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Comparative Studies Between Hydrophobic Catalysts Prepared for Isotopic Exchange Process

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The process for hydrogen isotopes separation is important for nuclear technology. Thus, the preoccupation is to find more efficient catalysts with a long operating time. The paper presents comparative studies between the physical and structural properties of hydrophobic catalysts. For the study of the isotope exchange water-hydrogen there were prepared the Pt/C/PTFE and Pt/SDB catalysts. These catalysts were manufactured by impregnation. For all the types of catalysts prepared there were determined the metal contain (active catalytically compound). The physical and texture properties such as specific surface, pore volume and its distribution after pore radius were also determined. The properties were determined by isotherm absorption to liquid nitrogen temperature 77.8 K through penetration with Hg. The specific surface was calculated according to BET equation; micro pore volume and its distribution after the range of radius were calculated using the approximation of cylindrical pore according to mathematical model Barret-Joyner-Haleda and nitrogen absorption data. One of the most important aspects for the utilization of hydrophobic catalyst in such processes is the stability radiation and impurities. Model catalysts are important because they offer a better prospect of controlling variables such as surface topography and composition. Using such a model catalysts it is better controlled the contamination of the surface structure.

Key Words: Catalysts, Hydrophobic, Isotope, Structure, Properties, Pore.

INTRODUCTION

Metals are used as catalysts for many technically important processes that the need to understand their structure as fully as possible hardly requires emphasis. Model catalysts are important because they offer a better prospect of controlling variables such as surface topography and composition than do normal technical catalysts[1].

Even when a catalyst support is viewed in a passive role, it is clear that its structure and surface chemistry are likely to be important in controlling the ultimate performance of a supported catalyst[2]. For instance, a number of factors including the pore structure, surface area and the nature of the surface influence the degree to which a metal is dispersed when it is introduced on to the support, while the pore structure will influence the accessibility of the supported metal to the reactant. However, a material which is used as a support for the metallic phase may have catalytic activity[3].

EXPERIMENTAL

In this paper, a comparative studies between the physical and structural properties of hydrophobic catalysts are presented. For the study of the isotope exchange water-hydrogen, there were prepared the Pt/C/PTFE. These catalysts were manufactured by impregnation.

The following steps are required to produce the Pt/C/PTFE catalyst: (a) active charcoal with a specific surface about 1100 m^2/g, (b) hexacloroplatinic acid, (c) polytetrafluoroethylene (PTFE) powder with a low granulation, (d) acetone, chemical reactive for analysis.

The samples were realized in small batches, in function of the necessary amount for physical and structural characterization[4].

Active surface area dispersion-metal surface-Pt, on C/PTFE support, the dimension of the Pt crystallite and the metal dispersion on the surface, all these were carried out through chemisorptions studies for H_2[5].

RESULTS AND DISCUSSION

The experimental data obtained from the interpretation of the chemisorption isotherms are given in the Table-1.

TABLE-1

Sample	%Pt	V_{H2ads}	H/Pt (at.H/at.Pt)	S_{Pt} (m^2Pt/gPt)	d_{Pt} (Å)
Cat. Pt/C/PTFE - 10 min	1.105	0.07	0.11	30.14	78
Cat. Pt/C/PTFE - 20 min	1.105	0.06	0.096	26.30	89
Cat. Pt/C/PTFE - 30 min	1.105	0.03	0.048	13.15	178

The analysis shows that the increase of the heating time is accompanied by a decrease of the dispersion parameters. The results shows, the chemisorptions H_2 volume decrease from 0.07 cm^3 for a heating time of 10 min to 0.03 cm^3/g cat., for a time of 0.5 h that means a decrease with 56.37 %.

The metal inserted in catalyst don't finely spread at the surface of the catalyst, the platinum dispersion decreasing with the increasing of heating time from 0.11 to 0.048. The Pt crystallites dimension increase from 78 Å for a heating time of 10 min to 178 Å for a heating of 0.5 h. These values indicates the existence of some metal concentration, pressed structures with different thickness which with the increasing of heating time can migrate in the support structure in inaccessible places for hydrogen[6].

To realize the studies using optical microscopy, the samples were photographed without any processing, in natural light, through reflexion on generatrix with a total enlargement about 23 times.

Sample 1 (10', stamp 1), Fig. 1: On a white radiant teflon support with grey shadows submicronic it make obvious black deposes, the great majority being 20 µ, but there are also *ca.* 1/3 with dimensions till 60 µ and rare random concentration in bunch form, about 100-200 m. The sponge support don't permits more enlargements.

Sample 2 (20', stamp 1), Fig. 2: The comparison with sample 1, the grey shadows submicronic include a big part from background, where it makes obvious points-like black deposes about 20-30 µ, random concentration about 50-60 µ and bunch or zigzag concentrations about 75-120 µ.

Sample 3 (30', stamp 1), Fig. 3: On a dark grey background it makes obvious light coloured islands with 150-200 µ. There can be observe black points-like deposes of 20-30 µ, random concentration of 40-50 µ and 100-120 µ concentrations.

Sample 4 (10', stamp 2), Fig. 4: On a white teflon support with some grey shadows it makes obvious black points-like deposes relatively often below 20 µ, zigzag deposes of 50-60 µ and random deposes of 100-150 µ.

Sample 5 (20', stamp 2), Fig. 5: On a grey background it makes obvious black points-like deposes of 30-40 µ, random deposes of 50-60 µ and black islands of 300-350 µ.

Sample 6 (30', stamp 3), Fig. 6: On a dark grey background it makes obvious rare light coloured islands with the diameter 100-200 µ where it can be observe black points-like deposes of 20-30 µ, random concentrations in bunch or zigzag form of 50-60 µ, big black deposes of 100-150 µ.

The physical and structural properties such as specific surface, pore volume and its distribution after the radius size of the pores were determined through adsorption isotherm for N_2 at 77.8 K and through penetration with Hg[7]. The data are presented in Table-2.

TABLE-2
TEXTURE PARAMETERS DETERMINED FROM ADSORPTION ISOTHERM FOR N_2 AT 77.8 K

Sample determination		M_1 10'	M_1 20'	M_1 30'	M_2 10'	M_2 20'	M_2 30'
Pores volume (R: 5-300 Å) cm^3/g		0.0127	0.0128	0.0172	0.0154	0.0087	0.0114
Distribution %	5-10	40.94	30.06	19.01	6.19	4.48	5.77
Pores vol	10-15	21.81	18.20	41.42	32.82	25.95	50.02
After	15-25	12.58	8.58	12.90	25.86	29.32	1.80
Radius	25-50	8.51	17.33	17.11	23.86	33.64	31.26
R (Å) =	50-100	8.98	14.96	5.79	6.89	4.41	6.69
	100-300	7.18	10.87	3.77	4.38	2.20	4.46
Specific surface (m^2/g)		16.81	14.72	15.88	13.84	5.84	7.50

Figure 1

Figure 2

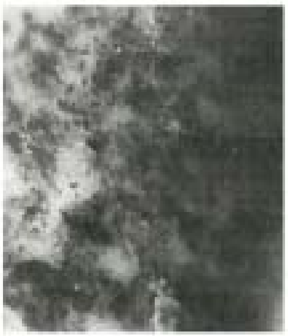

Figure 3

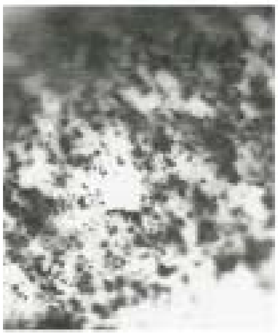

Figure 4

Figure 5

Figure 6

Specific surface was calculated with B.E.T. equation, pores volume and its distribution after the radius size were calculated in approximation of cylindrical pores, after the mathematical model Barret-Joyner-Halenda, using N_2 adsorption data[8].

The experimental data analysis shows that: (a) the catalysts obtained have the biggest porosity; the pores volume being of 0.2046 cm^3/g, for a heating time of 10 min; (b) the pores distribution after size is monomodal, with a single maximum, 63 % being in the radius range of 10^3-10^4 Å; (c) If we increase the heating time from 10 to 20 min, results the decrease at half of pores volume and changes in pores distribution after size. It also appears as a bimodal distribution of the porosity, with maximum between 10^3-10^4 Å and 10^5-2×10^5 Å; (d) the increasing of the heating time from 20 to 30 min don't significantly affect the porosity; (e) the medium radius of the pores decrease with the increasing of the heating time from 243 Å to 121 Å; (f) the pores distribution after radius is polyspreaded, with maximum between 5-50 Å (~ 18 %), 300-10000 Å (36-15 %) and 10^5-2×10^5 Å (20-30 %). The pores weight with radius of 2×10^5 Å, increases at once with heating time, while those with radius pores of 10^3-10^4 Å decreases from 18 % to -0.34 %; (g) medium radius of the pores increases from 103 to 153 Å.

The hydrogen isotopes separation represents a very important issue for nuclear energy[9]. So that, a major preoccupation is to find more efficient catalyst with a longer period of utilization. That's way the catalyst methods preparation is important because it is a check point for chemical composition and physical structure of the catalyst.

Regarding the study of the adequate catalyst used in the water-hydrogen isotopic exchange process there were prepared the catalysts PTFE and Pt/SDB[10].

These catalysts were prepared through impregnation and taken into account all data available until now. For all classes of catalysts prepared we determined the metal content (the active catalytic component). Physical and textural properties such as specific surface, pores volume and their distribution after the size of the pores radius were determined using absorption isotherm for nitrogen at 77.8 K and through penetration with Hg.

Specific surface was calculated using B.E.T., micro pores volume and its distribution after the size were calculated with the approximation of cylindrical pores using the mathematical model Barret-Joyner-Halenda and adsorption nitrogen data.

The next step it will be to test the catalysts in the isotopic exchange device from the experimental pilot plant from Rm.Valcea, Romania[11].

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