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Direct synthesis of copper faujasite

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Abstract

Copper containing faujasite has been successfully prepared for the first time using a direct synthesis method. Faujasite type zeolite can be prepared in the presence of copper species by tuning the synthesis conditions. Ammonium hydroxide was used to form a copper complex that was later mixed with the reacting gel. Sodium is required to obtain copper faujasite. The complete elimination of sodium ions from the starting gel produces amorphous material. Crystallization took place at 358 K for 11 days. Crystallization temperature of 373 K produces ANA type zeolite as an impurity. Increasing by two times the amount of copper complex added to the reacting gel increases the crystallization time of Cu-FAU from 11 to 20 days (the crystallization rate decreases). The copper containing faujasite obtained was characterized by XRD, FESEM, EDX, EPR, FT-IR, TPR, and BET. According to the XRD pattern only FAU type zeolite was obtained. According to TPR experiments, the reduction temperature for Cu^{2+} ions present in Cu-FAU prepared by direct synthesis was 70 K more than for Cu-FAU prepared by ion-exchange. This difference can be due to the different location of the copper ions in the supercages or in the sodalite cages of the faujasite.

Keywords: Cu-FAU; Zeolite Y; Direct synthesis; Copper tetraammonium nitrate

1. Introduction

Copper(II) exchanged Y zeolite has been used in different applications such as heterogeneous aziridination of styrene [1], to remove organo-sulfur species from commercial diesel fuel [2], as a heterogeneous catalyst for hydrogenations and cyclopropanation [3]. The development of zeolite based catalysts for reducing NO_x to N_2 by NH_3 have received a lot of attention for several reasons like (a) high NO_x conversion, (b) wide selective catalytic reduction window, (c) sulfur tolerance above 700 K, (d) less disposal problems for spent catalysts. The most efficient catalysts are based on transition metals ion-exchanged in zeolites. The reaction is regulated by the redox cycle of the transition metal ion (Cu^{+}/Cu^{2+}) [4]. Copper exchanged zeolites have also been used for CO hydrogenation and for N_2 storage [5]. Leaching of copper ions in the reacting media is one of the most common problems reported when copper exchanged zeolites are used as heterogeneous catalysts in liquid phase systems. Copper in solution generates undesirable reaction sub-products and limits the number of times than the catalyst can be reused [1].

New methods to synthesize copper containing zeolites are useful [2]. Direct synthesis of Cu-MFI has been previously reported. Copper species were present in the precursor gel and methylamine was used as a mineralizing agent to avoid precipitation of copper oxide. The reported advantages of the direct synthesis of Cu-MFI are the homogeneity of copper distribution in the sample and the reduction of copper leaching into solution [6]. In a different study, a copper(I) complex $[Cu(NH_3)_2]^+$ was formed in situ during the synthesis of the ferrierite framework [7]. Copper nitrate, ammonia, propylamine, silica, alumina, and hydrofluoric acid were used as raw materials [7].

Cu-zeolite Y (copper containing faujasite) has never been prepared in the presence of copper species by a direct synthesis method. We report the tuning of the synthesis conditions to prepare copper containing faujasite by a direct synthesis method. Ammonium hydroxide was used to form copper complexes and to avoid the precipitation of copper oxide in the alkaline media required to prepare the zeolite. Two different

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molar ratios of Cu/Al_2O_3 were used to synthesize FAU type zeolite. The experiments to produce Cu-zeolite Y by direct synthesis method and the characterization of the product are reported here.

2. Experimental

2.1. Synthesis procedure

Two different procedures were tried to synthesize copper containing faujasite. In the first method (experiments 9–12), copper tetraammine complex was prepared in situ. In the second method (experiments F34-1–F37) crystals of copper tetraammine nitrate were prepared and mixed with the reacting gel.

Synthesis of copper(II) tetraammine nitrate: Deep blue crystals of copper(II) tetraammine nitrate are formed by addition of ethanol to a solution of copper nitrate in ammonia [8]. Copper nitrate (20 g) was completely dissolved under stirring with concentrated ammonium hydroxide. Ethanol was added to the previously prepared solution to precipitate the copper(II) tetrammine nitrate salt. The crystals were filtered and washed with ethanol.

Synthesis of copper faujasite:

(a) Experiments 9–12 (see Table 1 for batch and crystallization conditions): The materials obtained were synthesized by preparing the copper tetrammine nitrate in situ. Solution 1 was prepared by dissolving copper nitrate in ammonia. Solution 2 was prepared by dissolving sodium hydroxide and aluminate in double deionized water (DDW). Colloidal silica (Ludox LS-30) and solution 2 were added simultaneously to solution 1. The mixture obtained was stirred for 2 h and crystallized at the conditions shown in Table 1. The products were recovered by centrifugation, washed until the pH was less than 8, and dried overnight at 373 K. The procedure to synthesize copper faujasite was modified, since the products obtained were amorphous with copper oxide, gismondine, and faujasite as impurities. Cobalt, nickel, and zinc faujasite were prepared with batch composition shown in Table 1 and following the same procedure used to synthesize experiments 9–12.

(b) Experiments F34-1–F37: Copper(II) tetraammine nitrate was dissolved in water and combined with colloidal silica (Ludox LS-30). After 2 h of stirring, the gel was mixed with an aqueous solution of sodium aluminate and sodium hydroxide. The batch composition used is presented in Table 1. The gel obtained was aged for 24 h and then crystallized at 358 K for 11–20 days. The product was recovered by filtration under vacuum and washed with deionized water until $pH < 8$. The solid was dried overnight at 373 K.

Cu,Na-zeolite Y ion-exchanged was prepared by mixing 5 g of Na-zeolite Y (Aldrich) with 50 mL of a 0.5 M aqueous solution of copper nitrate and stirred for 5 min. The solid was filtered, washed, and dried.

2.2. Characterization

The XRD studies were carried out with a Scintag XDS-200 powder diffractometer on finely powdered samples using Cu Kα radiation and 45 kV and 40 mA. The step size was 0.02° (2θ) and a count time of 0.6 s with a scan rate of 2.0°/min. The XRD patterns were recorded for 2θ values between 5° and 40°.

Field emission scanning electron microscopy (FESEM) experiments for the determination of morphology were performed on a Zeiss DSM 982 Gemini field emission scanning electron microscope (FESEM) with a Schottky Emitter at an accelerating voltage of 2 kV with a beam current of about 1 μA. An ECON IV energy dispersive X-ray (EDX) analyzer Model 9800 was used to determine the silica to alumina ratio and the Cu/Al ratio of the prepared zeolites.

Fourier transform infrared spectra (FT-IR) experiments were done on a Nicolet Magna IR system 750 FT-IR spectrometer with a resolution of 4 cm^{-1}. Potassium bromide (KBr) pellets of the samples were analyzed in the range 4000–400 cm^{-1}.

The TEM measurements were performed on a JEOL 2010 FasTEM 200 kV instrument equipped with an EDAX energy dispersive spectrometer and a Gatan EELS/GIF system.

Table 1
Batch compositions and conditions used to prepare metal,Na-zeolite Y

Run #	NH_4OH/SiO_2	Na_2O/SiO_2	Metal/Al_2O_3 (metal)	Crystallization conditions		Product (XRD)
				Temperature (K)	Time (days)	
9	1.2	0.4	0.67 (Cu)	373	6	GIS + amorphous
10	1.2	0.2	0.67 (Cu)	373	6	CuO + amorphous
11	0.6	0.4	0.67 (Cu)	373	6	FAU + ANA + amorphous
12	0.6	0.2	0.67 (Cu)	373	6	Amorphous
F34-1	0.6	0.4	0.11 (Cu)	373	4	FAU + ANA
F37a	0.6	0.4	0.11 (Cu)	358	11	FAU
F37	0.6	0.4	0.22 (Cu)	358	20	FAU
Na-FAU	0	0.4	0	358	6	FAU
Co-FAU	0.6	0.4	0.3 (Co)	358	13	FAU
Ni-FAU	0.6	0.4	0.3 (Ni)	358	13	FAU
Zn-FAU	0.6	0.4	0.3 (Zn)	358	13	FAU

SiO_2/Al_2O_3: 10; H_2O/SiO_2: 12. F37 at 358 K, 11 days produces a mixture of FAU + amorphous.

Samples for TEM analysis were ground with a mortar and pestle and sonicated in *n*-butanol prior to dropping on a 300 mesh holey carbon coated gold grid (SPI).

The surface area was measured on a Micromeritics ASAP equipped with ASAP 2010 V4 software. The sample (0.1 g) was loaded into the glass sample bulb and degassed at 423 K for 4 h. Surface areas of the samples were obtained using the Brunauer–Emmett–Teller (BET) model.

The temperature-programmed reduction (TPR) of the samples was done under the following conditions: 200 mg of sample was loaded on a quartz tube (1 cm diameter) and pretreated under flow of helium with a temperature program from 326 to 773 K in 16 min and kept at 773 K for 10 min, the sample was slowly cooled down to 306 K. A 5% mixture of hydrogen in nitrogen was used for the TPR experiment. The temperature was raised from 306 to 773 K in 45 min. The consumption of hydrogen was followed using an MKS-UTI PPT quadrupole mass spectrometer and is related to the presence and position of the copper species in the cages of the zeolite.

Electron paramagnetic resonance (EPR) measurements were performed using an X-band (9 GHz) EPR at 298 K on a Bruker EMX spectrometer. EPR samples were loaded into 3 mm o.d. × 2 mm i.d. quartz tube.

3. Results

3.1. XRD characterization

Table 1 shows the initial batch composition used to prepare metal,Na-zeolite Y by direct synthesis methods. XRD patterns (Fig. 1) show that experiments from 9 to 11, produced mainly amorphous material and some peaks corresponding to copper oxide, gismondine, or faujasite. Experiment 12 produced amorphous material (XRD pattern not shown). According to XRD patterns, the batch composition used in experiment 11 produced amorphous material and some faujasite. Experiment F34-1 had almost the same batch composition as experiment 11; the main difference is that the Cu/Al_2O_3 was reduced from 0.67 to 0.11. The XRD patterns in Fig. 2 show that faujasite and analcime were obtained when the conditions for experiment F34-1 were used. With the same batch composition (F34-1) and increasing the crystallization time, analcime became the predominant phase. FESEM micrograph of sample F34-1 (Fig. 3) confirms the presence of the two phases. There are two different types of spheres.

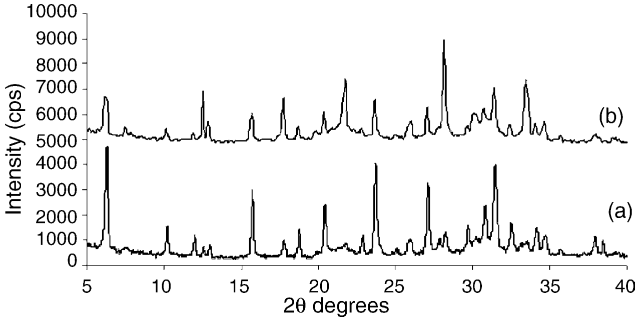

Fig. 2. XRD patterns of faujasite prepared with batch composition F34-1 (see Table 1) and crystallized at 373 K for: (a) F34-1: 4 days and (b) F34-2: 6 days.

Na-FAU was synthesized using the same batch composition as F34-1 but without copper to define the optimum temperature to obtain pure faujasite. Faujasite without analcime was obtained using the batch composition of Na-FAU at 358 K and for 6 days. Experiment F37a was performed using the same batch composition as experiment F34-1. The crystallization temperature was decreased to 358 K and the crystallization time was increased to 11 days. Crystalline faujasite was the only phase detectable by X-ray diffraction (Fig. 4). The XRD patterns for Na-FAU and for the copper-exchanged zeolite Y are also presented in Fig. 4 for comparison. Increasing the amount of copper added to the reacting gel causes a reduction in the crystallization rate. The experiment produced with batch composition F37 (twice the Cu/Al ratio compared to experiment F37a) was mainly amorphous after 11 days of crystallization at 358 K. Small faujasite peaks were detected.

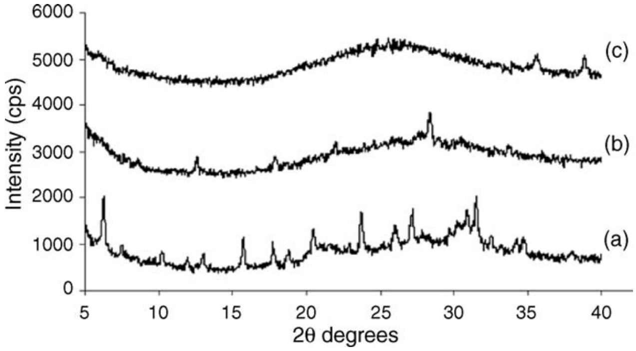

Fig. 1. XRD patterns for experiments: (a) faujasite and amorphous material (experiment 11—initial batch composition $0.67CuO:4Na_2O:Al_2O_3:10SiO_2:120H_2O:6NH_4OH$), (b) experiment 9 (initial batch composition $0.67CuO:4Na_2O:Al_2O_3:10SiO_2:120H_2O:12NH_4OH$), and (c) experiment 10 (initial batch composition $0.67CuO:2Na_2O:Al_2O_3:10SiO_2:120H_2O:12NH_4OH$). Crystallized during 6 days at 373 K.

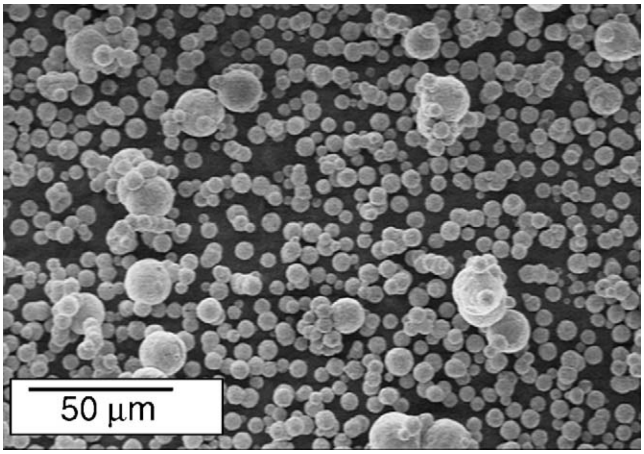

Fig. 3. FESEM of Cu,Na-faujasite produced in experiment F34-1.

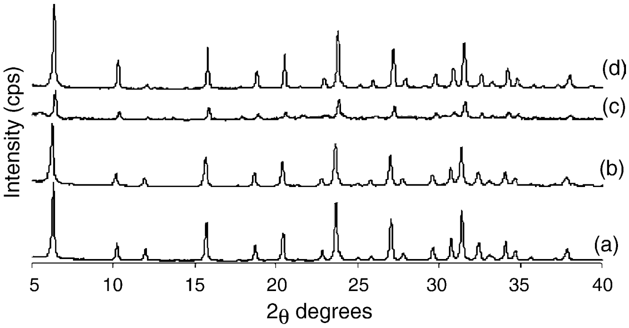

Fig. 4. XRD patterns of zeolites: (a) Na-FAU, (b) F-37a, (c) F-37, and (d) Cu,Na-Y ion-exchanged.

3.2. Morphology (FESEM/TEM) and surface area

Fig. 5 presents the FESEM micrographs for experiments Na-FAU, F37a, F37, and Cu,Na-Y ion-exchanged. In general, all the samples are aggregates. Na-FAU, F37a, and F37 are spherical aggregates. The particles forming the aggregates of F37a and F37 look smaller than the aggregates of Na-FAU (exact size is difficult to measure); the morphology is very uniform throughout the sample. TEM micrographs of samples Cu,Na-Y ion-exchanged and F37a are presented in Fig. 6. Particles of about 5 nm distributed on the surface of the zeolite were observed in both samples. TEM micrographs of cobalt and zinc faujasite are also shown in Fig. 6. Crystals of around 5 nm were observed on the surface of cobalt faujasite but not in the surface of zinc faujasite. Nickel faujasite also had crystals of around 5 nm on the surface (TEM not shown). Only faujasite was detected by XRD when cobalt, nickel, or zinc were used in the synthesis of faujasite (XRD not shown). Faujasite prepared in the presence of cobalt, nickel, or zinc had the same morphology as the faujasite prepared in the presence of copper. Particles of amorphous material were detected by SEM when cobalt, nickel, or zinc were used in the preparation of faujasite.

Table 2
Surface area (BET) of copper containing zeolites

	Surface area (m^2/g)
Cu,Na-Y ion-exchanged	625
F37a (358 K, 11 days)	675
Na-FAU	770

Table 3
EDX analyses of metal-zeolite samples (molar ratios)

Sample	SiO_2/Al_2O_3	Metal/Al (metal)	Na/Al
Cu,Na-Y ion- exchanged	4.33	0.34 (Cu)	0.57
F37a (358 K, 11 days)	3.97	0.12 (Cu)	0.80
F37 (358 K, 20 days)	3.91	0.16 (Cu)	0.80
Na-FAU	3.84	0	0.53
Co-FAU	3.61	0.2 (Co)	0.8
Ni-FAU	3.72	0.3 (Ni)	0.7
Zn-FAU	3.76	0.2 (Zn)	0.6

The surface area of samples F37a, Cu,Na-Y ion-exchanged and Na-FAU is presented in Table 2. The presence of copper in both zeolites causes a decrease in the surface area of the zeolites. The largest surface area reduction was observed for Cu,Na-Y zeolite prepared by ion-exchange (19%).

3.3. Evidence of the presence of copper

The SiO_2/Al_2O_3, Cu/Al, and Na/Al molar ratios (EDX) of the prepared zeolites are shown in Table 3. The sample with the highest content of copper ions is the sample prepared by ion-exchange. All the samples have sodium in the structure. The

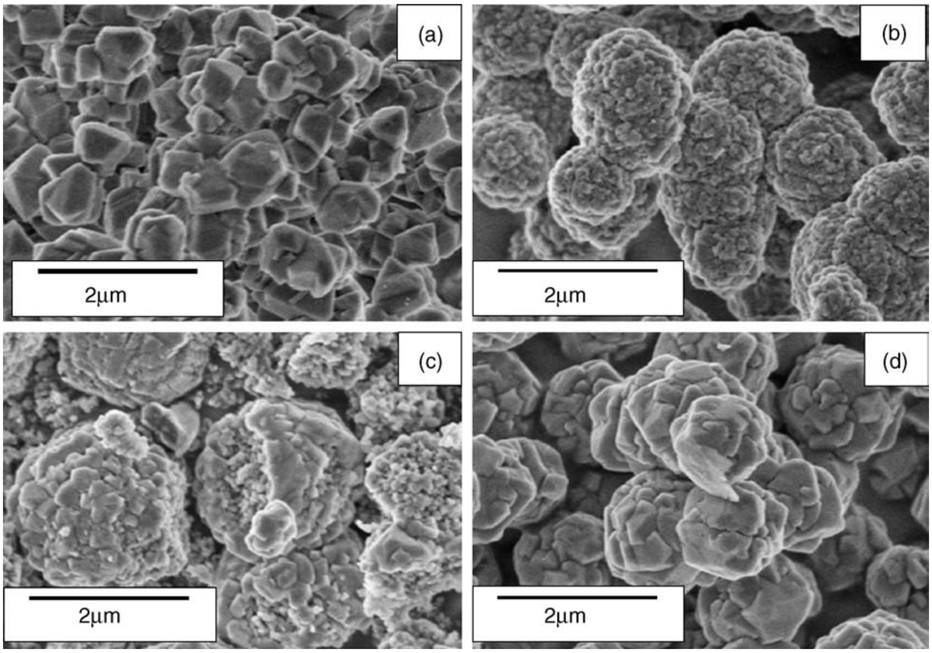

Fig. 5. FESEM micrographs of samples: (a) Cu,Na-Y ion-exchanged, (b) F-37a, (c) F-37, and (d) Na-FAU.

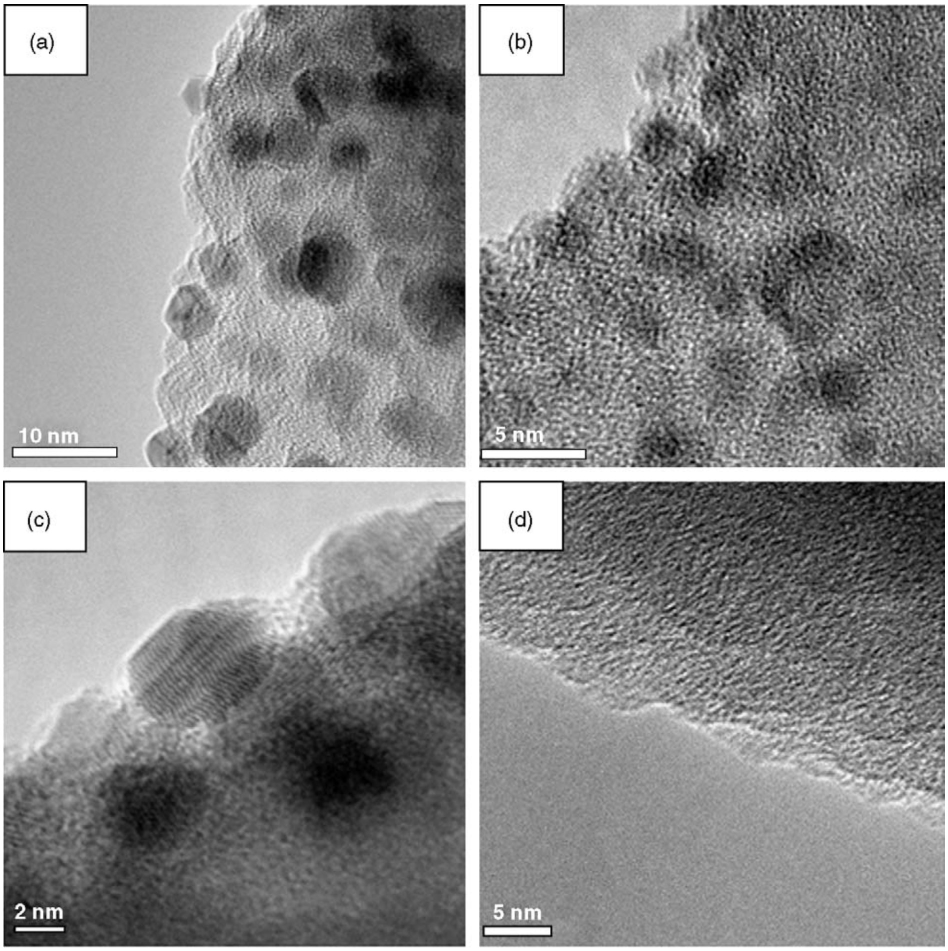

Fig. 6. TEM micrographs of samples: (a) Cu,Na-Y ion-exchanged, (b) F-37a, (c) Co-FAU, and (d) Zn-FAU.

copper content (wt.%) of the zeolite prepared in experiment F37 increased 25% when compared to the one produced in experiment F37a that had half the molar ratio of Cu/Al in the starting gel. Faujasite prepared with cobalt, nickel, or zinc had a metal/Al molar ratio of 0.2 or 0.3 compared to 0.12 or 0.16 for faujasite prepared with copper. Sodium was detected by EDX in all the samples.

The results of temperature programmed reduction for zeolites F37a, F34-1, and Cu,Na-Y ion-exchanged are presented in Fig. 7. The hydrogen consumption for samples prepared by direct synthesis is half than for the sample prepared by ion-exchange. The position of the reduction peak maximum is shifted 70 K to higher temperature for the samples prepared by direct synthesis. The maximum hydrogen consumption for sample prepared by ion-exchange is observed at around 500 K, while for the samples F37 and F34-1 the peak is closer to 570 K.

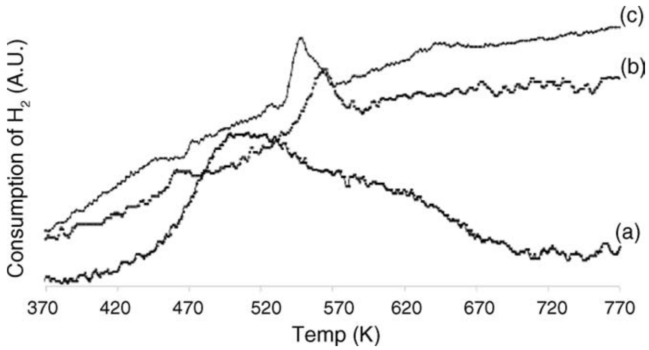

Fig. 7. TPR profile of zeolites: (a) Cu,Na-Y ion-exchanged, (b) F34-1, and (c) F37a.

EPR results are shown in Fig. 8. Cu,Na-ion-exchange faujasite has only one signal *g*: 2.16, for sample F37a there are several signals with *g* values 2.3 and 2.05. The only difference observed between samples Cu,Na-Y ion-exchanged, F37a, Zn-FAU, and Na-FAU by FT-IR analyses (Fig. 9) is that the samples containing copper or zinc have a band around 400 cm^{-1}.

4. Discussion

Experiments 9–12 produced amorphous material with copper oxide, gismondine, or faujasite. The Na_2O/SiO_2 ratio of experiment 10 is too low (0.2) to be able to dissolve and form enough silicate species from the solid gel. Also the addition of more ammonium hydroxide (NH_4OH/SiO_2: 1.2) increases the dilution of the system. Increasing the dilution of the system and decreasing the amount of silicate species generates more amorphous material. Copper(II) hydroxide can be prepared by addition of sodium hydroxide to copper nitrate. Copper

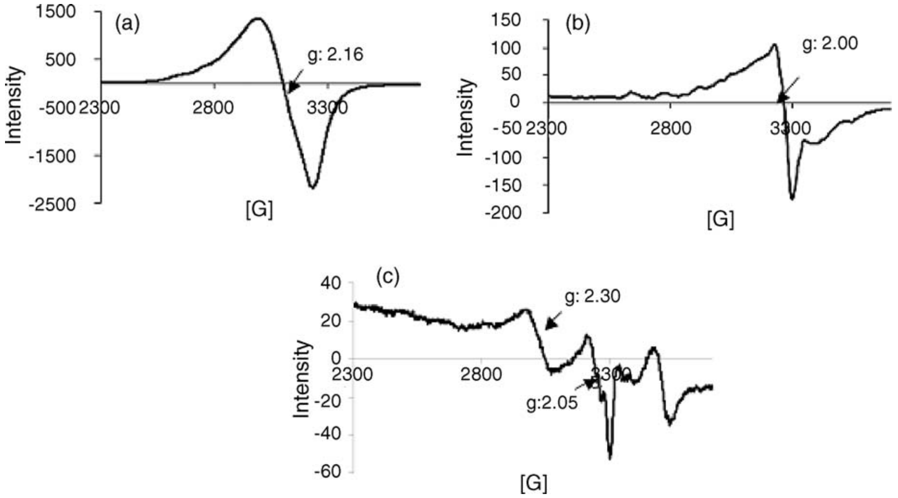

Fig. 8. EPR spectra for samples: (a) Cu,Na-Y ion-exchanged, (b) F34-1, and (c) F37a.

hydroxide decomposes at temperatures above 323 K in the presence of excess alkali to form copper oxide and water. Copper(II) hydroxide is soluble in ammonia solution [9]. In experiment 10, copper tetrammine nitrate was prepared in situ by dissolving copper nitrate in ammonium hydroxide. The copper nitrate that did not dissolve in ammonium hydroxide could react to form copper hydroxide. Under the crystallization conditions used (373 K, 6 days), the copper hydroxide could have decomposed to copper oxide. Those could be the reasons why experiment 10 produced copper oxide and amorphous material. Copper tetrammine nitrate was used as the source of Cu^{2+} to avoid the formation of copper oxide observed in experiment 10. Increasing the ratio of Na_2O/SiO_2 to 0.4 (experiment 9) helps to form more silicate species in solution and to form some gismondine. Experiment 11 has the right Na_2O/Al_2O_3 and NH_4OH/SiO_2 to produce faujasite but still with some impurities. Experiment F34-1 with a decreased CuO/Al_2O_3 molar ratio (0.11) compared to experiment 11 (0.67) produced faujasite with higher crystallinity (≈50% versus 20%).

Experiment F37 with twice the Cu/Al_2O_3 molar ratio as F37a requires twice the time to produce crystalline faujasite. Increasing the Cu/Al_2O_3 molar ratio affects the crystallization rate of faujasite. The surface of the faujasite crystals produced in the presence of copper ions is rougher than the surface of the crystals of sodium faujasite. Crystals of around 5 nm were detected by TEM on the surface of the faujasite. These crystals may correspond to copper oxide. TPR results showed a very small peak at around 470 K. This peak has been previously associated with the presence of copper oxide. Cu^{2+} ions in CuO are reduced to Cu^0 at lower temperatures (470 K) compared to Cu^{2+} at ion-exchange positions in the zeolite lattice [10]. Since the CuO crystals are so small (5 nm) they cannot be detected by XRD. The copper oxide crystals deposited on the surface of the faujasite may limit the crystal growth process. The aluminosilicate surface of the crystal is not fully available to interact with aluminosilicate species in solution.

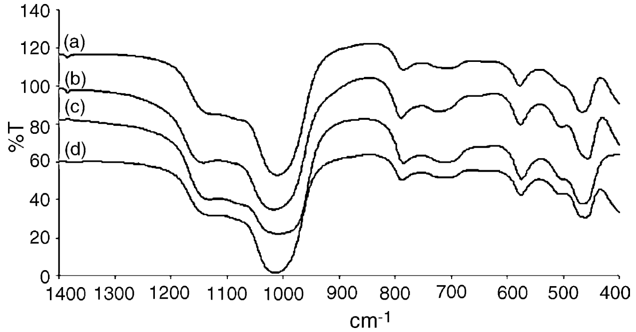

Fig. 9. FT-IR spectra for samples (a) Zn-FAU, (b) Cu,Na-Y ion-exchanged, (c) Na-FAU, and (d) F37a.

The position of copper ions in the faujasite structure is different to the position of copper ions in the ion-exchanged zeolite. In previous studies, two maxima for the reduction of Cu-Y zeolite prepared by ion-exchange have been observed. A two-step mechanism in the reduction of copper ions was proposed [10]. The two different peaks were also related to the reduction of Cu^{2+} in different positions in the zeolite framework. The ions in the supercage would be the most readily reduced, followed by the ions in the sodalite units. The most difficult ions to reduce would be those located in the hexagonal prisms [10]. The six rings are the preferred coordination sites for Cu^{2+} at small Cu^{2+} loadings [11]. According to TPR results, Cu,Na-Y ion-exchanged has most of the copper ions in the supercage of the zeolite, while for the zeolite prepared in experiment F37a the copper ions are located in a less accessible position for reduction. Higher temperature (570 K versus 500 K) is required to reduce the copper ions present in copper faujasite prepared by direct synthesis. Copper ions have more opportunity to interact directly with silicate and aluminate species during the gel formation and later crystallization of the zeolite.

The surface area of faujasite prepared by ion-exchange and by direct synthesis is reduced compared to the surface area of sodium faujasite. Copper ions inside the cages or the 5 nm particles detected by TEM on the surface of both samples of faujasite can block the access to the pores of faujasite causing the reduction in the measured surface area.

The copper content of faujasite prepared by direct synthesis is lower than for the sample prepared by ion-exchange. Decreasing the content of sodium in the synthesis of zeolite Y causes the formation of just amorphous material (results not shown). Sodium is a template for the formation of zeolite Y.

EPR spectra of Cu,Na-Y prepared by ion-exchange shows just one line. At high copper-exchange levels (>20%) spin-exchange interactions between paramagnetic ions makes it impossible to resolve the spectra. EPR spectra consist of a totally symmetric line [12]. In a previous report the EPR spectra of isolated Cu^{2+} ions were observed for samples of Cu,Na-Y with high degree of ion-exchange (54 and 74%) [13]. The relative intensity and the number of EPR parameters also depend on evacuation time and temperature prior to analysis of the sample. For zeolite Y with low levels (<17%) of Cu^{2+} (prepared by ion-exchange) the parameters found are $g^1_{\parallel}$: 2.38, $g^1_{\perp}$: 2.06, $g^2_{\parallel}$: 2.30, $g^2_{\perp}$: 2.03 [14]. The copper cations in the hydrated zeolite Y exist as free aquo-complexes located mainly in the supercages of the zeolite structure. Slow dehydration allows the Cu^{2+} ions to migrate into the small cages (sodalite and hexagonal prisms) [14]. Sample Cu,Na-Y ion-exchanged has an EPR with only one signal this maybe due to the high concentration of copper ions present in the sample as shown by EDX analysis (Cu/Al molar ratio: 0.34). Sample F37a presents several signals in the EPR spectrum, the g values are also similar to those reported for Cu,Na-Y ion-exchanged at low levels. The copper content of sample F37a is low according to EDX analysis (Cu/Al: 0.12). The differences in the g values reported and observed for F37a may be due to the different degree of dehydration of the samples. According to EPR results, there are low levels of copper ions in the sodalite cage and in the hexagonal ring of the copper containing faujasite prepared by direct synthesis.

FT-IR spectra of Cu-zeolite samples prepared by ion-exchange and by direct synthesis are similar to the spectrum of sodium zeolite Y. There are not distortions in the region between 1250 and 900 cm^{-1}. In case of isomorphous substitution, distortion of the FT-IR spectra in the region 1250–900 cm^{-1} is expected. This region of the spectrum is assigned to asymmetrical T–O–T stretching [15]. Cobalt, nickel, and zinc faujasite also present the characteristic bands of faujasite. There is no distortion of the bands in the region 1250–900 cm^{-1}. No isomorphous substitution of copper in the framework of faujasite was found.

5. Conclusion

Cu,Na-zeolite Y can be prepared by direct synthesis using the molar ratios: Na_2O/SiO_2: 0.4, $SiO_2/(Al_2O_3 + CuO)$: 0.9, H_2O/SiO_2: 12, Cu/Al_2O_3: 0.11-0.22 and crystallized at 358 K during 11–20 days. Increasing the amount of copper complex added to the reacting gel causes a reduction in the crystallization rate of zeolite Y.

Copper was detected by EDX. Nanoparticles (5 nm) of a copper compound were detected on the surface of Cu,Na-zeolite Y (ion-exchanged and prepared by direct synthesis) by TEM, these particles could be copper oxide. Copper ions (2+) in the cages of the zeolite should also be present according to the TPR result. Copper present in zeolite Y limits the possibility of nitrogen adsorption used to measure BET surface area. Cu,Na-Y prepared by ion-exchange and by direct synthesis present a reduction in surface area compared to sodium zeolite Y. Isomorphous substitution of copper in the framework of faujasite was not detected by FT-IR. The direct synthesis method reported here was also successfully used to synthesize cobalt, zinc, and nickel faujasite. The characterization used presents valuable information about the location of the copper species but additional characterization is foreseen to have a clearer picture.

Acknowledgements

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