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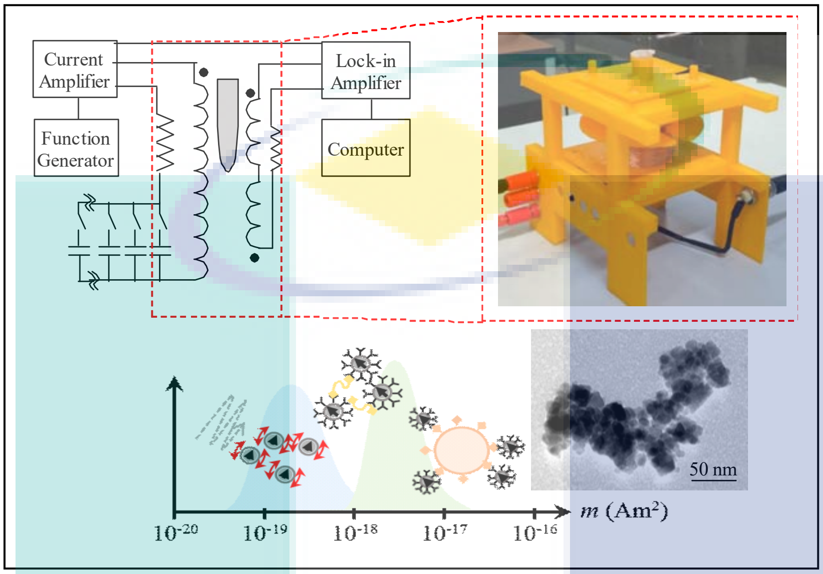

INVESTIGATING THE MAGNETIC FLUID CHARACTERISTICS IN LOW CONCENTRATION SOLUTIONS TO ENHANCE HARMONICS GENERATION

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ABSTRACT (120 words)

The generation of harmonics during application of AC and DC magnetic fields to multi-core iron oxide particles in low concentration solutions was studied based on magnetization curve and harmonics response of the nanoparticles in low concentration solutions. As a first step, the distribution of particle size was analyzed using a Non-Negative Inversion Method to investigate the relation between particle size and diamagnetic contribution of liquid carrier. The harmonics of nanoparticle solutions was evaluated using a highly sensitive AC susceptometer, which was developed during this research period. Based on harmonics response, distribution of particle size, and the derived models of AC susceptibility response, the fundamental harmonic generation was affected by the diamagnetic contribution when the iron concentration was lower than 6 microg/ml..

1. INTRODUCTION

Utilization of magnetic nanoparticles (MNPs), e.g., multi-core iron oxide nanoparticles, in bio-medical applications such as magnetic immunoassay and in-vivo imaging using magnetic particle imaging (MPI) technique, is greatly expected since their size is comparable to biological entities. Furthermore, their detection can be achieved via fast and non-invasive techniques. Due to these reasons, MNPs are expected to play important roles in many fields in future, thus, clarification of their properties is crucial for their targeted applications. In order to tailor MNPs for their specific applications, clarification on their magnetic characteristics and harmonics generation in diamagnetic liquid carrier is crucial. In many characterizations of MNPs, the diamagnetic contribution of liquid carrier is often disregard because it is constant and small. However, the diamagnetic contribution tends to deform the magnetization curve of low concentration MNP solutions. This deformation has been shown in references [1]–[3], thus it may affect the observed harmonics of MNP solutions. The objectives of this research are to investigate magnetic characteristics of MNPs in low concentration solutions and improve their harmonics generation. Commercial multi-core iron oxide nanoparticles are investigated so that comparison can be made with other studies. The AC and DC magnetizations of the nanoparticles in low concentration solutions is studied with respect to the magnetic moment distributions and the environmental factors. The harmonics generation and AC susceptibility models based on consideration to diamagnetic background, magnetization direction and magnetic anisotropy axis of MNP solutions will be developed so that MNPs can be optimized for the designed applications.

2. RESEARCH METHODOLOGY

In this study, the methodology was divided into 5 stages.

1. Validation of magnetization curve deformation in MNP solutions: A multicore iron oxide nanoparticle manufactured by Micromod Partikeltechnologie GmbH (nanomag D-spio D100) was used to prepare in a set of low concentration solutions and its magnetization curve was measured by a sensitive high-Tc SQUID magnetometer.

2. Reconstruction of magnetic moment distribution: The distribution of magnetic moment/particle size was modelled by a summation of superparamagnetic Langevin equations with different magnetic moment and the distribution was reconstructed by solving the inverse problem of the Langevin equations of the measured magnetization curve. A non-regularized non-negative inversion method was successfully developed to reconstruct the distribution. The distribution obtained from this method was then compared with the optically measured data from Transmission Electron Microscopy measurement to find their agreement. These two initial steps were performed to achieve number 1 objective.

3. Measurement of harmonics response: In this step, the actual performance of harmonic responses of the MNPs will be measured using a specially developed AC magnetometer. As a result, a sensitive AC magnetometer was successfully developed, having capability to measure harmonics from 3 Hz to 30 kHz with an excitation field up to 20 mTpp. The harmonics from the iron oxide nanoparticles were measured down to a concentration of 30 nanogram/ml.

4. Numerical simulation of harmonic generation: From the obtained distribution of particle size, time waveforms of AC magnetization upon excitation of DC and AC magnetic fields were numerically simulated. Then, the harmonics were extracted by applying Fast Fourier Transform method to the time waveforms with respect to different AC and DC fields with a consideration on the diamagnetic background. The simulated harmonics were compared to experimental data to clarify the mechanism of harmonic generation and effect of the environmental factors. This step was performed to achieve number 2 objective.

5. Modelling of MNPs in solution and optimization of harmonics: Based on data obtained from above steps, a magnetization model of colloidal MNPs under consideration of diamagnetic background and other factor was proposed based on the modification of the Langevin equation. The model was tested to find agreement between simulated and measured harmonics with respect to particle size distribution, viscosity and magnetism of liquid carrier. Based on the proposed model, a recommendation was made to optimize harmonic generations with respect to the diamagnetic background of the liquid carrier.

3. LITERATURE REVIEW

The magnetism of MNPs is influenced by several factors such as size distribution of magnetic cores, temperature, morphology (shape anisotropy), intrinsic and extrinsic surface effects, surface spin disorder and magnetic interactions. Assessing the information on these parameters is important in order to optimize MNPs for the target applications where they can be detected through magnetic susceptibility [1], [2], [4]–[6], relaxation [7]–[9], and remanence [10], [11] methods. Instrumental techniques such as DC and AC susceptibilities, magnetorelaxometry, Mössbauer spectroscopy, ferromagnetic resonance, neutron diffraction, transmission electron microscopy (TEM), and photon correlation spectroscopy have been used to assess and characterize the magnetic properties of MNPs [9], [10], [12], [13]. In the applications where detection is performed using magnetic susceptibility method, e.g., magnetic immunoassay and Magnetic Particle Imaging, the dynamic magnetic response of MNPs under excitation of AC magnetic fields is measured to detect MNPs. Furthermore, the non-linear magnetic characteristic of MNPs is utilized to obtain high sensitivity detection under strong excitation magnetic field. The noise resulted from the excitation magnetic field can be isolated by detecting harmonics, thus promising highly sensitive detection for bio-medical applications. The harmonics of MNP system have been investigated with respect to particle types, AC and DC magnetic fields, liquid carrier viscosity, and Brownian and Neel relaxations [14], where in many cases the weak magnetic characteristic of liquid carrier is often ignored. The preliminary studies in references [1], [2], [15] showed that the diamagnetic signal from the liquid carrier deforms the magnetization curve of low-concentration MNPs solution, where this may affect the static and dynamic magnetic characteristics of MNPs in solution. This project is expected to extend these preliminary studies so that it can contribute to enhancement of harmonic generation.

4. FINDINGS

1. The static magnetization curve in saturation region is deformed at iron concentration lower than 72 microgram/ml. A diamagnetic characteristic of carrier liquid was observed in this region for multi core iron oxide nanoparticles having an average particle size of 11 nm.

2. A sensitive AC magnetometer based on induction coils was developed specially for characterization of magnetic fluid. A resonant coil technique using Litz wire and capacitor network was developed for the excitation coil with high homogeneity (95%) and efficiency (5.2 mT/A). A large number of turns of detection coil was design and fabricated using a 0.25-mm copper wire. A sensitivity of 5*10^-7 Am^2 was shown at 100 Hz.

3. Simulated and measured harmonics showed that the diamagnetic carrier liquid affects the first harmonic when the iron concentration is lower than 6 microg/ml. However, higher harmonic components are transparent to diamagnetic background down to 30 ng/ml for particles having a magnetic moment peak higher than 10^-19 Am^2.

4. Selecting magnetic nanoparticles having a magnetic moment peak larger than 10^-19 Am^2 will permit detection of them up to sub nanogram of iron contents.

5. The AC response of MNPs showed two types of relaxation process, namely, Neel and Brownian relaxations. The use of Brownian relaxation characteristics enabled us to determine the hydrodynamics size of the MNPs. This permits to a technique for a fast estimation of MNPs' size.

6. The AC response can be used to extract magnetic parameters such as anisotropy energy, relaxation process, viscosity of the liquid carrier etc. We successfully showed that the harmonics can provide a simple and fast method to evaluate the viscosity of the liquid carrier.

7. Harmonics are affected by the anisotropy energy, relaxation process, magnetic moments of the MNPs etc., these factors might be studied in the future works.

5. CONCLUSION

The study successfully clarified the magnetization curve deformation of the multi-core iron oxide nanoparticles in low concentration solutions due to the diamagnetic contribution of the liquid carrier. An analysis method to estimate the core size of the nanoparticles was successfully developed and implemented based on DC magnetization characteristic, which will serve as a robust alternative method for magnetic nanoparticle standardization in future compared to the mainstream TEM imaging. From the proposed harmonic generation model and the measurement of harmonics intensity, it was found that the diamagnetic contribution affected the fundamental harmonic when the concentration and magnetic moment distribution peak were lower than 6 microgram/ml and 10^-19 Am^2, respectively. The harmonics could be used to determine other magnetic properties such as magnetic anisotropy, relaxations and hydrodynamic sizes. The study also successfully developed a sensitive AC magnetometer using inductive coils, in which established know-hows of its instrumentation technique and could be implemented for other magnetic property’s investigation in future.

ACHIEVEMENT

i) Name of articles/ manuscripts/ books published

Mohd Mawardi Saari, Nazatul Shareena Suhaimi, Mohd Herwan Sulaiman, Nurul Akmal Che Lah, Kenji Sakai, Toshihiko Kiwa, Keiji Tsukada: Influence of Viscosity on Dynamic Magnetization of Thermally Blocked Iron Oxide Nanoparticles Characterized by a Sensitive AC Magnetometer. Journal of Superconductivity and Novel Magnetism 02/2019;, DOI:10.1007/s10948-019-5031-6

Mohd Mawardi Saari, Nurul Akmal Che Lah, Kenji Sakai, Toshihiko Kiwa, Keiji Tsukada: Harmonics Distribution of Iron Oxide Nanoparticles Solutions under Diamagnetic Background. Journal of Magnetism and Magnetic Materials 12/2017; 452., DOI:10.1016/j.jmmm.2017.12.054

Mohd Mawardi Saari, Kenji Sakai, Toshihiko Kiwa, Keiji Tsukada, Nazatul Shareena Suhaimi, Nurul Akmal Che Lah: A Sensitive AC Magnetometer using A Resonant Excitation Coil for Magnetic Fluid Characterization in Nonlinear Magnetization Region. 2018 IEEE International Magnetic Conference (INTERMAG), Singapore; 11/2018, DOI:10.1109/INTMAG.2018.8508784

Mohd Mawardi Saari, Sharreena Nazatul, Saifudin Razali, Kenji Sakai, Toshihiko Kiwa, Keiji Tsukada, Nurul Akmal Che Lah: Development of A Resonant Excitation Coil of AC Magnetometer for Evaluation of Magnetic Fluid. Journal of Telecommunication, Electronic and Computer Engineering 2/2017; Vol. 10, 1-2.

Mohd Mawardi Saari, Mohd Aufa Hadi Putera Zaini, Hamzah Ahmad, Nurul Akmal Che Lah: An AC Magnetometer using Automatic Frequency Switching of a Resonant Excitation Coil for Magnetic Nanoparticles Characterization. 2018 9th IEEE Control and System Graduate Research Colloquium (ICSGRC); 08/2018, DOI:10.1109/ICSGRC.2018.8657632

Mohd Mawardi Saari, Nurul Akmal Che Lah, amir izzani mohamed, Mahendran Samykano, Nazatul Sharreena Suhaimi: Development of a compact and sensitive AC magnetometer for evaluation of magnetic nanoparticles solution. Control and System Graduate Research Colloquium (ICSGRC), 2017 IEEE 8th; 08/2017, DOI:10.1109/ICSGRC.2017.8070599

Nazatul Sharreena Suhaimi, Mohd Mawardi Saari, Hamzah Ahmad, Mohd Rusllim Mohamed, Nurul Akmal Che Lah: A Resonant Type AC Magnetometer for Evaluation of Magnetic Nanoparticles. Intelligent Manufacturing & Mechatronics, 01/2018: pages 93-99; , ISBN: 978-981-10-8787-5, DOI:10.1007/978-981-10-8788-2_9

Nurul A’in Nadzri, Chai Kar Hoe, Mohd Mawardi Saari, Saifuddin Razali, Mohd Razali Daud, Hamzah Ahmad: Vehicle Detection System Using Tunnel Magnetoresistance Sensor. Intelligent Manufacturing & Mechatronics, 01/2018: pages 547-555; , ISBN: 978-981-10-8787-5, DOI:10.1007/978-981-10-8788-2_49

Nurul A’in Nadzri, Mohd Mawardi Saari, Saifuddin Razali, Mohd Rusllim Mohamed, Hamzah Ahmad: Detection of Metallic Contaminant in Aluminium Soda Can Using

TMR Sensor. Intelligent Manufacturing & Mechatronics, 01/2018: pages 527-535; , ISBN: 978-981-10-8787-5, DOI:10.1007/978-981-10-8788-2_47

ii) Title of Paper presentations (international/ local)

Influence of Viscosity on Dynamic Magnetization of Thermally Blocked Iron Oxide Nanoparticles Characterized by a Sensitive AC Magnetometer, ICSM 2018, Turkey

A Sensitive AC Magnetometer using A Resonant Excitation Coil for Magnetic Fluid Characterization in Nonlinear Magnetization Region, INTERMAG 2018, Singapore

An AC Magnetometer using Automatic Frequency Switching of a Resonant Excitation Coil for Magnetic Nanoparticles Characterization, ICSGRC 2018, Shah Alam

Development of a compact and sensitive AC magnetometer for evaluation of magnetic nanoparticles solution, ICSGRC 2017, Shah Alam

Development of A Resonant Excitation Coil of AC Magnetometer for Evaluation of Magnetic Fluid, InECCE 2017, Langkawi

A Resonant Type AC Magnetometer for Evaluation of Magnetic Nanoparticles, SYMPOSIMM 2018, Pekan

Vehicle Detection System Using Tunnel Magnetoresistance Sensor, SYMPOSIMM 2018, Pekan

Detection of Metallic Contaminant in Aluminium Soda Can Using TMR Sensor, SYMPOSIMM 2018, Pekan

iii) Human Capital Development
Nazatul Sharreena Bt Suhaimi 920601-02-5900, Master of Science, Graduated May 2019

iv) Awards/ Others
Best Paper Award: Development of A Resonant Excitation Coil of AC Magnetometer for Evaluation of Magnetic Fluid, InECCE 2017, Langkawi

v) Others
Invitation to Okayama University, Japan for Japan-Asia Youth Exchange Program: Research Exchange on Magnetic Immunoassay using Magnetic Nanoparticles, funded by Japan Science & Technology Agency, July 2018

REFERENCES

[1] M. M. Saari, K. Sakai, T. Kiwa, T. Sasayama, T. Yoshida, and K. Tsukada, “Characterization of the magnetic moment distribution in low-concentration solutions of iron oxide nanoparticles by a high- T c superconducting quantum interference device magnetometer,” *J. Appl. Phys.*, vol. 117, no. 17, p. 17B321,

May 2015.
[2] M. M. Saari *et al.*, “Effect of diamagnetic contribution of water on harmonics distribution in a dilute solution of iron oxide nanoparticles measured using high-Tc SQUID magnetometer,” *J. Magn. Magn. Mater.*, vol. 394, pp. 260–265, Nov. 2015.
[3] M. M. Saari, N. A. Che Lah, K. Sakai, T. Kiwa, and K. Tsukada, “Harmonics distribution of iron oxide nanoparticles solutions under diamagnetic background,” *J. Magn. Magn. Mater.*, vol. 452, 2018.
[4] V. L. Calero-DdelC, D. I. Santiago-Quiñonez, and C. Rinaldi, “Quantitative nanoscale viscosity measurements using magnetic nanoparticles and SQUID AC susceptibility measurements,” *Soft Matter*, vol. 7, no. 9, p. 4497, 2011.
[5] F. Oisjöen, J. F. Schneiderman, A. P. Astalan, A. Kalabukhov, C. Johansson, and D. Winkler, “A new approach for bioassays based on frequency- and time-domain measurements of magnetic nanoparticles.,” *Biosens. Bioelectron.*, vol. 25, no. 5, pp. 1008–13, Jan. 2010.
[6] D. B. Reeves and J. B. Weaver, “Combined Néel and Brown rotational Langevin dynamics in magnetic particle imaging, sensing, and therapy,” *Appl. Phys. Lett.*, vol. 107, no. 22, 2015.
[7] T. Yoshida, K. Ogawa, T. Tsubaki, N. B. Othman, and K. Enpuku, “Detection of Magnetic Nanoparticles Using the Second-Harmonic Signal,” *IEEE Trans. Magn.*, vol. 47, no. 10, pp. 2863–2866, Oct. 2011.
[8] N. L. Adolphi *et al.*, “Characterization of magnetite nanoparticles for SQUID-relaxometry and magnetic needle biopsy,” *J. Magn. Magn. Mater.*, vol. 321, no. 10, pp. 1459–1464, May 2009.
[9] T. Yoshida, S. Bai, A. Hirokawa, K. Tanabe, and K. Enpuku, “Effect of viscosity on harmonic signals from magnetic fluid,” *J. Magn. Magn. Mater.*, vol. 380, pp. 105–110, 2015.
[10] N. Thanh, *Magnetic nanoparticles: from fabrication to clinical applications*. CRC Press, 2012.
[11] N. L. Adolphi *et al.*, “Characterization of single-core magnetite nanoparticles for magnetic imaging by SQUID relaxometry.,” *Phys. Med. Biol.*, vol. 55, no. 19, pp. 5985–6003, Oct. 2010.
[12] J. B. Weaver, A. M. Rauwerdink, C. R. Sullivan, and I. Baker, “Frequency distribution of the nanoparticle magnetization in the presence of a static as well as a harmonic magnetic field,” *Med. Phys.*, vol. 35, no. 5, p. 1988, 2008.
[13] J. Wells *et al.*, “Standardisation of magnetic nanoparticles in liquid suspension,” *J. Phys. D. Appl. Phys.*, vol. 50, no. 38, p. 383003, Sep. 2017.
[14] M. Graeser *et al.*, “Towards Picogram Detection of Superparamagnetic Iron-Oxide Particles Using a Gradiometric Receive Coil,” *Sci. Rep.*, vol. 7, no. 1, p. 6872, 2017.
[15] T. Mizoguchi *et al.*, “Highly sensitive third-harmonic detection method of magnetic nanoparticles using an AC susceptibility measurement system for liquid-phase assay,” *IEEE Trans. Appl. Supercond.*, vol. 26, no. 5, pp. 1–4, 2016.

APPENDIXES

Japan-Asia Youth Exchange Program: Research Exchange on Magnetic Immunoassay using Magnetic Nanoparticles
https://ssp.jst.go.jp/EN/report2018/vol12.html