The liquid-solid change phase method for the experiment of inverse ferrofluids topology

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Abstract Inverse ferrofluids refers to ferrofluids containing micron grade nonmagnetic particles. The typical inverse ferrofluids include nanowires, MAGIC fluids, magnetic complex fluids, magnetorheological fluids and bubbles. We will give a new kind of experimental method, namely liquid-solid phase change method. The liquid containing magnetic particles stirred uniformly is poured into the funnel of the experimental device, which forms a natural flow under the action of its own gravity. After that, the whole experimental device was placed in a low temperature environment. When the temperature of ferrofluids was lower than the crystal temperature of paraffin wax, the flow process stopped naturally. The experimental method is mainly through the experimental observation of the fluid dynamic behavior after the low transparency liquid or the opaque milky liquid with the addition of micron particles, which can overcome the limitation of the PIV, which can only observe the hydrodynamic behavior of particles added to the transparent liquid.

1. Introduction
Ferrofluids refers to a magnetic suspension materials, which have been studied for nearly 50 years[1-3]. It is formed by ferromagnetic particles (FPs) about 10 nm in size dispersed in a suitable base liquid, a Since the surface of FPs can be adsorbed by surfactant molecule about 2 nm in size, the ferrofluid is stable. Compared with general fluids, ferrofluids not only have liquid fluidity, but also have magnetization properties. We can control the movement of ferrofluids by applying an external magnetic field. Although the micro structure, particle interaction and phase behavior of ferrofluids are still controversial, many macroscopic properties have been widely used. [4-5].

Ferrofluids containing micron-sized non-magnetic particles are called inverse ferrofluids [6-8]. Skjeltorp [6] added a non-magnetic colloid into a ferrofluid and then mixed it to get an inverse ferrofluid in 1983. Magnetic holes refers to the particles added to inverse ferrofluids. The typical inverse ferrofluids include nano-wires (NWs) [9], MAGIC (MAG-netic Intelligent Compound) fluids [10], magnetic compound fluids [11], magnetorheological fluids and bubbles containing ferrofluids. The magnetization curve remains overlapping and has no microstructure change on the magnetic phase after the non-magnetic particles are dispersed.

Because the micron-sized non-magnetic particles is much larger than the nano-sized magnetic particles in the inverse ferrofluids, the interaction between the non-magnetic particles is mainly the magnetic dipole moment, therefore the van der Waals force is not considered. It indicate that when a large amount of non-magnetic particles are placed in ferrofluids, the particles in an external magnetic field exhibit anisotropy under the action of a dipole force, and are assembled into a chain in the direction of the magnetic field[6-9]. When micron-sized organic solvent particles are placed in
ferrofluids, magnetic holes are forming some tree and circle structures under the action of a magnetic field. Since the macroscopic properties of inverse ferrofluids are affected by nonmagnetic particles, the special structural properties of non-magnetic particles have important applications value in the fields of engineering thermal physics, chemical engineering and bioengineering [9, 12].

Researchers have studied the complex geometry of non-magnetic particles through experimental simulations and theoretical analysis. In 1976, Kaiser[13] first found that in the inverse ferrofluids, micron-sized non-magnetic particles generated mutual attraction in the direction of the magnetic field and mutual exclusive in the vertical direction of the magnetic field. In 1987, Fujita [14] verified the Kaiser phenomenon through experimental measurements and dipole force model calculations. Zhu[10] performed Stokesian kinetics simulation of experiment by adding micron-sized circular magnetic particles and circular non-magnetic particles to magnetic fluids. It showed that non-magnetic particles can be driven by magnetic particles and eventually dispersed in the voids of corps of magnetic particles, moreover the manipulation of non-magnetic particles can be achieved in this way. Then Ido[15] used the same method to mix different proportions of spherical particles and capsule-shaped magnetic and non-magnetic particles to study the self-assembled morphology respectively. He found that the non-magnetic particles also slightly self-assembled along the lines of magnetic field. Peng[16] used two-dimensional Monte Carlo method to simulation of magnetic particles and non-magnetic particles suspended in ferrofluid under an uniform magnetic field. They found that the properties of the particles have more influence on the movement of the particles than the external magnetic field. Recently, Iwamoto[9] found that some changes in thermal conductivity can be made by adding silver nano-wires to the magnetic fluid. By using silver nano-wires in magnetic fluids, it expressed that thermal conductivity reduced in the direction parallel to the direction of thermal gradient but increased in the vertical direction of the magnetic field. Visualization of bubble behavior in inverse ferrofluids is one of the important issues of inverse ferrofluids in scientific and engineering applications. Ueno and Korlie [17-18] used the VOF method to calculate the bubble behavior in inverse ferrofluids. Yamasaki[19] has studied the bubble deformation of inverse ferrofluids by the combination of the Volume of Fluid (VOF) method and the lattice Boltzmann (LBM) method.

The above works are studied in the macroscopic view. In the context of continuum mechanics, the mechanical behavior of non-magnetic particles is mainly observed through experiments. Non-magnetic particles (such as bubbles, two-ball settlement, etc.) and fluid interactions are no longer scaled separately. Numerical simulation and various algorithms are usually used on the macro scale to couple level set method, the immersion boundary method and the elastic collision model, respectively. In fact, three-dimensional problems of ferrofluids need to be considered.

The macro scales are covered by macroscopic physical quantity[1-2]. The collision and stress are not affected by the macroscopic model between the fluid and the ferromagnetic particles, between the fluid and the non-magnetic particles, and between the ferromagnetic particles and the non-magnetic particles.

The nano-space scale with some nano-particles. In the beginning the ferrofluids statics theory was analyzed by the stresses of ferromagnetic particles, such as gravity, dipole interaction force and van der Waals force [4-5]. When the ferrofluids achieve statistical equilibrium, ferromagnetics nano-particles has usually only 2-15 nm, and ferromagnetics particles are coated with a 2 nm dispersant.

Subsurface space is a micron-scale nonmagnetic particle[20-22]. Compared with nano ferromagnetic particles, there is a strong interaction between the non-magnetic particles. The scale effect also appears between the base carrier fluid and the ferromagnetic particles. Therefore, the inverse ferrofluids is no longer simply regarded as a continuous medium.

2. Experiment of the magnetic field affecting the distribution of ferroferric oxide in liquid paraffin

2.1 Experimental apparatus
As in Fig.1, the experiment apparatus is mainly divided into three parts. 1, mixer, it includes ultrasonic mixer and normal mixer; 2, external magnetic field, where a permanent magnet is generally used with magnetic field intensity of 400-1000 Gauss; 3, thermostat.

![Figure 1. the experimental apparatus](image)

In figure1, (a) is conventional mixer, (b) is external magnet, and (c) is thermostat.

2.2 Experimental process
The ferroferric oxide particles ($\text{Fe}_2\text{O}_3$) were placed in liquid paraffin, stirred in an ultrasonic stirrer for 10 minutes, and then ferroferric oxide particles were dispersed to form inverse ferrofluids to prevent particle agglomeration rapidly. After that, the inverse ferrofluids is placed in a conventional agitator and stirred for 2 to 6 hours. It is obvious that the viscosity of the liquid paraffin itself is large, we suggested stirring for more than 3 hours.

We first pour the inverse ferrofluids into the funnel, and the inverse ferrofluids flow naturally under gravity. Then, refrigerate the entire experimental device, and the temperature of the inverse ferrofluids rapidly reduced to the paraffin curing temperature, the flow process naturally stopped. At this time, the pipe is filled with the solidified inverse ferrofluids.

Taking out the solid paraffin containing ferroferric oxide particles to slice and measure it, the process is shown as following:

Slice: We cut the pipe which is filled with solid paraffin containing ferroferric oxide particles symmetrically, remove the solid paraffin, and cut it into thin slices by ultra-thin blades. Therefore we obtain the production of five millimeter-thick paraffin sheets.

We first place the paraffin sections on the slide and cover it with coverslips, and then place the slices in a higher temperature environment to allow it soften naturally. As long as the paraffin section starts from white to light blue, the ambient temperature increased to make the paraffin slightly harden. In the last step we placed the paraffin sheet in a metallographic microscope for observation, and a final image was obtained. Usually, we refer to the above experimental observation method as liquid-solid phase method.

| Experimental Materials | Density (g/cm$^3$) | Softening temperature ($^\circ$C) | melting point ($^\circ$C) | transparency | Particulates' Diameter |
|------------------------|-------------------|----------------------------------|--------------------------|--------------|------------------------|
| paraffin               | 0.8 (liquid)      | 30~40                            | 56                       | Low transparency (liquid) |            |
| (a) $\text{Fe}_2\text{O}_3$ | 5.18              | /                                | 1594.5                   | Black and opaque | 6μm       |
| (b) $\text{Fe}_3\text{O}_4$ | 5.18              | /                                | 1594.5                   | Black and opaque | 20μm      |

2.3 Analysis of experimental results.
(1) Distribution of Fe3O4 particles with a diameter of 6 microns in paraffin
Fig. 2 and Fig. 3 both show the complex geometry of the particles in the hydrodynamic behavior of liquid paraffin respectively. Fig.2(a) and Fig.3(a) denote inverse ferrofluids paraffin liquid, we can also observe the ferromagnetic particles under the additional magnetic field. From Fig.2(a) and Fig.3(a), it can be seen that the ferromagnetic particles are in a disorderly distribution without an external magnetic field. From Fig. 2(b) and Fig. 3(b), it can be seen that under the action of an external magnetic field, the ferromagnetic particles form a chain geometry during the flow. Especially if the ferromagnetic particles are 6μm, the phenomenon is particularly remarkable.

3. Conclusion
The liquid-solid phase change method is a feasible and effective method, which is summarized through the process of experimental research. This method can help us to effectively observe the hydrodynamic behavior of very low transparency or opaque liquids after adding the particles. In general, people can only observe the hydrodynamic behavior of liquids with higher transparency by PIV. Experimental studies in this paper show that the solid-liquid phase-change method can effectively overcome this difficulty.

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