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ORIGINAL ARTICLE

INCAPSULATION OF Fe2O3 NANO-PARTICLES IN A MODIFIED FAUJA-SITE Y-ZEOLITES

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Abstract:

The solid zeolites are modified to increase their usefulness under the different reaction conditions. The Different metals incorporate in to the zeolite gives different catalytic activities of the samples prepared hence are useful in carry out different catalytic reactions depending upon the Catalytic active metal incorporated in to the zeolite. The synthesized and highly crystalline form of Na-Y sample is used for further modification. As it is the largest zeolite in use in petrochemical industries and for different purpose in the word even today. The synthesized Na-Y is ion exchanged to converted in to NH4-Y by reflux method using Ammonium Nitrate and calcined at 5000C,The process were repeated three times to convert it in to highly crystalline low silica H-Y zeolite. The sample H-Y zeolite was treated carefully under controlled hydrothermal conditions at 550OC, at 700 OC \and 850 OC in succession one after the other by using previous sample as the base sample for each of the next samples After cooling at each temperature interval the above procedure of ion exchange and calcinations is repeated thrice for each of the sample to obtain the final product. This high silica H-USY sample thus prepared is used for encapsulation of nano particle Fe in oxidized form in stable position of host Zeolite (H-USY as zeolites pores are of nano size). and modified to Fe-USY products. These samples are characterized by XRD, ESR, UV-VISIBLE, AAS, XRF techniques etc. The confirmation of incorporation of Fe in the sample at stable position and in the other environments, ESR studies are discussed in the present paper.

KEYWORDS:

(1) Fe -source(2)controlled Steam chamber(3) Faujasite Na-Y zeolite.

INTRODUCTION

What are Zeolites?

Zeolites are crystalline hydrated alumino- silicates having rigid three dimensional infinitely extended framework structure. It encloses the cavities and channels of molecular dimensions. The framework structure contains corner sharing of [SiO4] -4 and [AlO4] -5 Tetrahedral linked through common oxygen atoms as the primary building units. The general empirical crystallographic unit cell formula of zeolite is expressed as

M x/n[(AlO2)x (SiO2)y] zH2O

where

M= charge compensating cat ions of valency.

x and y represents the no. of moles of SiO2 & AlO2 where $y \geq x$ and

z= no. of water molecules.

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Most of the zeolites are very unstable and lose their catalytic activity easily under the severe conditions of catalytic reactions and hydrothermal conditions at high temperatures. This is due to large dealumination of zeolite framework which leads to collapse of zeolite structure. To overcome this problem synthesized zeolites are dealuminated carefully in a very controlled manner under hydrothermal conditions Which is found to be a best method for increasing catalytic activity as well as stability of the zeolite. The extra lattice aluminum species formed are removed by acid leaching. Abundant literature is available on Y type zeolite as it is most promising zeolite due to the 3D pore structure. it is best suited to produce large numbers of petrochemicals. Such zeolites are treated under steam at different temperatures and are called as H ultra stable Y zeolite. The zeolite fujasite type-Y is low silica,large3D pore zeolite, which can be easily synthesized in the laboratory and in the industry.The unit cell of Y type zeolite is cubic with a large dimension of 250A and contains 192(Si, Al) O4 tetrahedral, has remarkably stable and rigid framework structure with largest void space, which amounts to be nearly40% by volume of the dehydrated crystal.

Experimental:

10g of Na-Y was added in to 200 ml 2N NH4NO3 and ion exchanged for Preparation of NH4-Y zeolite from it. The Solution was taken in a round bottom flask. This mixture was heated at 1000C by stirring it for 12h by using reflux method. The sample was then filtered, washed several times with hot de-ionized water till free from any traces of nitrate ions and dried at 1000C over night, the above procedure was repeated at list for three times to obtain more than90% of NH4-Y from Na-Y by above method of 2N NH4NO3 ion exchange.As Sodium was added to the zeolite the total no. of acid sites was reduced. Effect of sodium poisoning was found to be dramatic over the entire range of Si/Al ratio examined.one sodium atom could effectively poison 5 non frame work acidic Al atoms which found to reduce the catalytic activity of the zeolite. In view of this report Na-Y was converted into NH4-Y zeolite and used for Fe incorporation in it. Still It was used in the Synthesis of zeolite, because it can be easily ion exchanged with any metal.

Preparation of high silica Fe-Y zeolites by using NH4-Y and (NH4)3Fe(C2O4)3.3H2O in multy step (MSTP)method:

Series of samples identified as MSTP-X-Y.where X stands for temperature of preparation and Y stands for mole fraction of (NH4)3Fe(C2O4)3.3H2O per mole of Al used in preparing them.These samples were prepared one after the other by the following procedure.

2) Preparation of high silica USY zeolites;

Most of the zeolites are very unstable and lose their catalytic activity easily under the severe conditions of catalytic reactions and in hydrothermal conditions. Due to large dealumination of zeolite framework which leads to collapse of zeolite structure to overcome this problem synthesized zeolites are dealuminated in controlled manner and extra lattice aluminum species formed were removed by acid leaching .The above prepared NH4 Y zeolite was further dealuminated under carefully controlled steam in the cylindrical reactor to increase hydrothermal severity at 550 OC for 4 h, 700 OC for 4 h, 850 OC for 4 h, in succession by using the previous sample as the mother sample for the next sample. After hydrothermal treatment at each temperature interval the samples were ion exchanged with 2N NH4-Y as per above procedure and finally sample was calcined at 500 OC to obtain H-USY zeolite the procedure is narrated in the Table-1.The above condition of the sample was standardized before introducing Fe in to the sample.

Table-1 Preparation of USY by hydrothermal dealumination of NH4 -Y

S.N.	Name of the sample	Steaming temperature	Exchange with NH_4NO_3
1	USY-550-MSTP	$550\ ^0C$	Yes
2	USY-550-700-MSTP	550^0C 700^0C	Yes Yes
3	USY-550-700-850 MSTP	550^0C 700^0C 850^0C	Yes Yes Yes

Preparation of very high silica Fe-USY from very high silica USY(550-700-850-MSTP), For incorporation of Fe in the USY zeolite, we have adopted similar procedures which lead to very high silica USY zeolite. It is a variation of multiple step method of preparation. In this method, the very high silica USY was first prepared as per sample no. 3in the table-1. It was then reacted with ammonium Iron (III) oxalate (NH4)3Fe(C2O4)3.3H2O solution with its appropriate mole fraction as shown in table-2 at 2000C for 18 hours, filtered and washed it by de-ionized hot water. It was then treated with dilute 0.01N HNO3 solution at 1000C for 4 hours, filtered , washed several times with hot de-ionized water and dried. The whole cycle was repeated for 10 times to get the final sample. In order to incorporate Fe along with Ti in USY by using K2TIO(C2O4)2 2H2O as a Ti sault and Prepation of very high silica Fe-USY from USY(550-700-850MSTP, and (NH4)2Fe(C2O4)23H2O .by taking their required mole fraction along with Ti-Sault as shown in Table-2

Table-2 Study of Changes obtained by XRD, Spectroscopy:

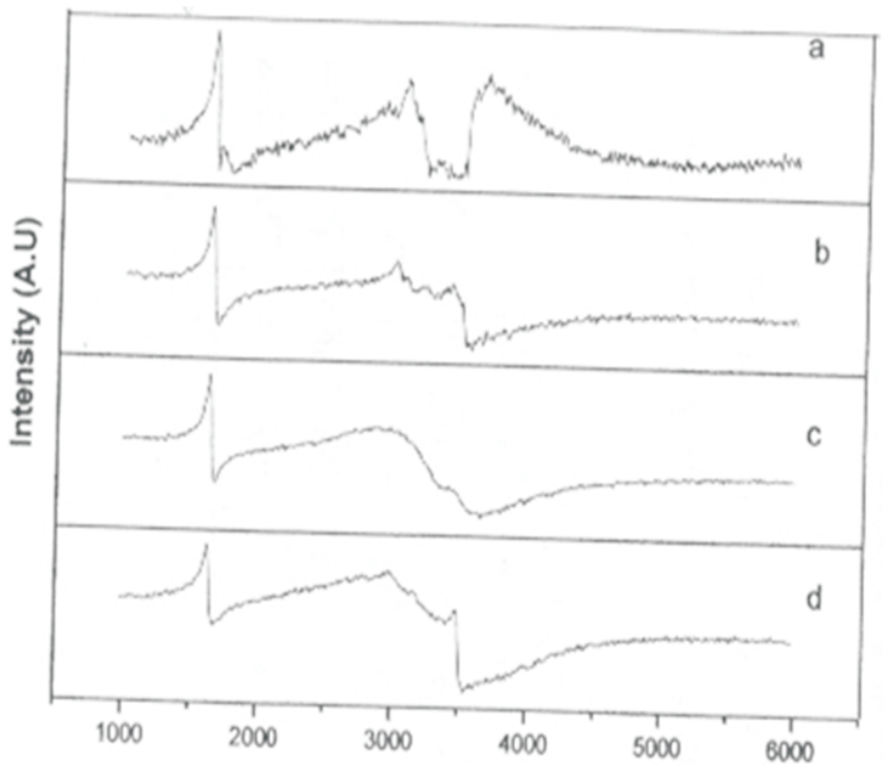
S No	Name of Sample	Unit cell const.
1	Na-Y	24.71
2	NH4Y	24.68
3	550-MSTP	24.38
4	550-700MSTP	24.34
5	550-700-850MSTP	24.21
6	550-700-850MSTP-10	24.21
7	Fe-USY	24.25

Increase in unit cell constant to 24.25 from 24.21 in 550-700-850MSTP to Fe-USY confirms the in corporation of Fe in the frame work location as the bond length of Fe-o bond(= 1.97) which is bigger than the bond length of both Al-o bond(=1.75) and Si-o bond(=1.62).

Table-3, Prepation of very high silica Fe-USY from USY(550-700-850MSTP, and (NH4)2Fe(C2O4)23H2O .

Sr. No.	Name of the sample	HNO_3 Exchange	$NH_4)_2Fe(C_2O_4)_23H_2O$ treatment	Reaction temp.
1	550-700-850-MSTP-10	0.001N	-	100^0C
2	Fe-USY	0.001N	0.05	150^0C
3	Fe –USY	0.001N	0.05	200^0C

Fig-1



g-values

ESR Spectra of acid treated very high silica Fe-USY samples, Prepared by using (NH4)3Fe(C2O4)2(Oxa).at different temp.

Result and discussion:

From the table-2 It is clear that. the unit cell constant was decreasing as the amount of dealumination increases. This is because depending upon the difference in the metal oxide bond length. As bond length of Al-O is 1.75 0A and that of Si-O bond length is very short it is 1.620A Hence there was shrinkage of unit cell constant. Because re-insertion of Si-O takes place at the position of Al-O.but it is not found to in proporation because of reinsurtation of Fe at the place of Al dealuminated from frame work location. The g value is the proportionality factor which is a function of the electron environment. It is sometimes called spectroscopic splitting factor. It was observed that the intensity of the signal at g=4.3 increases with the increase in the frame work location of Fe. The Fe3+ correspond to g=2 is assigned to the hexa- coordinated complex located at the cationic sites of the hydrated USY zeolite. The signal at g=2.3 was assigned to randomly oriented Fe3+ions with the characteristics of hydrated oxide species which does not belong to the frame work structure of the zeolite. The remaining three different weak signals present in the spectra of some of the sample may be arising due to the traces of 3 different iron species occluded in the zeolite cavity. The spectra of these species may only be responsible for broadening of the ESR spectra of the samples.

Conclusions:

1),The substitution of Fe at the frame work location of the USY Zeolites was confirmed by ESR,Spectroscopic techniques.

2) Shrinkage of unit cell constant was due to re-insertion of Si-O takes place at the position of Al-O bond of higher length.

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