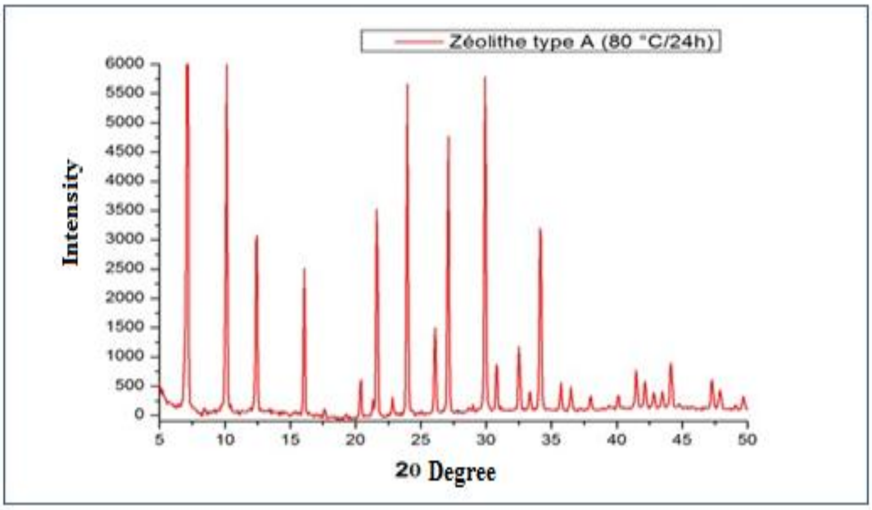


Fig 8: XRD of zeolitic type A (T = 80 ° C / 24h)

The figure 8 represented the X-rays of the synthesized zeolite type A. The decomposition of the spectrum and the indexing indicated that the structure is cubic. This result is confirmed by the morphology of the particles obtained by the SEM.

d. The flux through the membranes as a function of the pressure of N_2:

The study by SEM of the membrane deposited on clay support indicated that a continuous membrane is formed on the support, but they not provide information on the permeation performances of the membrane. So to say the quality of the zeolite membranes is to perform gas permeation.

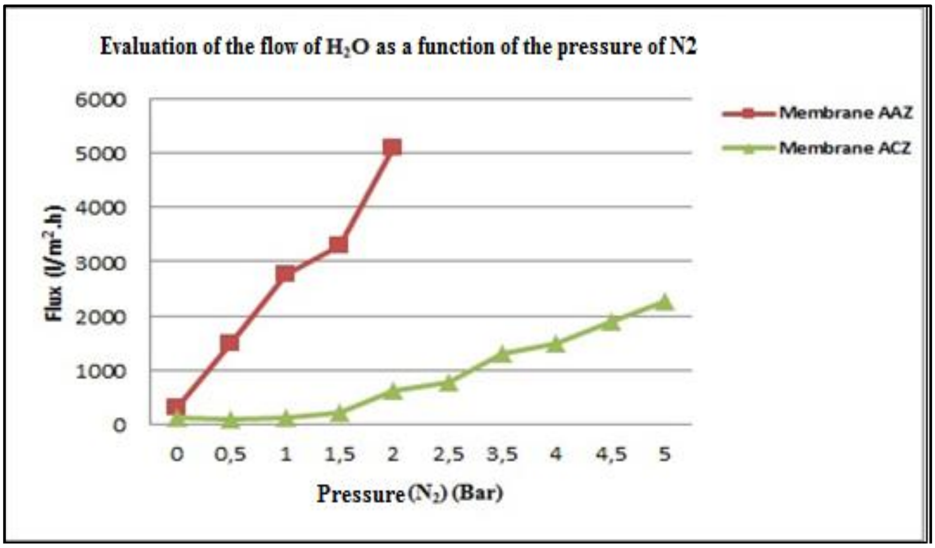


Figure 9: Flow through membranes ACZ and AAZ

Figure 9 indicate that the flow through for the zeolithe membrane (AAZ) is more important than the membrane (ACZ) by using ultra pure water of a resistivity of 18 MΩ.

N.B: In the rest of the experiment, we chose membrane ACZ-80 to carry out the tests of the retention of heavy metals.

Table 3: Pore diameter and flux through AAZ and ACZ membranes.

Membranes	Ø of pores	Flux obtained at 0,5 bars in L/m^2.h
AAZ	520 nm	1514
ACZ	420 nm	81

E . Filtration tests of metals:

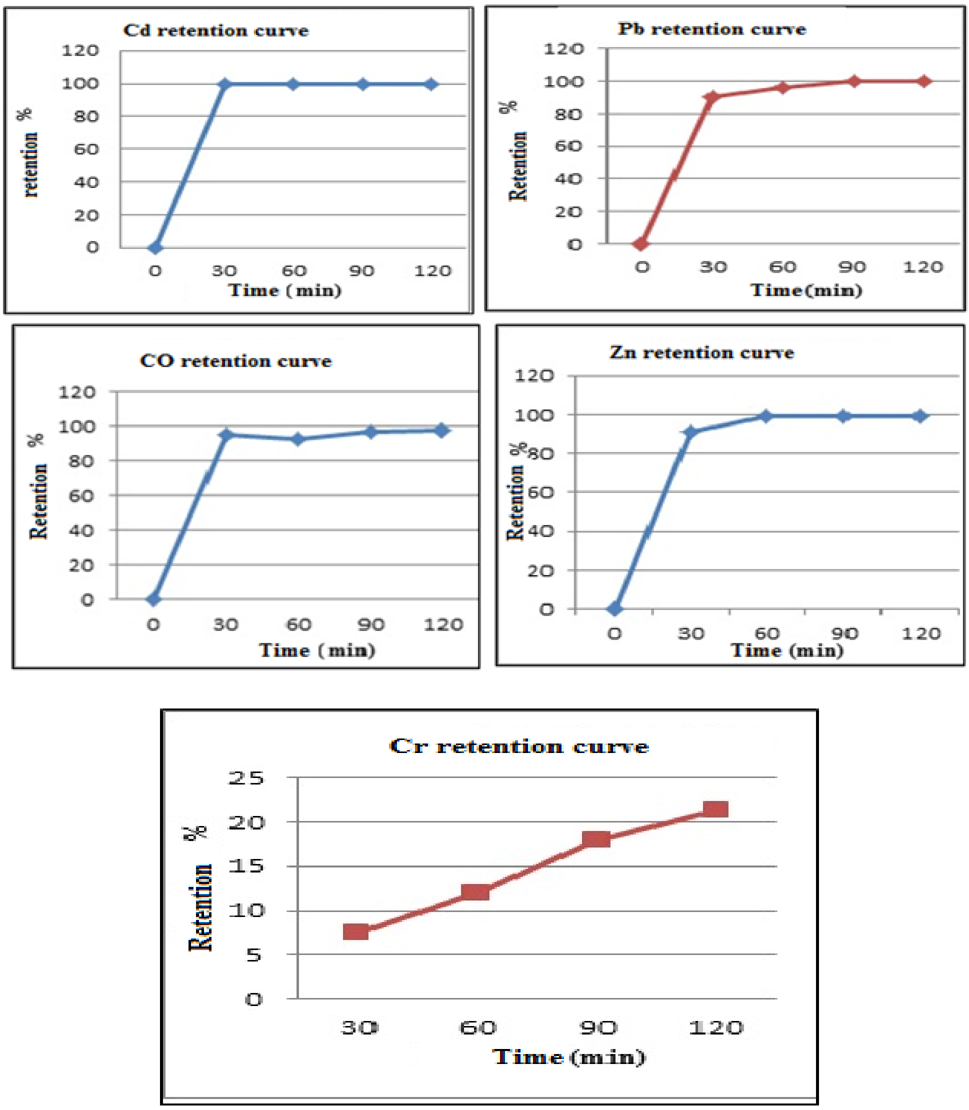


Figure 10 : Metal retention curves on ACZ-80 membrane.

Figure 10 represented the retention of some heavy metallic cations by the ACZ-80 membrane as a function of time , shows that Zn(II), Pb (II), Co(II), Cd (II) rejection increased to 98 % after 30 min , while the rejection of chromates it is 25 %, because the surface of the NaA zeolite membrane charged positively, for Zn(II), Pb (II), Co(II), Cd (II) of their positive charge , will be repulsed by the membrane a fact which explains their rejection about 98 % after 30 min of filtration , Therfore, the chromates species which are charged negatively reject 25% of the Cr(VI). The increase in the retention rate of ion Cr (VI) during filtration can be explained by partiel filling at the surface or inside the pores of the membrane

IV. Conclusion

In this study, we synthesized NaA zeolite membranes on a low cost seeded clay obtaind from Méknés region (Morocco). Temperature played an important role in the performance of the membrane During this work the NaA zeolite membrane was obtained for a synthesis 80°C of 24 h . Then the result of the experiments test for the retention of the metals indicated that zeolite membrane, useful for the separation of heavy metals from solution.

References

[1]. P. Blanc, A. Larbot, M. Persin and L. Cot, Preparation of hafnia ceramic membranes for ultrafiltration, J. Membr. Sci., 134 (1997) 109.

[2]. R.J. Baker, A.G. Fane, C.J.D. Fell and B.H. Yod, Desalination, 53 (1985) 4.

[3]. A.Bouisfi (1997) : Elaboration et caractérisation des supports à base d'argile pour membranes de microfiltration et d'ultrafiltration, Diplôme des études supérieures, Université Sidi Mohammed ben Abdellah, Facultés des Sciences Dhar el Mehraz Fès, Maroc, 103pp.

[4]. S. Iaich, D. Lahmami, L. Messaoudi, A.Chahlaoui et N.Tijani :ScienceLib Editions Mersenne : Volume 5 , N ° 130513, ISSN 2111-4706.

[5]. Y. Anbri, N. Tijani, J. Coronas, E. Mateo, M. Menéndez, J. Bentama, Desalination, 2008, 221, 419 – 424

[6]. N. Tijani , H. Ahlafi, M.Smaihi and A. El Mansouri " Preparation and Characterization of Na-A Zeolite Membranes and its Application for Removal of Heavy Metals" Mediterranean Journal of Chemistry 2012, 2(3), 484-492

[7]. N. Tijani , H. Ahlafi, M.Smaihi and A. El Mansouri " Preparation and Characterization of Na-A Zeolite Membranes and its Application for Removal of Heavy Metals" Mediterranean Journal of Chemistry 2012, 2(3), 484-492

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