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Preparation of Narrow Size Distribution Superparamagnetic γ-Fe₂O₃ Nanoparticles in a Sol−Gel Transparent SiO₂ Matrix

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γ-Fe₂O₃/SiO₂ nanocomposites have been prepared using a sol−gel procedure, starting from iron nitrate and triethyl orthosilicate. The addition of acids to the sols resulted in a way to increase particle size, keeping low iron concentrations, and to narrow the particle size distribution of γ-Fe₂O₃ in the glass composite. Thus, the addition of 0.56 mmol HCl (≈0.1 M) to a solution having an Fe/Si molar ratio of 18% leads to an increase of γ-Fe₂O₃ particle size from 6 to 13 nm together with a remarkable decrease in the polydispersity degree of the particle size from 66% to 15%. The iron oxide crystalline phase, the particle size and shape, and the homogeneity of the resulting nanocomposites were studied by X-ray diffraction, Mössbauer spectroscopy, and transmission electron microscopy. ²⁹Si NMR and Fourier transform infrared techniques were used to study the changes in matrix structure due to the presence of acids. The influence of acid addition on the size of the resulting magnetic particles was found to depend on the matrix microstructure, the charge environment, and the presence of the counteranions of the acids. Magnetization studies at room temperature showed superparamagnetic behavior for all the nanocomposites, except for the sample with the largest particle size (13 nm), which exhibits ferromagnetism. Optical measurements showed that as the size of the magnetic particle decreases, a blue shift in the UV−vis absorption spectra occurs, which is related to quantum confinement effects.

Introduction

Magnetic nanocomposites are currently the subject of intense research work due to their novel properties, which could have promising applications in technology.[1] The properties of these materials depend strongly on the particle size, the particle−matrix interactions, and the degree of dispersion of the nanoparticles in the matrix.[2] The ability to control these parameters by chemical modifications of the method of preparation is of crucial importance nowadays from both a fundamental and an industrial point of view. For example, γ-Fe₂O₃/SiO₂ nanocomposites, showing very interesting magneto-optical properties,[3−6] have been recently proposed for the fabrication of optical fiber sensors for low magnetic field measurements. For this purpose, large magnetic particles within the superparamagnetic range (around 10 nm)[7] with narrow particle size distribution (PSD) are required to

obtain the maximum susceptibility at low fields. To use this material as a magneto-optic sensor based on the Faraday effect, high light throughput is required while maintaining the magnetic properties of the nanocomposites. Therefore, a compromise must be taken with regard to the concentration of nanoparticles in the composite.

The sol−gel process has been shown to be a useful preparative route for the synthesis of γ-Fe₂O₃/SiO₂ nanocomposites. They can be obtained either by dispersing coprecipitated iron oxide particles in different sol−gel matrixes[6,8] or by "in situ" precipitation of magnetic nanoparticles during the matrix formation.[9−12] An important advantage of the latter is the excellent optical properties of the resulting materials.[3] Nevertheless, there are many parameters in the sol−gel process that need to be controlled in order to obtain pure γ-Fe₂O₃ nanoparticles with a narrow PSD. Recently, the influence of several factors affecting the size of the nanoparticles and the iron oxide phases obtained in sol−gel preparations has been studied, such as the surface of evaporation/volume ratio of the sol and the thermal treatment temperature.[13] These experimental factors affect the amount of the organic residue in the matrix pores, which plays an important

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role in the reduction/oxidation process of the iron oxide particles during the calcination of the samples.[12] If the amount of alcohol in the pores is low, small γ-Fe$_2$O$_3$ particles are obtained (4−7 nm) requiring temperatures of about 700 °C. However, if the amount of alcohol in the pores is kept high enough, larger particles can be obtained (6−15 nm) at a much lower temperature (400 °C) but with a very broad PSD.[12] The addition of acids to sol−gel preparations has not yet been considered as a way to control the PSD of the resulting nanocomposites. Acid catalysis is expected to give rise to more dense, homogeneous gels with smaller pore size.[14] In this sense, Niznansky et al. obtained particles as small as 5 nm using nitric acid in the preparation of a γ-Fe$_2$O$_3$/SiO$_2$ nanocomposite, but the role of the acid was not clear.[5]

The aim of this work is to control the particle size and PSD of transparent γ-Fe$_2$O$_3$/SiO$_2$ magnetic nanocomposites prepared by the sol−gel method. These will be controlled by the addition of acids to the sol−gel preparation. The effect of the nature and concentration of the acids on the properties of the resulting nanocomposites such as particle size, PSD, and the microstructure of the silica network will be studied. The magnetic and optical properties of the nanocomposites will be measured, and a mechanism for the formation of the particles in the silica network will be proposed, accounting for the role of the acid. The ability to control the size and PSD of magnetic nanoparticles in a transparent silica matrix opens new horizons on the design and development of new materials for a wide range of magneto-optical applications.[3]

Experimental Section

Materials and Preparation. All samples were prepared by the sol−gel method. Fe(NO$_3$)$_3$·9H$_2$O (2.72 mmol) (Fluka) was dissolved in 35 mmol of ethanol (spectroscopic grade, Merck). Tetraethyl orthosilicate (TEOS, 15 mmol) (99%, Aldrich) was added to the iron solution under vigorous stirring, followed by the addition of the corresponding amount of nitric or hydrochloric acid. The Fe/Si molar ratio was initially adjusted to 18%, due to the excellent optical properties reported previously for γ-Fe$_2$O$_3$/SiO$_2$ nanocomposites at this concentration.[3,12] The water used to produce the hydrolysis reaction of the silica alkoxide comes only from the hydration water of the iron salt and from the dilute acid used in the initial solution, the water/TEOS molar ratio being 2.6. The sols were covered and kept for 10 days in an oven at 50 °C to obtain the gels. Different acidic environments were obtained by the addition of nitric or hydrochloric acids in amounts ranging between 0.15 and 0.56 mmol. Samples at this stage (gel samples) will be named in the text as G0 for samples without acid addition, GN$_x$ for samples prepared with nitric acid, and GCl$_x$ for samples prepared with HCl, x being the mmol of acid added to the initial solution. The gels were then heated to 400 °C for 4 h resulting in hard, brittle solids. The calcined samples were named as C0, CN$_x$, and CCl$_x$. To determine the effect of the salt concentration under acid conditions, samples with Fe/Si molar ratios of 15%, 25%, and 30% were prepared, varying the amount of iron salts while keeping constant the amounts of TEOS (15 mmol) and hydrochloric acid (0.56 mmol).

Characterization. The resulting iron oxide particles in samples C0, CN$_x$, and CCl$_x$ were characterized by X-ray diffraction (XRD), transmission electron microscopy (TEM), and Mössbauer spectroscopy. X-ray diffraction patterns were recorded from 10 to 60° (2θ) using Cu Kα radiation in a Philips 1710 diffractometer. Mean particle diameters (D_{RX}) were estimated from the full width at half-maximum height of the most intense peak, corresponding to the (311) reflection of the spinel, using the Debye−Scherrer equation.[15] The lattice parameter a_0 was found by extrapolating the a value obtained at different 2θ degrees

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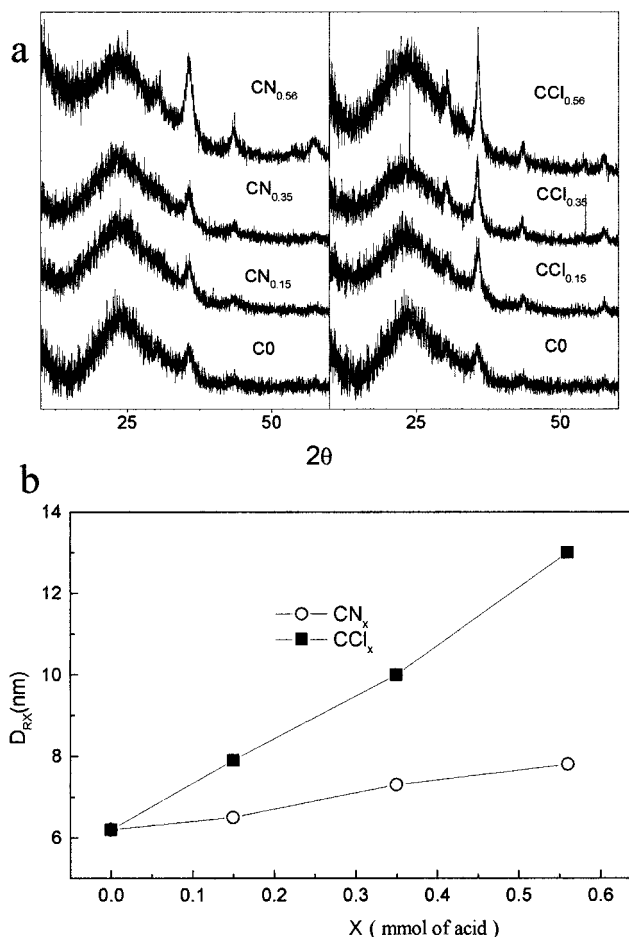

Figure 1. XRD data of samples C0, CN$_x$, and CCl$_x$ with different amounts of acid: (a) diffractograms and (b) crystalline size vs mmols of added acid.

against sin^2 θ through a linear fitting. Particle size and morphology were also examined by TEM in a JEOL electron microscope operating at 200 keV. The samples were scratched, and the powder obtained was dispersed in acetone and dropped on a conventional carbon-coated copper grid. Mean particle size (m) and standard deviation (σ) were evaluated from TEM micrographs, at 250 000× magnification, counting around 100 particles. From these data, the degree of polydispersity, defined as σ/m, was calculated. Mössbauer spectra were recorded in a conventional transmission spectrometer operating in constant acceleration mode with a ^{57}Co source in Rh, from room temperature to 4.2 K. The calibration was undertaken using a 25 μm thick α-Fe foil, giving the Mössbauer parameters relative to this standard at room temperature.

The microstructure of the silica matrix in the gel samples (G) was examined by ^{29}Si NMR measurements and infrared spectroscopy. The ^{29}Si NMR measurements (solid samples) were carried out on a Bruker MSL 400 MHz spectrometer operating under a static magnetic field of 9.4 T. The radio frequency of 79.4 MHz was used for the detection of the ^{29}Si resonance. The peaks were assigned to Q_m (m is the number of bridging oxygen atoms). To obtain the Q_m distributions, Gaussian functions were chosen to deconvolute the NMR spectra by a mathematic fit. The concentration of each species was obtained from the relative intensity of the NMR resonances. Infrared spectra were carried out in a Nicolet FT-IR spectrophotometer model 20SXC using KBr pellets (2 wt % of sample).

Optical properties of the nanocomposites were measured by reflectance on a Varian 2300 UV−vis−NIR between 300 and 800 nm. The samples were ground to a fine powder and placed on a 1 mm wide quartz cuvette. The magnetic properties of samples C were measured using a VSM magnetometer at room temperature. Hysteresis loops were obtained after applying a saturating field of 5000 Oe. Magnetic initial susceptibility χ was calculated

Table 1. Crystallite Size, Particle Size, and Magnetic Parameters of γ-Fe$_2$O$_3$–SiO$_2$ Nanocomposites Prepared with an Fe/Si Molar Ratio of 18% Using Different Acid Conditions

sample (18%Fe/Si)	acid	D_{RX} (nm)	D_{TEM} (nm)	σ/m (%)	Ms (emu/g)	coercitivity (Oe)	χ_i (emu/gOe)
C0		6.2 ± 0.5	6 ± 4	66	1.69	0	0.0032
CN$_{0.15}$	HNO$_3$	7.8 ± 0.8	8 ± 4	50	3.02	0	0.0053
CN$_{0.35}$	HNO$_3$	8.2 ± 0.8	8 ± 4	47	3.51	0	0.0080
CN$_{0.56}$	HNO$_3$	9.4 ± 0.5	8 ± 3	35	4.56	0	0.0111
CCl$_{0.15}$	HCl	7.9 ± 0.5	7 ± 2	28	5.56	0	0.0129
CCl$_{0.35}$	HCl	10.0 ± 0.6	10 ± 2	20	6.75	0	0.0173
CCl$_{0.56}$	HCl	13.1 ± 1.0	13 ± 2	15	7.64	10	

from the linear fitting of M (magnetization) versus H (magnetic field) between -100 and 100 Oe. The value of the saturation magnetization was obtained by extrapolating M versus $1/H$ at high magnetic field.

Results and Discussion

Composite Characterization. The iron oxide phase in the nanocomposites was identified by X-ray diffraction and Mössbauer spectroscopy. Diffraction patterns of samples C0, CN$_x$, and CCl$_x$ at different acid concentrations are presented in Figure 1a. A characteristic broad band of amorphous SiO$_2$ can be observed at $2\theta = 22°$. The rest of the peaks have been assigned to a spinel structure with the characteristic reflections of γ-Fe$_2$O$_3$ or Fe$_3$O$_4$. The calculated value of the lattice parameter ($a_0 = 8.33 \pm 0.02$) and the brown-ochre color of the samples indicate the presence of γ-Fe$_2$O$_3$ ($a_0 = 8.351$) rather than Fe$_3$O$_4$ ($a_0 = 8.3967$, black color). As the acid concentration increases, both a progressive decrease of the broadening and an increase of the peak intensity are observed for samples CN$_x$ and more remarkably for samples CCl$_x$ (Figure 1a). Averaged mean crystal diameters calculated from the peak broadening are shown in Table 1 and Figure 1b. The values are in accordance with the particle size measured from TEM micrographs given in Table 1.

All of the nanocomposites consist of well-dispersed γ-Fe$_2$O$_3$ particles, nearly spherical, embedded in the SiO$_2$ network as shown in Figure 2a for sample CCl$_{0.56}$. This nanocomposite, obtained at an Fe/Si molar concentration of 18%, has a narrow PSD with an average particle size of 13 nm. It should be emphasized that the addition of acid affects not only the particle size but also the PSD, as can be observed by the values of the polydispersity degree (σ/m) obtained from TEM micrographs (Table 1). Thus, the σ/m value of 66% obtained for the sample prepared with no added acids is drastically reduced to 35% due to the addition of nitric acid and further reduced to 15% when hydrochloric acid is used (Figure 2b).

The Mössbauer spectra recorded at 4.2 K showed that no Fe^{2+} ions are present in samples C0, CN$_{0.56}$, and CCl$_{0.56}$. Figure 3a shows the Mössbauer spectra at 4.2 K for sample CCl$_{0.56}$ as an example. Isomer shift values in the range between 0.44 and 0.46 mm/s and very low values of the quadrupole splitting confirm that γ-Fe$_2$O$_3$ is the iron oxide phase present in these nanocomposites. In addition, the hyperfine fields for A (tetrahedral) and B (octahedral) sites were 50.4 and 47.6 kOe for sample CCl$_{0.56}$, 50.8 and 48.1 kOe for sample CN$_{0.56}$, and 50.0 and 46 kOe for sample C0. These values are slightly smaller than the theoretical values for γ-Fe$_2$O$_3$ in bulk but close to the experimental values observed for γ-Fe$_2$O$_3$ particles of about 10 nm.[16] The effect of the acid on the increase in particle size was also confirmed by Mössbauer data at 70 K (Figure 3b). The spectrum for sample CCl$_{0.56}$ is entirely split at this temperature in contrast to the spectra for samples CN$_{0.56}$ and C0, which show, together with a sextet, a central doublet assigned to unblocked (smaller) particles. The
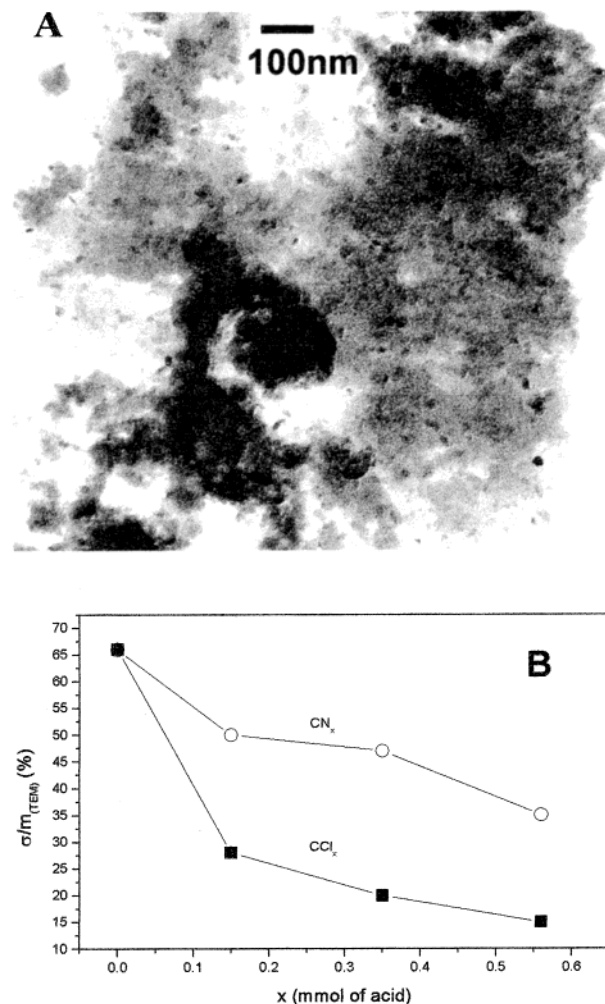
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Figure 2. (a) TEM image of CCl$_{0.56}$ nanocomposite and (b) polydispersity degree (σ/m) vs mmol of HCl and HNO acids in CN$_x$ and CCl$_x$ samples.

doublet contribution to the spectra is much more important for sample C0, which corresponds to the smallest particles. Therefore, the spectra clearly illustrate the difference in particle size between the samples. The difference between the PSD of the samples is also suggested from the Mössbauer spectra at 70 K (Figure 3b), showing blocked (large) and unblocked (small) particles in samples CN$_{0.56}$ and C0, in contrast to sample CCl$_{0.56}$, where only blocked (large) particles are observed.

X-ray diffraction, TEM, and Mössbauer measurements agree that the resulting magnetic particles are larger due to the addition of HNO$_3$ and even larger if HCl is used: $D(C0) < D(CN_{0.56}) < D(Cl_{0.56})$. Moreover, the PSD is substantially reduced by the addition of acids especially in the case of HCl. The degree of polydispersity for sample CCl$_{0.56}$ ($\sigma/m = 15\%$) is lower than the limit established for monodispersed solids (20%)[17] and provides a direct measure of the narrow PSD of the sample. Therefore,
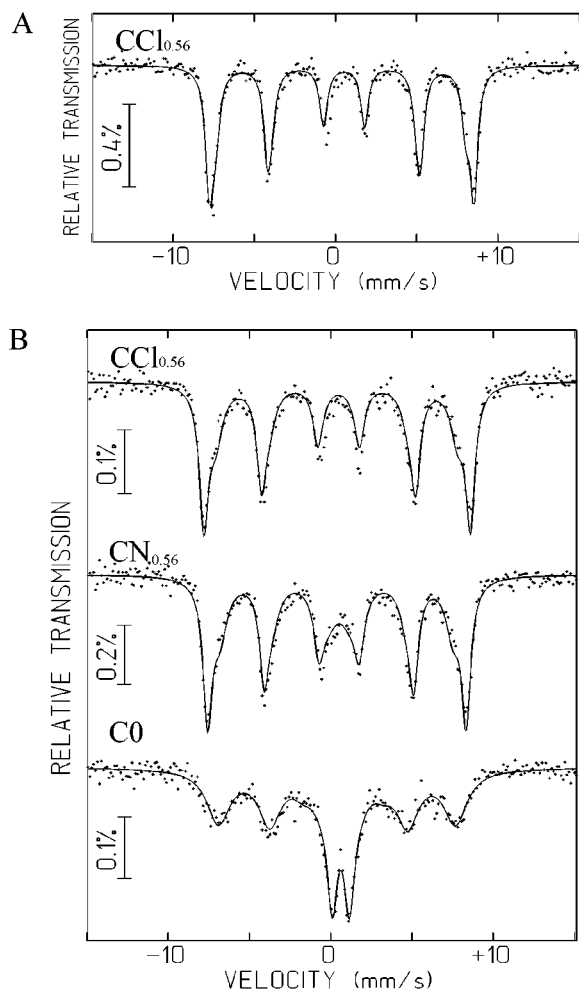
Figure 3. Mossbauer spectra of sample (a) $CCl_{0.56}$ at 4.2 K and (b) samples C_0, $CN_{0.56}$, and $CCl_{0.56}$ at 70 K.

important improvement in terms of narrower PSD of the nanocomposite has been attained by the addition of acids, as compared with nanocomposites prepared by other methods.[18,19]

The concentration of iron in the sample was also shown to affect the size of the resulting magnetic particles. It has been reported that changes in the Fe/Si molar ratio from 15 to 40% resulted in an increase of particle diameter from 6 to 15 nm.[12] These nanocomposites, however, were prepared without the addition of acids and showed a broad PSD with σ/m higher than 60%. The effect of the concentration of the iron (Fe/Si molar ratio of 10−30%) on the size of the resulting nanoparticles was studied by X-ray diffraction (Figure 4). All samples were prepared with 0.56 mmol of HCl. At Fe/Si molar ratios lower than 15%, only a broad band corresponding to SiO_2 amorphous material is present, while concentrations higher than 18% lead to the formation of traces of nonmagnetic α-Fe_2O_3 in addition to γ-Fe_2O_3. The latter is probably due to the γ-Fe_2O_3 to α-Fe_2O_3 phase transition taking place at low temperatures for large γ-Fe_2O_3 particles.[20] The (311) reflection band narrows as the Fe/Si molar ratio is increased from 10 to 30%, indicating an increase of
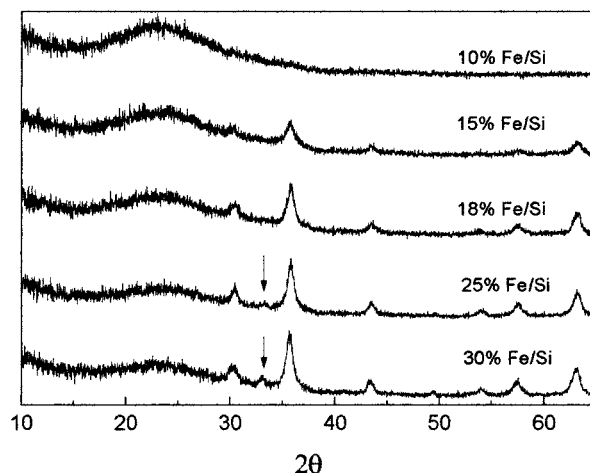
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Figure 4. XRD patterns of γ-Fe_2O_3–SiO_2 nanocomposites prepared with different Fe/Si molar ratios and 0.56 mmol of HCl. Arrows indicate the presence of α-Fe_2O_3.
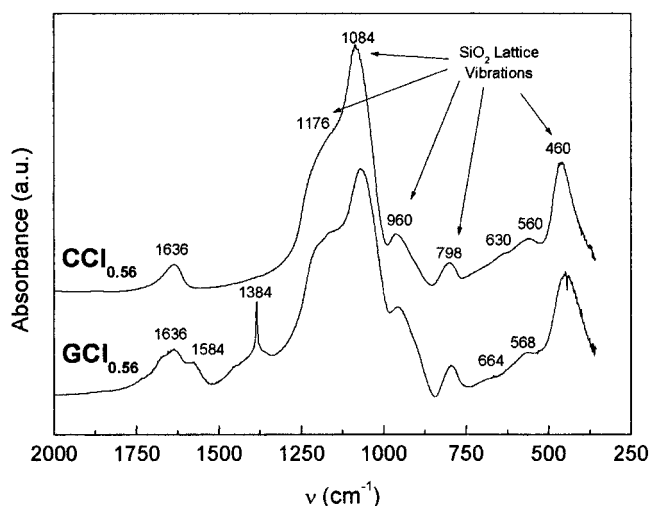

Figure 5. FTIR spectra of samples $GCl_{0.56}$ and $CCl_{0.56}$.

crystallite size. Therefore, an Fe/Si molar ratio of 18% should be used to obtain pure γ-Fe_2O_3/SiO_2 nanocomposites with the largest particle size, at the given acidic conditions.

The changes in the matrix microstructure and the pore environment before and after the heat treatment at 400 °C (G to C transformation) were followed by Fourier transform infrared (FTIR) spectroscopy on samples $GCl_{0.56}$ and $CCl_{0.56}$ (Figure 5). The lattice vibrations of the matrix, marked with an arrow in the figure, showed no significant changes due to calcination. The main difference between both spectra was observed in the bands related to the iron oxide particle formation. Thus, a sharp band at 1384 cm^{-1} attributed to nitrate ions and a band at 1584 cm^{-1} assigned to FeO−H are observed only in sample $GCl_{0.56}$, indicating that nitrate ions are decomposed and the hydrolyzed Fe^{3+} ions form iron oxide particles upon heating. In addition, a shift of the band at 668 cm^{-1} to 630 cm^{-1} is observed from the gel sample to the composite, supporting the transformation of the iron polymers into oxides. The latter band together with the band at 560 cm^{-1} corresponds to Fe−O−Fe bonds in maghemite.[21−22] It is also important to note that the absence of a band at 857 cm^{-1} in the $CCl_{0.56}$ spectrum suggests that no Si−O−Fe bonds are

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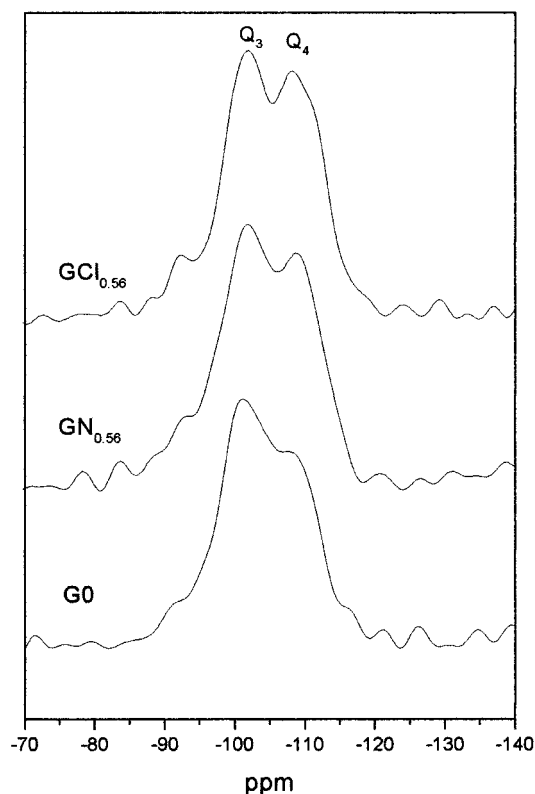
Figure 6. ^{29}Si NMR spectra for samples G0, GN$_{0.56}$, and GCl$_{0.56}$.

formed, in contradiction to the results obtained for Fe-silicalites.[23] This is probably due to the low temperature used for the heat treatment (400 °C). On the other hand, bands associated with water and hydroxyl groups show similar features for the gel (GCl$_{0.56}$) and calcined (CCl$_{0.56}$) samples.

^{29}Si NMR measurements were carried out in samples G0, GN$_{0.56}$, and GCl$_{0.56}$ in order to evaluate the effect of the acid on the structure of the silica matrixes (Figure 6). Due to the fact that NMR measurements can be done only on nonmagnetic materials, samples were measured at the gel stage of the preparation. ^{29}Si NMR measurements show that the acidic matrixes have higher Q4/Q3 ratios (Q$_3$ = (RO)Si(OSi≡)$_3$ (R = H or C$_2$H$_5$) species and Q$_4$ = Si(OSi≡)$_4$ species), suggesting a more cross-linked and consequently more dense structure with fewer unreacted groups. From these results, we can conclude that the presence of acid plays an important role in the microstructure of the matrix at the gel stage. The latter is also true for the calcined samples (C), since no changes in the matrix were observed during the gel to composite transformation, as mentioned before.

Optical and Magnetic Properties. UV−vis diffuse reflectance measurements were carried out for samples C0, CN$_{0.56}$, and CCl$_{0.56}$ (Figure 7). The UV−vis spectrum of γ-Fe$_2$O$_3$ nanoparticles prepared by coprecipitation is given for comparison.[24] A blue shift of the absorption edge from sample CCl$_{0.56}$ to sample C0 is observed, which is attributed to the decrease in particle size[25] as previously observed in γ-Fe$_2$O$_3$ nanocomposites prepared by other

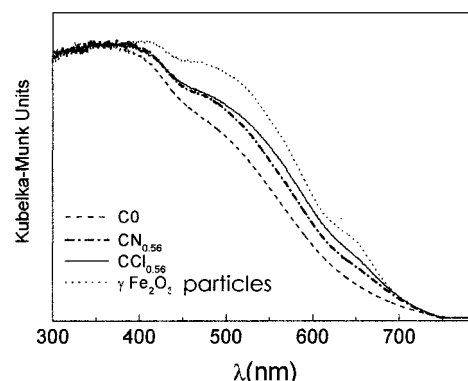(23) Scarano, D.; Zecchina, A.; Bordiga, S.; Geobaldo, F.; Spoto, G.; Petrini, B.; Leofanti, G.; Padovan, M.; Tozzola, G. *J. Chem. Soc., Faraday Trans.* **1993**, *89*, 4123.

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Figure 7. Normalized UV−vis diffuse reflectance spectra of γ-Fe$_2$O$_3$ particle "bulk sample" and C$_0$, CN$_{0.56}$, and CCl$_{0.56}$ γ-Fe$_2$O$_3$ nanocomposites.

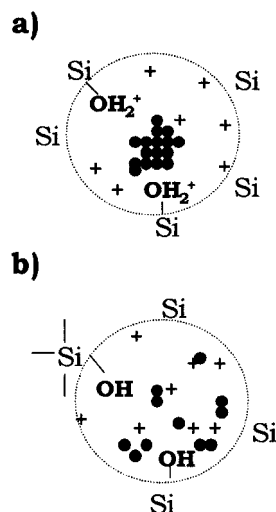**Figure 8.** Scheme of the mechanism of formation of γ-Fe$_2$O$_3$ nanoparticle precursors in a silica matrix for samples prepared (a) with and (b) without the addition of acids.

methods.[5,26] The red shift of the cutoff position of the absorption spectra was also observed by Niznansky et al.[5] and Lei Zhang et al.[8] for similar nanocomposites and assigned to crystal growth of the iron oxide particles in the silica matrix and therefore to a quantum confinement effect in the system. The quantum confinement effect is based on the opening of the electronic band gap between the valence and conduction bands as the particle size decreases resulting in a blue shift in the absorption spectra.[25]

Magnetic properties, such as saturation magnetization (Ms) and initial susceptibility (χ), also reflect the differences between these nanocomposites. The magnetization as a function of the applied field for samples C0, CN$_{0.56}$, and CCl$_x$ is presented in Figure 9. According to their macroscopic magnetization behavior, all samples except CCl$_{0.56}$ behave as superparamagnetic materials, in accordance with their particle size (13 nm). Different behavior was observed for other γ-Fe$_2$O$_3$/SiO$_2$ nanocomposites prepared with nitric acid, which show a remanent magnetization when the applied field goes to zero at room temperature.[5]

The hysteretic parameters of all samples are shown in Table 1. The initial magnetic susceptibility and the saturation magnetization increase as the mean particle

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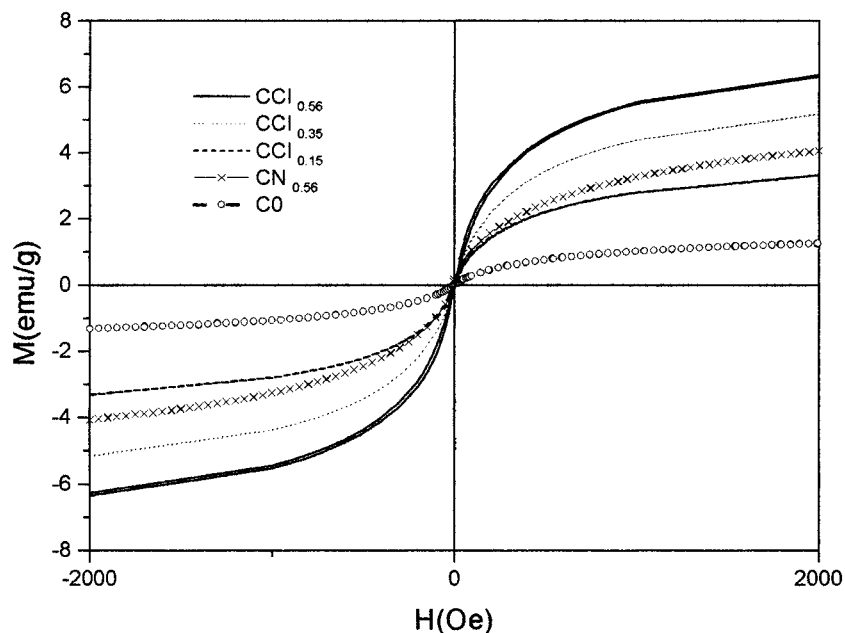
Figure 9. Magnetic hysteresis loops of the samples C0, $CN_{0.56}$, and $CCl_{0.56}$. The inset shows Ms versus mmol of HCl acid.

size increases.[27,28] In this particle size range, there is a clear transition from superparamagnetic (<13 nm) to ferromagnetic (>13 nm) behavior. Sample $CCl_{0.35}$ showing large superparamagnetic particles of 10 nm and a polydispersity degree of 20% exhibited the maximum susceptibility at low field.

Effects of the Acid Addition. The origin of the effect of the acid addition on the size and PSD of the iron oxide particles in the silica matrix can be explained in terms of the changes in the initial iron polymer formation and the changes in the pore surface of the matrix.

The addition of acids to the initial solution of the sol–gel preparation affects the charge environment and introduces new anions to the matrix where the particles are about to be formed. During the gelation process, the hydrolysis of the iron salt leads to the formation of cationic iron hydrolytic polymers (potential of zero charge (pzc) = 7).[29] Taking into account the isoelectric point of the silica (pzc = 2)[29] and considering the initial acid conditions of the solution, an electrostatic repulsion between the protonated matrix pore surface (pH $<$ 2) and the growing iron polymers will take place. This repulsion is expected to be stronger in more acidic conditions. In that case, the repulsion causes iron monomers and dimers to form larger polymers (Figure 8a), which will produce larger particles after heating at 400 °C (13 nm). On the contrary, for samples prepared without acid, the repulsion forces between the matrix surface and the iron polymers are weaker, and therefore a higher dispersion of the iron polymeric species is favored, resulting in smaller particles of about 6 nm after the heat treatment (Figure 8b).

As mentioned before, the addition of acids leads also to a more dense and homogeneous matrix. An increase in the Q4/Q3 ratio in the NMR spectra indicates a decrease in the number of OH groups on the matrix surface and therefore a decrease in the available sites for nucleation of iron oxide particles. Moreover, protonated OH groups (OH–H⁺) will not act as nucleation sites due to the

repulsion forces between positively charged species. This assumption supports the observed effect of the acid on the size of the particles, since an increase in the acid concentration will result in fewer iron nuclei and therefore larger particle size. In addition, the fact that the matrix is more dense, making matter diffusion more difficult, justifies the formation of nanocomposites with narrower PSD, as observed by TEM (Figure 2a).

The nature of the counteranions of the added acid plays an essential role in the iron hydrolytic process[30–32] and can also have an effect on the size of the resulting nanoparticles. Nitrate ions are considered "inert" in the formation of iron hydrous oxides (nonspecific adsorption),[33] while chloride ions are known to substitute OH groups in the iron polymers (specific adsorption), forming larger particles as the chloride ion concentration is increased.[31] Since samples prepared with added HNO_3 showed also an increase in particle size, it can be concluded that both the increase in the proton concentration and the nature of the counteranion of the acid play an important role in the formation of iron oxide particles in the silica matrix.

Conclusions

The addition of acids in the sol–gel preparation of γ-Fe₂O₃/SiO₂ nanocomposites has been shown to be a very interesting way to obtain size-tailored magnetic nanoparticles (between 6 and 13 nm) homogeneously dispersed in the silica matrix, with very narrow PSD.

The increase in particle size has been found to depend on the amount and nature of the acid added. Both protons and chloride anions lead to an increase in particle size of the resulting particles. The effect of the acid and chloride addition on the particle size is attributed to the charge environment (repulsion between iron polymers and pore surface), the matrix microstructure, and the specific adsorption of chloride on growing iron polymers.

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The resulting nanocomposites show a blue spectral shift in the UV−vis absorption spectra as the particle size decreases, associated with a quantum confinement effect in the nanocomposites. Samples C0 and $CN_{0.56}$ behave as superparamagnetic materials according to their particle size. However, ferromagnetic behavior is observed for sample $CCl_{0.56}$ prepared with the same concentration of acid. Large superparamagnetic nanoparticles of about 10 nm, with a polydispersity degree of 20%, were obtained showing the maximum magnetic susceptibility at low field. The ability to obtain single-phase γ-Fe_2O_3 magnetic nanoparticles with controllable particle size and size distribution opens a new route for the design of nanocomposites for different applications.

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