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# Formation of magnetite nanoparticles in poly(acrylamide) gels

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## Abstract

Magnetic gels with magnetite nanoparticles incorporated in a matrix of poly(acrylamide) gel were studied. Magnetite was synthesized through coprecipitation of Fe(II) and Fe(III) in the gel phase, in the solution of linear polymer and in aqueous solution without polymer in alkaline media. The effects of network structure and of the concentration of iron salts in the swollen networks on the composition, structure and properties of magnetic gels have been studied by electron diffraction, XRD, transmission electron microscopy and vibration sample magnetometry. The average size of magnetite nanoparticles,  $D$ , is of the order of 10 nm. It decreases with the increase of polymer concentration in the gel phase. In the dried gels the particles form spherical aggregates (diameter about 150 nm), whereas in the solution of linear polymer, in the aqueous solution of iron salts and in the gel with high content of polymer the aggregates have irregular shape.

## 1. Introduction

Magnetic gels are a new class of soft polymer materials with the properties controlled by magnetic field. They are constituted of swollen polymer networks with incorporated magnetic particles. The first magnetic gels were synthesized by cross-linking of the mixtures of the solutions of polyvinyl alcohol with magnetic fluids [1–3]. Later, magnetic gels based on poly(acrylamide) (PAM) [4] and poly(*N*-isopropylacrylamide) [5] were obtained by three-dimension polymerization in the presence of magnetic fluids. Typically the ferrogels contains several per cent of magnetic colloid particles and about one cross-link per hundred monomer

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**Table 1.** Composition of polymerizing mixtures and characterization of the gel samples.

Sample	AM (wt%)	BAM/AM (mol/mol)	$Q_{H_2O}$	$Q_{SALT}^a$	$Q_{MD}$	$Y$ (%)	$P$ (wt%)
1	6	1/500	35.0	38.8	0.4	99.7	86.5
2	6	1/100	20.3	23.6	0.8	99.4	79.5
3	6	1/30	13.7	17.2	—	98.7	73.5
4	15	1/100	8.7	11.3	1.94	—	61.0
5	30	1/100	5.0	6.5	2.18	103.0	54.0
6	50	1/100	3.4	4.3	1.64	110.5	46.5

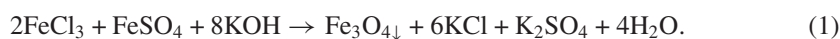
<sup>a</sup> Concentrations of iron salts was 60 wt%.

units of the network [6]. At a size of magnetic particles of the order of 10 nm the gels exhibit superparamagnetic properties [6]. Magnetic gels can be deformed in magnetic field [1–3]. Magneto-elastic properties of magnetic gels can be used to construct sensors, switches and artificial muscles [5]. Their possible applications also include different separation, membrane and drug delivery systems. However, the concentration of magnetic substances in magnetic fluids is limited. Moreover, the formation of the cross-linked network in the presence of magnetic fluid can affect the properties of magnetic particles, for instance causing their aggregation. In this work magnetic particles were synthesized by *in situ* formation of magnetite in the gel phase through coprecipitation of Fe(II) and Fe(III) in alkaline media. The study is devoted to synthesis and characterization of the structure and properties of magnetic gels composed of cross-linked poly(acrylamide) (PAM) and magnetite. The effects of cross-link density and of the concentrations of polymer and salts in the swollen networks on the composition, structure and properties of magnetic gels and magnetite particles were studied by electron diffraction, x-ray diffraction (XRD), transmission electron microscopy (TEM) and vibration sample magnetometry (VSM).

## 2. Experimental section

### 2.1. Synthesis

Acrylamide (AM), N,N'-methylene-(bis)-acrylamide (BAM), ammonium persulfate (PS) and N,N,N',N'-tetramethylethylenediamine (TEMED) were purchased from Sigma-Aldrich Fine Chemicals. PAM gels were prepared by radical copolymerization of AM and BAM in aqueous solutions. 15  $\mu$ l of TEMED and 75  $\mu$ l of 10 wt% aqueous PS solutions were added to 10 ml of the solution of monomers. The concentration of AM during the three-dimensional polymerization and the ratio of BAM to AM are listed in table 1. The copolymerization was performed at 40 °C for 24 h. The gels were obtained between glass plates separated with polyethylene gaskets with a thickness of 0.3 mm. After washing in water for several days the prepared films were dried at 80 °C to constant weight. The dried films were swollen in concentrated aqueous solutions containing FeCl<sub>3</sub> hexahydrate and FeSO<sub>4</sub> heptahydrate taken in the molar ratio 2:1 for 24 h. After swelling in salt solutions the free standing films were treated with an excess of a concentrated solution of potassium hydroxide under mixing. Magnetite was obtained according to the reaction



The yield of reaction (1) in the gel phase depends on the concentration of iron salts and alkali. It can also be affected by the structure and the properties of a swollen PAM network and by the dynamics of diffusion of the components within the gel in the course of the reaction. Analyses of the cuts of the films after the reaction have shown that for obtained PAM films

30 min of treatment with KOH is enough to complete the reaction (1) in the whole volume of the gel. At significantly smaller time the central part of the gel films remains untreated due to rather slow diffusion of KOH into the gel. In the study the concentration of alkali KOH was always kept constant (47 wt%). The obtained magnetic gels were washed with distilled water down to neutral pH. The degree of swelling of the initial PAM gels was defined as  $Q = (m - m_N)/m_N$  where  $m$  is the weight of the sample in water ( $Q_{H_2O}$ ) or in the solution of iron salts ( $Q_{SALT}$ ) and  $m_N$  is the weight of the dried PAM network. The drying of the samples was performed at 90 °C to constant weight. The ratio  $Q$  was determined gravimetrically. For magnetic gels ( $Q_M$ ), instead of  $m_N$  we used the values of  $(m_N + m_M)$ , where  $m_M$  is the amount of precipitate in the gel. The values of  $m_M$  were calculated from the known weight of initial dried PAM network and the weight of the obtained dried magnetic gel. The weight fraction of precipitate (in wt%) in dried magnetic network,  $P$ , was defined as  $P = 100m_M/(m_N + m_M)$ . The theoretical amount of precipitate in the gel  $m_T$  was calculated from the known values of the concentration of iron salts  $C$  and known amount of salt solution in the gel phase using chemical equation (1). The calculation was based on the assumption that the concentrations of salts in the solution absorbed by the gel phase and in the outer solution are equal. The effective yield of reaction (1),  $Y$  (%), was calculated by the formula  $Y = 100m_M/m_T$ .

Together with the gels, magnetite particles were synthesized in linear PAM. The value of  $M_w$  for linear PAM was determined by static light scattering and was equal to 1 070 000. 1 ml of the solution of PAM (concentration 4.0 wt%) in 60 wt% solution of iron salts was added to 10 ml of the solution of potassium hydroxide. The black gel-like precipitate was washed in distilled water down to neutral pH. An analogous procedure was used for obtaining magnetite without polymer. In the latter case black powder was obtained.

## 2.2. Methods

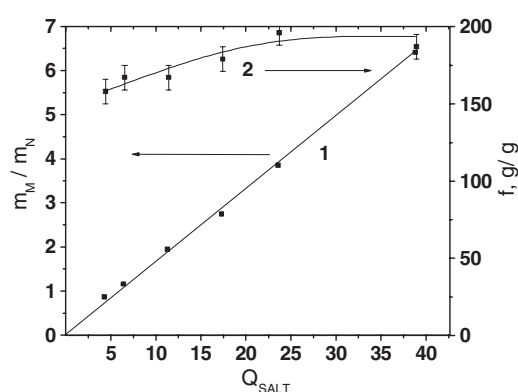
Electron diffraction measurements were performed with an EMP-102 electron diffraction camera ( $U = 75$  kV). TEM images were obtained with a JEM-100C microscope ( $U = 100$  kV).

X-ray diffraction (XRD) measurements were performed with a Rotaflex D/max-RC diffractometer (Rigaku, Japan) at the wavelength  $\lambda = 0.15418$  nm using Cu K $\alpha$  radiation. Average diameters of magnetite nanocrystals were estimated from the full width at half maximum intensity of the peaks corresponding to the crystal crystallographic planes {311} using Scherrer's equation [7]:

$$D_{hkl} = \frac{k\lambda}{\Omega \cos(\Theta_{hkl})}, \quad (2)$$

where  $hkl$  corresponds to the Miller index of the reflecting crystallographic planes,  $D_{hkl}$  is the average diameter of a particle estimated from the peak ( $hkl$ ),  $k$  is the form factor ( $k = 1$  in our case),  $2\Theta_{hkl}$  is the mean angle of scattering peak ( $hkl$ ),  $\Omega$  is the full width at half maximum intensity of the peak (in radians) observed at a mean scattering angle  $2\Theta$ .

The relative magnetic properties of the gels were roughly characterized by the value of the intrinsic force,  $f$  (g/g) exerted by 1 g of precipitate in the magnetic field of given configuration. Discs of equal diameters were cut from magnetic gel films and put in the field of a permanent magnet. The magnet to gel distance  $L$  was fixed constant; the thickness of the films was much smaller than  $L$ . The values of  $f$  were measured using balances. Magnetic properties of the powders of the dried gels were measured using a VSM in magnetic fields up to 8 kOe at room temperature. The obtained values were normalized to unity.



**Figure 1.** Dependences of the ratio  $m_M/m_N$  (1) and of the relative magnetic force  $f$  (2) on the value of  $Q_{\text{SALT}}$ .

### 3. Results and discussion

#### 3.1. Swelling behaviour of magnetic gels

After treatment of the gels containing iron salts by the concentrated alkali, black precipitate is formed in the gel films and they begin to provide magnetic properties. The effect of the cross-linking density and of the concentration of monomers at gel synthesis on the swelling ratio of the gels is shown in table 1. The increase of the cross-linking density of the network and especially the increase of AM concentration at synthesis leads to a decrease of the values of the swelling ratio of PAM gels in water ( $Q_{\text{H}_2\text{O}}$ ). The latter effect is due to significant increase of the concentration of entanglements between the polymer chains in the swollen PAM network. In concentrated solution of mixed salts the swelling ratio  $Q_{\text{SALT}}$  of the gels is higher than in water. This result can be explained by the higher density of the solution of salts in comparison with water.

The comparison of the values of  $P$  for swollen networks with different  $Q_{\text{SALT}}$  shows that the gels having higher degrees of swelling before the reaction of precipitation contain a larger fraction of iron oxides  $P$  in the dried magnetic gels. Figure 1 (curve 1) shows the dependence of the amount of precipitate  $m_M$  formed in the gels and normalized to 1 g of the dried networks  $m_M/m_N$  on the degree of swelling of the network  $Q_{\text{SALT}} = (m - m_N)/m_N$ . The amount of precipitate in the gels with different network structures is directly proportional to the amount of absorbed solution. Thus, in the solution of concentrated iron salts the presence of polymer matrix has only a small effect on the direction and the yield of reaction (1). However, at a small degree of swelling a slight positive deviation from linearity is observed. This deviation can be explained by the formation of a certain amount of iron hydroxides (e.g.  $\text{FeOOH}$ ) in parallel with magnetite in the gel phase [8]. The formation of iron hydroxides is supported by the fact that the experimental yield of the precipitate calculated for reaction (1) for the gels with low degree of swelling becomes higher than 100% (table 1).

The results listed in table 1 show that after magnetite formation the degree of repeated swelling of magnetic gels in water ( $Q_{\text{MD}}$ ) strongly decreases. This effect is more pronounced for the gels with a higher concentration of magnetite. The data of table 2 demonstrate the effect of concentration  $C$  of iron salts in the solution, the effect of  $P$  (which increases with the increase of  $C$ ) and the effect of gel prehistory on the swelling behaviour of gel 2. The dilution of the initial concentrated solution of iron salts does not strongly affect the swelling ratio of the gel  $Q_{\text{SALT}}$ . After magnetite formation in the gels two types of behaviour are observed. The swelling ratio  $Q_M$  of the gels containing high concentration of iron oxide lowers, while that

**Table 2.** Effect of total concentration  $C$  of iron salts on the characteristic parameters of gel 2.

$C$ (wt%)	$Q_{\text{SALT}}^a$	$Q_M$	$Q_{\text{MD}}$	$P$ (wt%)	$f$ (g/g)
60.0	23.6	9.1	0.8	80	$190 \pm 8$
34.4	29.4	11.4	2.8	80	$190 \pm 8$
18.5	28.9	20.3	3.2	68	$148 \pm 8$
4.9	25.3	78.5	20.9	41	$50 \pm 8$
1.6	24.8	102.3	66.3	25	$<10$
0	20.3	100.1	—	—	

<sup>a</sup> Concentration of AM at synthesis was 6 wt%, BAM/AM = 1/100.

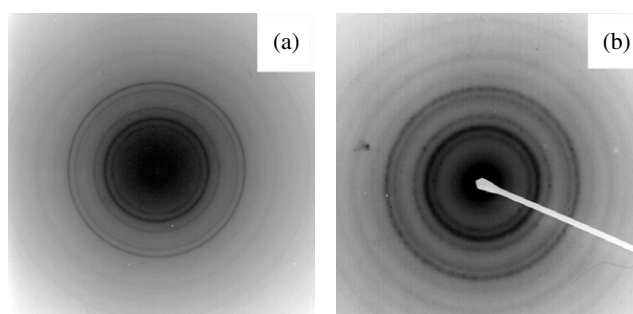
of the gels with low concentration of iron oxide increases markedly. The drying and repeated swelling of all the gels containing magnetite results in a strong decrease of their swelling ratio  $Q_{\text{MD}}$  in comparison with the values of  $Q_M$ . The decrease of the swelling ratio of the gels containing magnetic precipitate is explained by their additional cross-linking by the physical cross-links. The comparison of the swelling ratios of initially swollen magnetic gels  $Q_M$  and those after repeated swelling  $Q_{\text{MD}}$  shows that the physical cross-linking proceeds in two stages. It can be assumed that firstly the primary small isolated magnetic particles are formed. Secondly, during the drying further aggregation of magnetite particles occurs. The comparison of the swelling ratios of initially swollen magnetic gels  $Q_M$  and those after repeated swelling  $Q_{\text{MD}}$  shows that the effective cross-linking and immobilization of the polymer chains of the network occur mainly at the stage of drying. A similar effect was described for the composite gels containing incorporated suspensions of clays [9]. This is explained by the irreversible aggregation of the colloid particles in the gel phase during drying.

The decrease of the concentration of iron oxide in the gel leads to disappearance or marked weakening of the effect of inorganic particles on the swelling behaviour of the gels. The values of  $Q_M$  and  $Q_{\text{MD}}$  become even higher than before the reaction with alkali. The latter result is explained by the fact that in parallel with reaction (1) hydrolysis of PAM occurs. Hydrolysis is accompanied by the appearance of ionized carboxylic groups on the chains of PAM. Due to appearance of the anionic groups together with their counter-ions on the polymer chains the osmotic pressure in the gel phase strongly increases and the gels swell. Reaction of hydrolysis of PAM networks can be used for the regulation of the degree of swelling of magnetic gels.

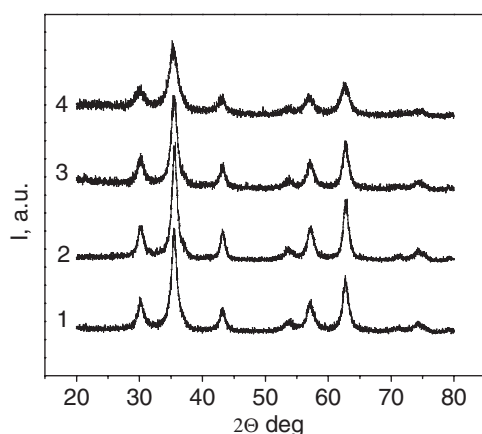
### 3.2. The structure of magnetic particles in the gels

The structure of the precipitate in the gels was studied by electron diffraction. The typical electron diffraction pattern obtained for magnetic gel 2 is shown in figure 2(a). The presence of continuous concentric rings in electron diffraction patterns demonstrates a random polycrystalline structure of the precipitate in the gel. The values of interplanar spacing calculated from the diameters  $2r$  of the rings correspond to magnetite. The characteristic size of the crystallites was roughly estimated from the full width at half maximum intensity of the peak using Scherrer's formula. The characteristic size of the crystallites was about 10 nm. The electron diffraction patterns obtained for magnetic precipitates synthesized in the presence of linear PAM and in aqueous solution in the absence of polymer give the same picture and the same characteristic size of the crystallites.

The other method used in the study of the structure of magnetic particles was XRD. Figure 3 shows XRD profiles obtained for magnetite synthesized in aqueous solution without polymer (1), for magnetite obtained in the presence of linear PAM (2) and for magnetic gels



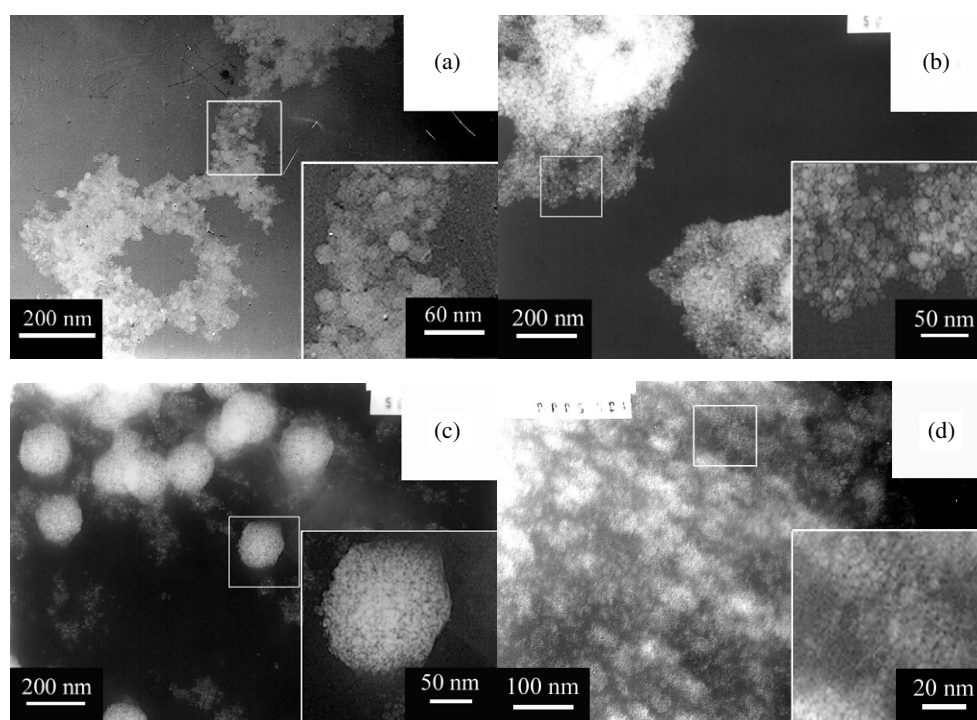
**Figure 2.** Electron diffractograms obtained for magnetic gel 2 at electron beam diameter  $0.2 \mu\text{m}$  (a) and for an individual spherical aggregate of magnetic particles at beam diameter  $0.1 \mu\text{m}$  (b).



**Figure 3.** XRD profiles obtained for magnetite synthesized in aqueous solution without polymer (1), for magnetite obtained in the presence of linear PAM (2) and for magnetic gels 2 (3) and 6 (4).

2 (3) and 6 (4). The position and intensity of the peaks coincide with the data for magnetite (JCPDS-PDF2 card No 75-33). All the peaks on the scattering curves are rather broad, showing a small size of crystalline areas. From the full width at half maximum intensity of the peaks the characteristic diameters of the crystallites  $D$  can be calculated using equation (2). The results of the calculations are shown in table 3. For the particles obtained in water, in 4.0 wt% solution of linear PAM and in PAM gel 2 with the polymer concentration of 4.2 wt% ( $Q_{\text{salt}} = 4.3$ ) the average size of magnetite crystallites practically coincides and lies within 9–11 nm. The increase of PAM concentration in the gel phase during the synthesis from 4.2 to 23 wt% ( $Q_{\text{salt}} = 23.6$ ) leads to the decrease of the size of magnetite crystallites from 9 to 6.0 nm. The difference in the diameters of magnetic crystals in this range of characteristic  $D$  values is important because the transition from ferromagnetic to superparamagnetic properties in magnetic materials is very sensitive to the particle size and shape [10].

More detailed information about the structure of magnetic particles in the gels was obtained by TEM. Figures 4(a)–(d) show TEM images obtained for the magnetite nanoparticles prepared in aqueous solution without polymer (a), prepared in the presence of linear PAM (b), and synthesized in gels 2 (c) and 6 (d). In figure 4(c) large white round circle areas with average size  $L = 155 \pm 17 \text{ nm}$  are observed. The electron diffraction pattern for one of these objects obtained at a small diameter of the electron beam is shown in figure 2(b). It demonstrates a picture analogous to the picture shown in figure 2(a). Hence, the large round objects observed by TEM have polycrystalline structure and consist of randomly distributed magnetite domains.



**Figure 4.** TEM micrographs obtained for the magnetite particles synthesized without polymer (a), in the solution of linear PAM (b), in gel 2 (c) and in gel 6 (d).

**Table 3.** Average particle size calculated from x-ray and TEM data (concentration of iron salts in water during precipitation was 60 wt%).

Sample	$D_{\text{XRD}}^{\text{a}}$	$D_{\text{TEM}}$
Gel 6	6	$3.3 \pm 0.8$
Gel 2	9	$5.3 \pm 1.7$
Linear PAM	11	$6.6 \pm 2.2$
Without polymer	10	$7.7 \pm 2.9$

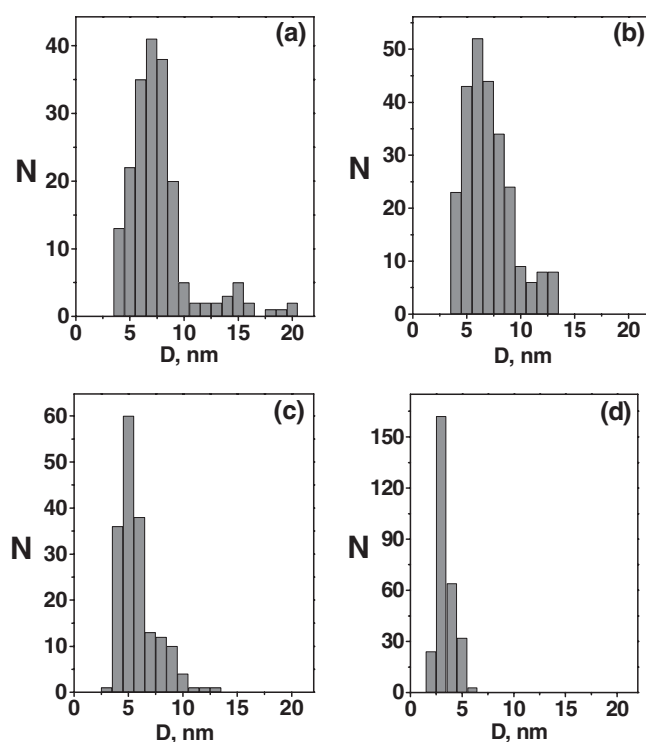
<sup>a</sup> The values of  $D_{\text{XRD}}$  were calculated from the half-width of the main maximum (311) at  $2\theta = 36.0^\circ$ .

The high magnification TEM image of one of the domains is shown in the bottom right corner of figure 4(c). It can be observed that large spherical aggregates are composed of small spherical domains with the characteristic size of several nanometres. In the case of PAM gel 6 with high polymer content (figure 4(d)) the distribution of the particles in the sample is inhomogeneous, but the aggregates do not have a regular shape.

TEM images of the precipitates synthesized in the solution of linear PAM (figure 4(b)) and in polymer-free solution (figure 4(a)) show that both precipitates have an irregular shape. The high magnification TEM images of these precipitates demonstrate that both precipitates are also aggregates of a large number of small domains.

The measurements of the diameters of the domains in the TEM images give their size distributions  $N(D)$  shown in figure 5. The histograms constructed from TEM images for the precipitates in gels 6 and 2 show that in both cases the distributions are unimodal.





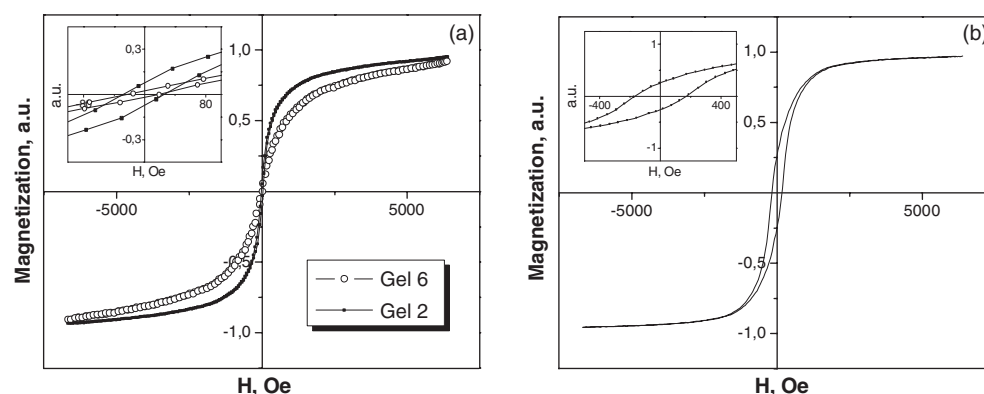
**Figure 5.** Histograms of size distribution  $N(D)$  of the magnetite particles synthesized in aqueous solution without polymer (a), in the solution of linear PAM (b), in gel 2 (c) and in gel 6 (d).

Analogous histograms constructed from TEM images for the precipitates obtained in the solution of linear PAM and in polymer-free solution show that together with the small particles some fraction of larger particles of magnetite is observed.

The average diameters of magnetite nanoparticles in different samples and standard deviations obtained by TEM are shown in table 3. From the obtained data it can be concluded that the narrowest size distribution is observed for the particles obtained in the gel phase with high concentrations of polymer. Magnetite domains obtained in the aqueous solution have the largest average size. Together with small particles a marked fraction of large particles with characteristic size  $D > 16$  nm is present in the precipitate.

### 3.3. Magnetic properties of the gels

For the practical applications it is interesting to compare the magnetization of the magnetic particles in the gels in relatively weak magnetic field i.e. far from saturation. The relative magnetization of the gels was characterized by the value of the intrinsic force,  $f$  (g/g), that acted on 1 g of the precipitate contained in the sample immersed in the magnetic field of a weak enough constant magnet (see experimental section). Curve 2 in figure 1 shows the dependence of  $f$  on  $Q_{\text{SALT}}$  of PAM gels in the concentrated solution of iron salts, i.e. on the initial content of iron ions in the gels. A weak tendency of the magnetic force to decrease with the decrease of  $Q_{\text{SALT}}$  is observed. This effect may be a result of two factors, one of which is the formation of iron hydroxides in the gels. The other reason may be the observed decrease of the size of magnetite crystals in the gel matrixes with enhanced content of the polymer.

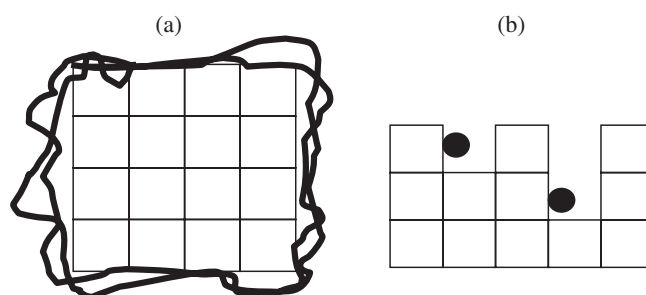


**Figure 6.** Hysteresis loops for the magnetite particles synthesized in gels 2 and 6 (a), and for the powder of magnetite particles with average size diameter of  $0.5 \mu\text{m}$  (b).

The dilution of the solution of iron salts and the formation of iron hydroxides in the gels lead to a strong decrease of the magnetic properties of precipitates in the gel phase. In table 2 the values of  $f$  are listed as a function of the concentration of iron salts  $C$  in the gels before magnetite formation. In the region of salt concentration 34–60 wt% the value of  $f$  is not changing. Further decrease of  $C$  leads to lowering the values of  $f$  which finally falls down to zero at  $C = 1.6 \text{ wt}\%$ . The magnetic susceptibilities of the samples of magnetic gels swollen in water are rather stable. After two months of observations the average decrease of the values of  $f$  was within 4%, i.e. did not exceed the experimental error.

Hysteresis loops were obtained for magnetic gels 2 and 6 and for magnetic particles synthesized in the solution of linear PAM and in water. Figure 6(a) shows hysteresis loops for gels 2 and 6. For comparison figure 6(b) shows hysteresis loops for commercial spherical magnetite particles with average diameter of  $0.5 \mu\text{m}$ . The values of the coercive force  $H_c$  for gels 2 and 6 coincide;  $H_c$  is 24 Oe. For comparison the analogous value for the large particles of commercial magnetite (figure 6(b))  $H = 180 \text{ Oe}$ . The existence of weak coercivity manifests the presence in samples 2 and 6 of some fraction of ferromagnetic particles of magnetite. Much smaller values of remanence and lowering of magnetization loop for sample 6 in comparison with sample 2 shows that the fraction of superparamagnetic particles of magnetite in the former is higher. It should be noted that the low value of the coercive force—about 20 Oe—is typical for single-domain ferromagnetic magnetite nanoparticles. Hysteresis loops obtained for magnetite particles synthesized in the presence of linear PAM and in water without polymer practically coincide with that obtained for gel 2 (figure 6(a)) and therefore are not listed.

Let us discuss the possible mechanisms of limitation of the crystal size in PAM gels. The best known reason for termination of the crystal growth is adsorption of polymer macromolecules on the surface of the growing crystal. Due to the cooperative character of polymer adsorption even weak attraction between the surface of the crystal and the monomer units results in strong adsorption of linear macromolecules [11]. However, in the case of PAM and magnetite this effect is small because the average size of magnetite particles obtained in 4 wt% solution of linear PAM is close to that of the particles synthesized in polymer free solution. At the same time significant decrease of the crystal size is observed in gel 6 with the concentration of polymer in the solution of iron salts 23.0 wt%. We assume that the limitation of growth of magnetite crystals in concentrated gel is due to the fact that polymer chains create physical hindrances for the crystal growth. This situation is schematically shown in figure 7. The polymer chains cut the growing surface in small growing areas with size close to that of



**Figure 7.** Scheme of termination of the crystal growth due to adsorption of polymer macromolecules on the crystal surface (a) and due to creation of physical hindrances for the crystal growth (b). The crystal is represented by squares; the polymer is represented by a black line and circles.

a nucleus. Thus, the rate of growth of the crystals becomes strongly reduced while new nuclei are forming in the system.

#### 4. Conclusion

Hydrogels of PAM are appropriate media for synthesis of magnetic gels by the coprecipitation method. Varying the structure of the gel and the conditions of synthesis, one can obtain magnetic gels with different chemical structures and magnetic properties. The content of magnetite in the dried networks can be varied from low up to very high values (more than 80% by weight). Their swelling degree can be easily regulated by varying the concentration of the cross-links and physical entanglements, by changing the amount of precipitate in the gels, and by varying the polymer chains *via* hydrolyses of amide bonds. Magnetite particles in magnetic gels have an average size of several nanometres. In contrast to the particles synthesized in the solution magnetic domains in dried networks form spherical aggregates of a regular shape.

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