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# Study of magnetisation behaviours for binary ionic ferrofluids

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The magnetisation curves of ionic fluids of three types ($CoFe_2O_4$ ferrofluids, *p*-$NiFe_2O_4$ paramagnetic fluids and $CoFe_2O_4$–*p*-$NiFe_2O_4$ ferrofluids prepared by Massart method) are measured at room temperature and their magnetisation behaviours are studied. Comparison of the experimental data of $CoFe_2O_4$ ferrofluids with the Langevin theory curve demonstrates a considerable difference between them, but the curve fitted by model of a gas-like compression agrees with experimental data very well. The experimental results show that the magnetisation of the $CoFe_2O_4$–*p*-$NiFe_2O_4$ binary ferrofluids is larger than the sum of the magnetisation of the two single ferrofluids in high field. The magnetisation behaviour of the binary ferrofluids is explained by self-assembled ring-like aggregates of $CoFe_2O_4$ ferrofluids particles breaking.

**Keywords:** ferrofluids; binary; magnetisation; aggregate

## 1. Introduction

Generally, ferrofluids are suspensions containing single-domain ferromagnetic or ferrimagnetic nanoparticles about 10 nm in diameter dispersed in a carrier liquid. Under an external magnetic field, the physical behaviours of the ferrofluids, e.g., viscosity behaviour, optical behaviour, etc., would be reasonably changed. The field-induced physical effects of ferrofluids result from transitions of the microstructure, and such transitions depend on the interaction between the particles. Polydispersity of ferrofluids appears naturally since the particles in real ferrofluids always possess a size distribution. In recent years, a bidisperse theoretical model which contains the large and small two size's particles with the same chemical composition is advanced [1–5]. It has been concluded that the large particles constitute the main structure of the ferrofluids under an external magnetic field, and the small particles depending on relative content either suppress or enhance the formation and variation of field-induced structure. However, such a size bidisperse system is difficult to achieve in experiment. In physical nature, the interaction between particles is related to the interaction between magnetic moments. Therefore, for ferrofluids, large and small size (different $d$) particles also can be viewed as strong and

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weak magnetism dipole (different $m$). Nanoparticles' magnetic moment $m$ is proportional to its volume $V$, and the magnetic moment of spherical particles can be given by

$$m = \pi d^3 M/6, \tag{1}$$

where $M$ is the magnetisation of particles, $d$ is the diameter of particles. Ferromagnetism or ferrimagnetism nanoparticles have an inherent magnetic moment: in Equation (1), $M$ is the saturation magnetisation $M_s$ [6]. Paramagnetic particles have an induced magnetic moment: $M$ is expressed by $\chi B$, where $\chi$ is susceptibility and $B$ is the magnetic field strength [7]. Thus, the difference of magnetisation property (different $M$) between the two single systems can be used to synthesise a bidisperse system. In other words, one can use a mixture system of two single systems with different magnetisations, such as ferrimagnetic and paramagnetic systems, as a magnetic bidisperse system instead of a size bidisperse system. The magnetic bidisperse system may have different properties compared with the single magnetic system. We studied the coercive force for a $ZnFe_2O_4$–$CoFe_2O_4$ magnetic nanoparticles system [8]. In this article, the magnetisation behaviours of $CoFe_2O_4$ ferrofluids, $p$-$NiFe_2O_4$ paramagnetic fluids, and $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary ionic ferrofluids prepared by Massart method are studied.

## 2. Experiment

In the present work, the fluids used are ionic ones and prepared by the Massart method [9,10]. The preparation consists of two steps. First, $p$-$NiFe_2O_4$ nanoparticles and $CoFe_2O_4$ nanoparticles are prepared by chemical co-precipitation. The $FeCl_3$ solution and $Ni(NO_3)_2$ solution are well-mixed in a molar ratio of 2 : 1. Then, the mixed solution is added into the appropriate proportion of NaOH solution and heated to boiling. After boiling for 1 min, the heat solution is cooled to room temperature and $Ni\ (OH)_2 \cdot 2Fe(OH)_3$ precipitation, which is the precursor to prepare $NiFe_2O_4$ materials, is obtained [11–13]. The precipitation is cleaned to $pH = 7$. Finally, the precipitation is added to the $Fe(NO_3)_3$ solution and kept boiling for half an hour to obtain the necessary nanoparticles (called $p$-$NiFe_2O_4$ nanoparticles in this article). Using a similar method, $CoFe_2O_4$ nanoparticles are obtained by replacing $Ni(NO_3)_2$ with $Co(NO_3)_2$. Second, the synthesis of $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary ionic fluids. These two nanoparticles are used to produce 1.2% mother fluids, respectively, then the two fluids are mixed with the same volume to obtain the binary ionic fluids.

According to the definition of particle volume fraction, the particle volume fraction in the $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary ionic fluids is written as

$$\phi = \frac{V_1}{V_1 + V_2} = \phi_{\mathrm{Co}} + \phi_{p\text{-}\mathrm{Ni}}, \tag{2}$$

where $V_1$ is the volume of all particles, $V_2$ is the volume of carrier liquid, $\phi_{\mathrm{Co}}$ and $\phi_{p\text{-}\mathrm{Ni}}$ are the volume fractions of $CoFe_2O_4$ and $p$-$NiFe_2O_4$ nanoparticles, respectively, which are dispersed in the $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary ionic fluids. They are defined as

$$\phi_{\mathrm{Co}} = \frac{V_{\mathrm{Co}}}{V_1 + V_2}, \quad \phi_{p\text{-}\mathrm{Ni}} = \frac{V_{p\text{-}\mathrm{Ni}}}{V_1 + V_2},$$

where $V_{Co}$ and $V_{p\text{-Ni}}$ are $CoFe_2O_4$ and $p$-$NiFe_2O_4$ particles volume in the binary fluids. Therefore, in this binary fluids, $\phi_{Co}$ and $\phi_{p\text{-Ni}}$ is 0.6%, respectively. To compare the magnetisation behaviour of the binary system with single systems, $CoFe_2O_4$ fluids and $p$-$NiFe_2O_4$ fluids with particle volume fraction as 0.6% are produced, respectively, by diluting the mother fluids.

X-ray diffraction (XRD, XD-3) is used to analyse the crystal structure of the particle samples, and transmission electron microscopy (TEM, JEM-100CX2) is used to observe the particles morphology and determine the particles diameter. Vibrating sample magnetometer (VSM, HH-15) is used to measure the magnetisation curves of the $CoFe_2O_4$ and $p$-$NiFe_2O_4$ particles, $CoFe_2O_4$, $p$-$NiFe_2O_4$, and $CoFe_2O_4$–$p$-$NiFe_2O_4$ fluids. In the measurement, the magnetisation per unit mass $\sigma$ is measured since it is easier to accurately measure mass [14]. The magnetisation per unit volume is given by $M = \sigma\rho$, where $\rho$ is the density.

## 3. Experimental results

The results of XRD measurement are as shown in Figure 1. It can be seen that the diffraction peak of $p$-$NiFe_2O_4$ particles is unclear and that of $CoFe_2O_4$ particles is broad. These indicate that the particle size should be very fine. Using the Debye–Scherrer formula, the average size of $CoFe_2O_4$ grains is calculated from the (311) plane as 10.06 nm.

The morphology of both the $p$-$NiFe_2O_4$ and the $CoFe_2O_4$ particles are basically spherical as shown in Figure 2, but directly observing under TEM is clearer than the photograph. For the $p$-$NiFe_2O_4$ particles, the median diameter is 3.34 nm and the distribution function is

$$f(x) = \frac{2.22}{x}\exp\left[-15.52(\ln x - 1.21)^2\right]. \tag{3}$$

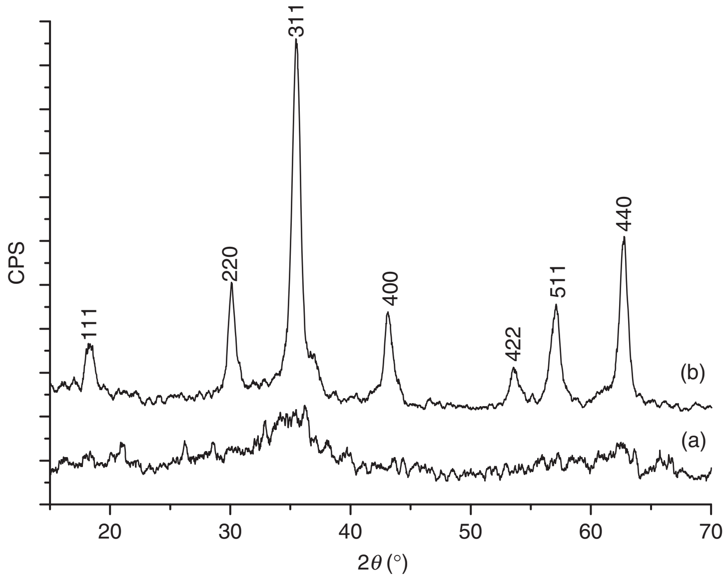


Figure 1. XRD patterns of $p$-$NiFe_2O_4$ particles (a) and $CoFe_2O_4$ particles (b).

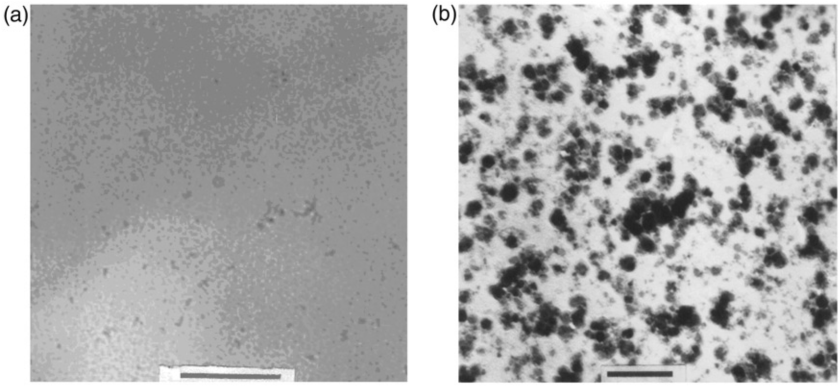


Figure 2. TEM photographs of $p$-$NiFe_2O_4$ particles (a) and $CoFe_2O_4$ particles (b). The scale bar is 100 nm.

For the $CoFe_2O_4$ particles [8], the median diameter is 12.76 nm which agrees with the result of XRD, and the distribution function is

$$f(x) = \frac{1.14}{x}\exp[-4.06(\ln x - 2.55)^2]. \tag{4}$$

The VSM's measurement results indicate that $CoFe_2O_4$ particles are strong magnetic nanoparticles, $p$-$NiFe_2O_4$ nanoparticles are the weak magnetic particles. Two types of particle's magnetisation curves are as shown in Figure 3. According to the linear relation between saturation magnetisation $M_p$ and $1/B$ under high field, when $1/B$ equals to zero, the $M_p$ of $CoFe_2O_4$ particles is estimated to be $220\,\mathrm{kA\,m^{-1}}$. $p$-$NiFe_2O_4$ particles are paramagnetic ones and $M$ is $13.18\,\mathrm{kA\,m^{-1}}$ at 1 T. The magnetisation curves of three types of fluids are as shown in Figure 4. Obviously, the magnetisation behaviour of the $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary fluids are similar to those of the $CoFe_2O_4$ ferrofluids, so that the fluids formed by mixing the $CoFe_2O_4$ ferrofluids and $p$-$NiFe_2O_4$ paramagnetic fluids can be called binary ionic ferrofluids.

## 4. Analysis and discussion

### 4.1. *Magnetisation behaviours of single fluids*

The $p$-$NiFe_2O_4$ paramagnetic fluids magnetisation behaviour can be described by

$$M = \chi B, \tag{5}$$

where $\chi$ is susceptibility. From the experimental result, the susceptibility of the $p$-$NiFe_2O_4$ paramagnetic fluids is $0.05\,\mathrm{kA(m\,T)^{-1}}$. From the measured result as shown in Figure 4, the $CoFe_2O_4$ ferrofluids' saturation magnetisation $M_f$ can be estimated to be $0.95\,\mathrm{kA\,m^{-1}}$ with an error of about 1.05%. While interaction between the particles can be neglected, magnetisation of ferrofluids is described with Langevin theory. If the particles' diameter is the same, then the Langevin magnetisation expression is

$$M = M_f\left(\coth(\alpha) - \frac{1}{\alpha}\right), \tag{6}$$

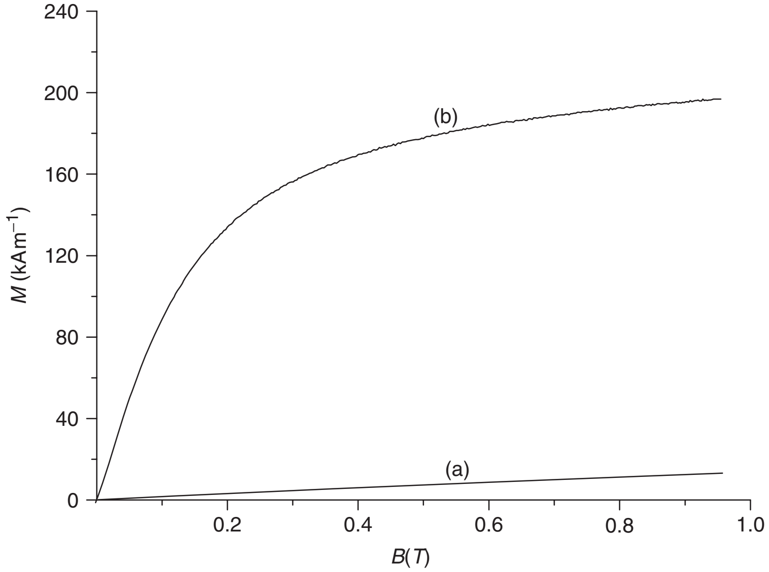


Figure 3. The magnetisation curves of $p$-$NiFe_2O_4$ particles (a) and $CoFe_2O_4$ particles (b).

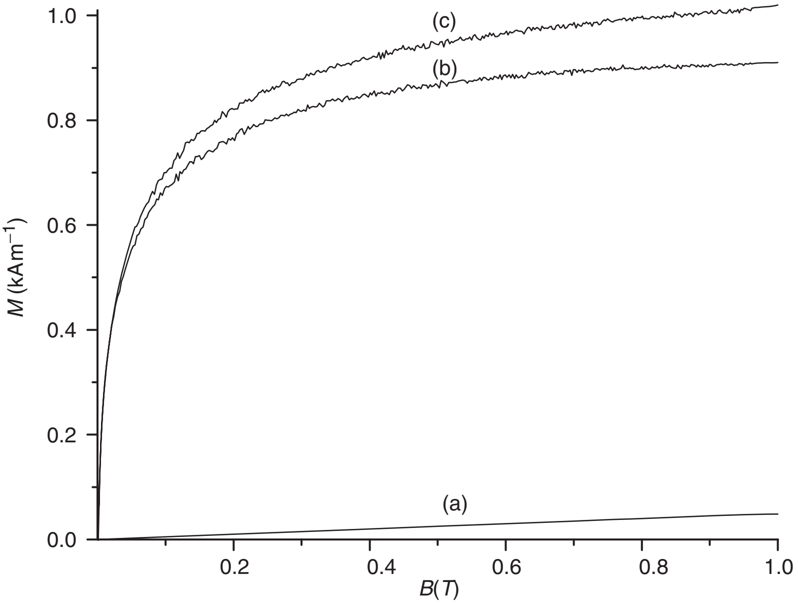


Figure 4. The magnetisation curves of three types of fluids: (a) $p$-$NiFe_2O_4$ fluids with $\phi = 0.6\%$; (b) $CoFe_2O_4$ fluids with $\phi = 0.6\%$; (c) $CoFe_2O_4$–$p$-$NiFe_2O_4$ fluids with $\phi = 1.2\%$.

where $M_f$ is the saturation magnetisation of ferrofluids, $L(\alpha) = \coth(\alpha) - 1/\alpha$ is the Langevin function, $\alpha = mB/kT$ is the Langevin parameter, $m$ is the moment of particle, $B$ is the magnetic field strength, $k$ is the Boltzman constant, and $T$ is the absolute temperature. For calculating the effect of field-induced aggregates on the magnetisation of ferrofluids, a model of gas-like compression (MGC) has been proposed [15]. In this model, the magnetisation is described by

$$M = M_{\mathrm{f}}\left(\coth(\alpha) - \frac{1 + \ln(\phi_H/\phi)}{\alpha}\right), \tag{7}$$

where $\phi_H$ is the volume fraction of aggregate particles and is given by $\phi_H = (0.638 - \phi)\tanh(\gamma(\phi\alpha)^2) + \phi$, $\gamma$ is defined as the compressive parameter and can be got by simulation and, $\gamma$ parameter shows the extent of measured magnetisation curve deviating from Langevion theory from the effect of field-induced aggregates. While $\gamma = 0$, formula (7) is just formula (6). From Figure 5, it can be seen that Langevin theoretical curve has a consideration deviation with the experimental result, but the model of gas-like compression ($\gamma = 12$, the error is estimate at ±1) fits the experimental result very well. The result shows that the $CoFe_2O_4$ ferrofluids could form a chain-like aggregate during the magnetising process.

The relationship between the saturation magnetisation of ferrofluid and the particle is regarded as $M_{\mathrm{f}} = \phi M_{\mathrm{p}}$ [6]. But, for the $CoFe_2O_4$ system, $M_{\mathrm{f}}$ ($=0.95\,\mathrm{kA\,m^{-1}}$) is less than $\phi M_{\mathrm{p}}$ ($=1.32\,\mathrm{kA\,m^{-1}}$). This could be due to the presence of self-assembled ring-like aggregates in the ferrofluids in absence of magnetic field. The experiment and computer-simulation indicated that ferrofluids' particles can self-assemble into chain-like and ring-like structures [16,17]. The chain-like aggregates could not affect the saturation magnetisation, but the magnetic flux from the ring-like structure is a closed loop so that it does not make any contribution to the magnetisation [18]. Therefore the measured saturation magnetisation $M_{\mathrm{f}}$ is less than the $\phi M_{\mathrm{p}}$ for the ferrofluids.

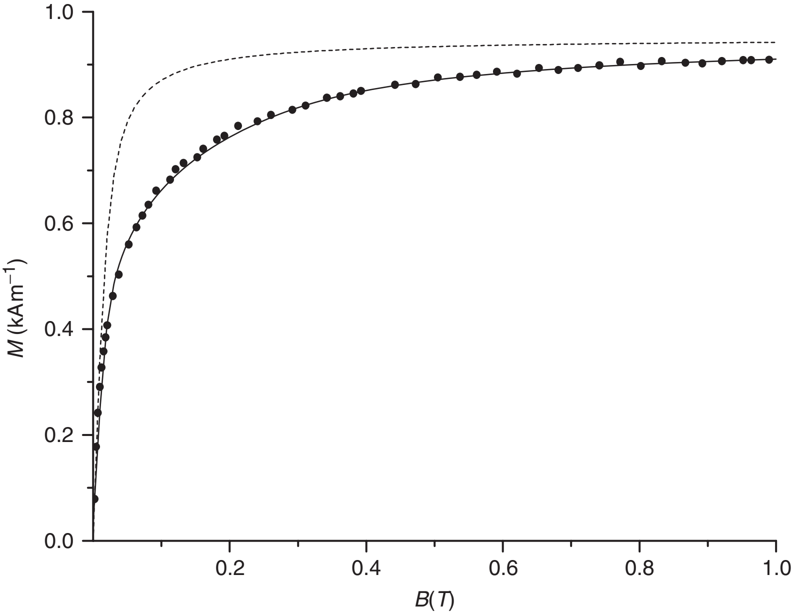


Figure 5. The $CoFe_2O_4$ ferrofluids' magnetisation data as well as the curves of both Langevin theory and MGC. ——— Langevin theory curve . . . Experimental date ---- MGC curve ($\gamma = 12$).

### 4.2. *Magnetisation behaviours of binary ferrofluids*

$p$-NiFe$_2$O$_4$ particles are paramagnetic and the greatest average magnetic moment in the magnetic field is

$$\langle m\rangle = \frac{\pi}{6} M_{1T} \int_0^{\infty} x^3 f(x)\mathrm{d}x,$$

where $M_{1\mathrm{T}}$ is the magnetisation at 1 T. Substituting $M_{1\mathrm{T}}$ and $p$-NiFe$_2$O$_4$ diameter distribution function (3), thus $\langle m\rangle = 3.01 \times 10^{-22}(\mathrm{A\,m^2})$.

CoFe$_2$O$_4$ particles are ferrimagnetic, according to diameter distribution function (4), the average magnetic moment is

$$\langle m\rangle = \frac{\pi}{6} M_p \int_0^{\infty} x^3 f(x)\mathrm{d}x = 4.22 \times 10^{-19}(\mathrm{A\,m^2}).$$

Because interaction between single-domain magnetic nanoparticles can be regarded as the interaction between magnetic moments, the interaction energy between particles $i$ and $j$ is given by

$$E = \left(\mu_0 m_i m_j / 4\pi r_{ij}^3\right)[\cos(\theta_i - \theta_j) - 3\cos\theta_i \cos\theta_j], \tag{8}$$

where $\mu_0$ is the vacuum permeability, $r_{ij}$ is the particles' distance, $\theta_i$ and $\theta_j$ are the angles between orient of the moments and central connections line [6]. When $\theta_i = \theta_j = 0$, two $p$-NiFe$_2$O$_4$ particles ($r_{ij} = d = 3.34$ nm) linked by contact, the greatest interaction energy ($B = 1$ T) is about $4.86 \times 10^{-25}$ J. The interaction energy of two CoFe$_2$O$_4$ particles ($r_{ij} = d = 12.76$ nm) is about $1.71 \times 10^{-20}$ J. While one CoFe$_2$O$_4$ particle and one $p$-NiFe$_2$O$_4$ particle are linked by contact ($r_{ij}$ = (diameter of CoFe$_2$O$_4$ particles + diameter of $p$-NiFe$_2$O$_4$ particles)/2), the interaction energy ($B = 1$ T) is about $4.87 \times 10^{-23}$ J. At room temperature($T = 300$ K), the thermal energy is about $k_B T = 4.14 \times 10^{-21}$ J. Obviously, only dipole interaction between CoFe$_2$O$_4$ particles is larger than thermal energy. So, only the particles in CoFe$_2$O$_4$ ferrofluids can form aggregates during the magnetising process. Thus, CoFe$_2$O$_4$–$p$-NiFe$_2$O$_4$ binary ionic ferrofluids' magnetisation can be expressed by the sum of two single ferrofluids' magnetisation.

The magnetisation of the sum of both CoFe$_2$O$_4$ and $p$-NiFe$_2$O$_4$ ferrofluids as well as the measured magnetisation of the CoFe$_2$O$_4$–$p$-NiFe$_2$O$_4$ binary ionic ferrofluids are as shown in Figure 6. The summed magnetisation is taken from the results at the experiments and from theoretic description, respectively, i. e.,

$$M = M_{\mathrm{Co}}\ \text{(experimental value)} + M_{p\text{-Ni}}\ \text{(experimental value)}, \tag{9}$$

where $M_{\mathrm{Co}}$(experimental value) is the magnetisation data of CoFe$_2$O$_4$ ferrofluids and $M_{p\text{-Ni}}$(experimental value) is the magnetisation data of $p$-NiFe$_2$O$_4$ paramagnetic fluids from Figure 4(a) and (b), respectively, and

$$M = M_{\mathrm{Co}} + M_{p\text{-Ni}} = M_{\mathrm{f}}\left(\coth(\alpha) - \frac{1 + \ln(\phi_H/\phi)}{\alpha}\right) + \chi B. \tag{10}$$

From Figure 6, it is seen that in low magnetic field ($B < 0.15$ T) the experimental data of binary ferrofluids agrees with the summed data or the fitted curve from two single fluids, but in high magnetic field ($B > 0.15$ T) the experimental data is higher than the summed

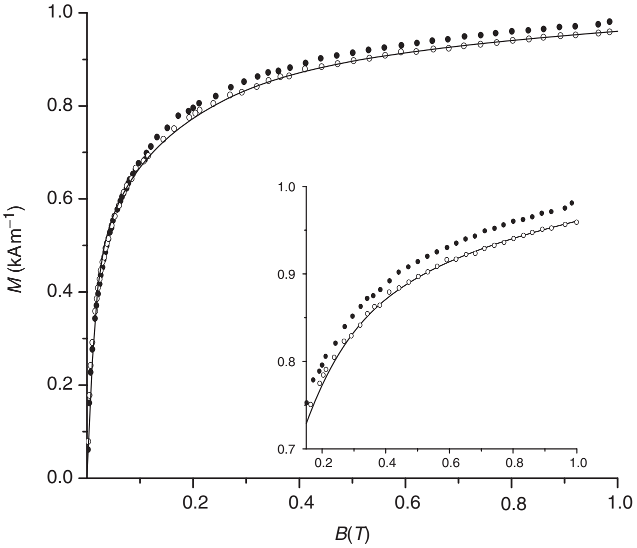


Figure 6. The comparison of both experimental data and fitted magnetisation curve for binary ferrofluids.

Notes: . . . Experimental date of binary ferrofluids.
——Binary magnetisation curve fitted by Equation (10) ($M_f = 0.95\,\mathrm{kA\,m^{-1}}$).
○○○○Summed date of both Figure 4(a) and (b).

data of two single fluids. This shows that the binary ferrofluids' magnetisation behaviours are not a simple summation of those of the $CoFe_2O_4$ ferrofluids and *p*-$NiFe_2O_4$ paramagnetic fluids. In the high magnetic field, the binary ferrofluids' magnetisation curve can be written as $M = M_f(1 - 1/\alpha) + \chi B$. It can be seen from Figure 6 that the experimental data exhibit a different slope from the summed data in the high magnetic field ($B > 0.15\,\mathrm{T}$). So, it is judged that the saturation magnetisation of $CoFe_2O_4$ ferrofluids component $M_f$ in the binary ferrofluids could increase. By fitting, it is found that Equation (10) can agree well with the experimental result while the $M_f$ is taken as $0.97\,\mathrm{kA\,m^{-1}}$, but other parameters are unchanged, as shown in Figure 7. In Figures 6 and 7, the insets show clearly the difference of both the experimental data and the magnetisation curve fitted with different $M_f$ value under the high field. The increment could result from the breaking of the self-assembled ring-like aggregates of $CoFe_2O_4$ ferrofluids' particles while the binary ferrofluids are magnetised as shown in Figure 8. It can be explained as follows.

While the binary ferrofluids are synthesised with $CoFe_2O_4$ ferrofluids and *p*-$NiFe_2O_4$ paramagnetic fluids, the self-assembled chain-like and ring-like aggregates from $CoFe_2O_4$ particles keep still and some *p*-$NiFe_2O_4$ particles could be in the rings. During the magnetising process, the *p*-$NiFe_2O_4$ particles would be the induced moments orienting as the direction of the applied magnetic field, and they act as magnetically polarised gas. The induced moments will enhance with the magnetic field arising, so that the *p*-$NiFe_2O_4$ particles can repel each other because magnetic interaction among them make the rings break into particles and chains, as shown in Figure 8. This is similar to the 'frozen treatment' effect, in which expansion of water molecules can damage the closed circular

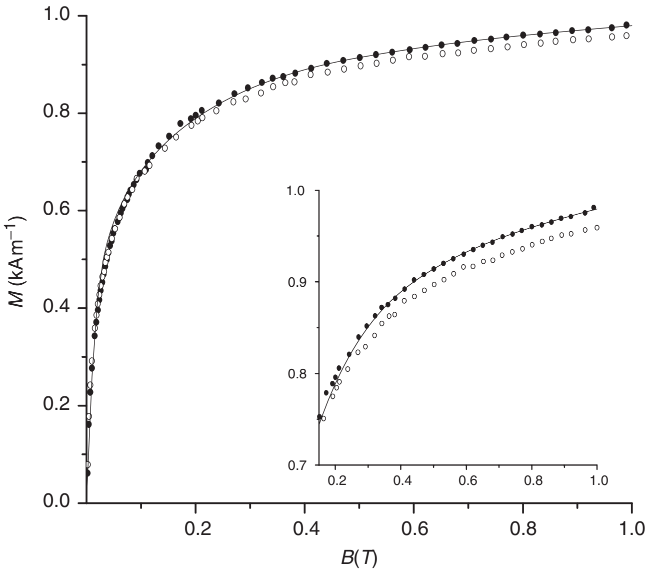
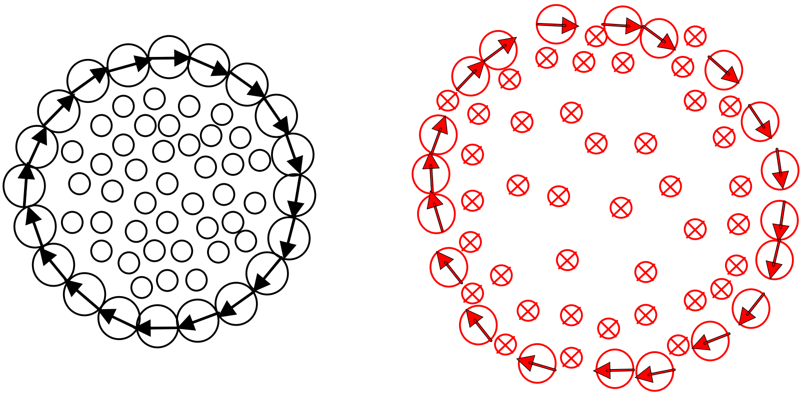


Figure 7. The comparison of both experimental data and fitted magnetisation curve of $M_f$ being amended for binary ferrofluids.

Notes: • • •Experimental date of binary ferrofluids. ——Binary magnetisation curve fitted by Equation (10) ($M_f = 0.97\,\mathrm{k\,A\,m^{-1}}$). ○○○○Summed date of both Figure 4(a) and (b).



Under zero magnetic field

Under magnetic field
(The directions of both the magnetic field and the induced moments perpendicular to the plane of the ring)

Figure 8. Schemetic drawing of the break of $CoFe_2O_4$ magnetic moment closed ring of self-assembly structure in the binary ferrofluids.

structure [18]. The particles and chains from the broken rings will contribute to the magnetisation of the ferrofluids, so the saturation magnetisation of $CoFe_2O_4$ ferrofluids component in the binary ferrofluids is enhanced. But, the broken rings may be only few since the $p$-$NiFe_2O_4$ particles may remove the plane of the rings from the affection of Brownian motion.

## 5. Conclusions

The inherent moments of ferrimagnetic $CoFe_2O_4$ particles are far larger than the induced-moments of paramagnetic $p$-$NiFe_2O_4$ particles in the present experiment. The $CoFe_2O_4$ particles would form field-induced aggregates which can result in the ferrofluids' magnetisation deviating from Langevin theory. The magnetisation of $CoFe_2O_4$ ferrofluids can be well described by MGC. The $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary ionic ferrofluids can be synthesised with both $CoFe_2O_4$ ferrofluids and $p$-$NiFe_2O_4$ paramagnetic fluids prepared by the Massart method. Since there is not enough magnetic interaction to aggregate between the $p$-$NiFe_2O_4$ particle and the $CoFe_2O_4$ particle, the magnetisation can be written as the sum of the magnetisation of both the $CoFe_2O_4$ ferrofluids component and the $p$-$NiFe_2O_4$ paramagnetic fluids component. But, the saturation magnetisation of the $CoFe_2O_4$ ferrofluids component in the binary ferrofluids is larger than the single $CoFe_2O_4$ ferrofluids. This could be because the ring-like aggregates of the $CoFe_2O_4$ ferrofluids' particles break, partially from the 'expansion' effect of the polarised $p$-$NiFe_2O_4$ particle gas into the rings during the magnetising process. For the magnetisation, the effect of field-induced aggregates from the $CoFe_2O_4$ ferrofluids' particles is the same between the binary ferrofluids and the single ferrofluids. In summary, in the present study, the microstructure transition of the $CoFe_2O_4$–$p$-$NiFe_2O_4$ binary ionic ferrofluids mainly results from the $CoFe_2O_4$ ferrofluids component, and the $p$-$NiFe_2O_4$ paramagnetic fluids component does not produce sensitive affection to the field-induced aggregates, but could do so to the self-assembled aggregates of the $CoFe_2O_4$ ferrofluids under zero field. This could be very important to investigate the physical behaviour, e.g., magneto-optical effect, for the binary ferrofluids. It needs to be pointed out that the microstructure of particular fluids is difficult to observe experimentally [19]. The electron microscope is a very useful tool for nanostructure observation, but, for the ferrofluids' structure investigation, only information about the size of separate particles can be obtained because the structure would undergo uncontrollable change while a sample was prepared for the examination [20]. For ferrofluids, especially binary ferrofluids, the microstructure as well as the relation between the microstructure and the physical behaviour should be further studied.

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