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論 文 要 旨
THESIS SUMMARY

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要旨（英文 300 語程度）
Thesis Summary (approx.300 English Words)

Introduction

Superparamagnetic Iron Oxide Nanoparticles (SPIONs) are pivotal in biomedical applications, particularly in biosensing, due to their unique magnetic properties and modifiable surface chemistry. However, their behavior under varying physiological conditions, influenced by interparticle and inter-cluster interactions, remains inadequately understood. This study aims to elucidate these interactions, focusing on the effects of different ionic environments and incubation times on the dynamic magnetic susceptibility (DMS) of citric acid-coated SPIONs (CA-SPIONs). Understanding these factors is crucial for optimizing SPIONs in biosensing technologies, where stability and precise control over magnetic behavior are essential.

Method

CA-SPIONs were synthesized and surface-modified to enhance their stability in aqueous environments. The characterization of these nanoparticles was carried out using a combination of analytical techniques. Dynamic light scattering (DLS) was employed to measure the hydrodynamic size distribution, while zeta potential analysis provided insights into the surface charge and stability of the nanoparticles. Dynamic magnetic susceptibility (DMS) analysis was utilized to investigate the magnetic properties of the CA-SPIONs under various conditions. Additionally, transmission electron microscopy (TEM) was conducted to obtain detailed morphological images, offering a visual confirmation of the particle size and structure. The study systematically varied incubation times, ionic strengths, and buffer solutions to evaluate their effects on nanoparticle stability and magnetization dynamics. Precise control over incubation times allowed for an in-depth assessment of how these variables influenced the aggregation behavior and magnetic properties of the SPIONs in different ionic environments.

Results and Discussion

Interparticle and Inter-Cluster Interactions:

The results indicate that interparticle and inter-cluster dipole-dipole interactions play a critical role in determining the magnetic behavior and stability of CA-SPIONs. These interactions were found to induce significant changes in nanoparticle aggregation, which were captured in a two-step process observed in the DMS spectra. The first step involves signal broadening, which is indicative of initial aggregation where nanoparticles begin to interact more closely, yet remain relatively dispersed. This broadening suggests the onset of interparticle interactions. The second step is marked by peak shifting in the DMS spectra, occurring at higher ionic strengths and prolonged incubation times. This shift reflects a more substantial level of aggregation, as confirmed by TEM images, which show increased clustering of SPIONs. The changes in magnetization dynamics associated with these shifts underscore the importance of controlling these interactions to optimize the performance of CA-SPIONs in biomedical applications.

Quantification of Dipolar Interactions:

Interparticle and inter-cluster dipolar interactions were quantified by calculating the broadness of the DMS signal. This broadness was determined by the ratio between the peak frequency (f_peak) and the frequency at which the normalized imaginary part of DMS equals 0.5 (f_half) in the lower frequency region. This method provided a straightforward yet effective way to quantify the extent of aggregation, with greater broadening indicating stronger interactions and more significant clustering of SPIONs.

Order of Influence on DMS:

A comparative analysis established a hierarchy of influence among different ionic environments on

the DMS of CA-SPIONs:

1. **Potassium ions (K+):** Exhibit the strongest influence, causing significant peak shifts at lower concentrations, indicative of substantial nanoparticle aggregation.
2. **Sodium ions (Na+):** Also induce peak shifts and broadening in DMS spectra but require higher concentrations compared to K+ to achieve similar effects.
3. **Phosphate-buffered saline (PBS):** Primarily causes broadening in DMS spectra without significant peak shifts, suggesting a stabilizing effect due to phosphate ions.
4. **HEPES buffer:** Exhibits the weakest influence on DMS, with minimal broadening and peak shifts, indicating minimal destabilization of CA-SPIONs.

Magnetic Dipole Interactions:

The analysis of magnetic dipole interactions within SPION clusters reveals their critical role in determining dynamic behavior. These interactions become more pronounced with extended incubation times and higher ionic strengths, underscoring the need for precise control to optimize SPION performance in biosensing applications.

Conclusions

This study concludes that the stability and magnetic properties of CA-SPIONs are significantly influenced by interparticle interactions, ionic strength, buffer composition, and incubation time. The findings highlight the importance of understanding these factors to develop highly effective SPION-based biosensors, particularly in applications requiring high sensitivity and specificity.

Future Perspectives

Future research should explore the long-term stability of CA-SPIONs under varying physiological conditions to further optimize their application in biosensing. Investigating the combined effects of incubation time and complex biological environments on SPION behavior will be essential for advancing their use in medical diagnostics.

Novel Contributions

This dissertation makes several key contributions:

- It provides a comprehensive analysis of interparticle and inter-cluster dipole-dipole interactions on the dynamic behavior of CA-SPIONs, addressing a gap in the literature.
- The study introduces a novel, simplified approach to DMS analysis by focusing on a narrower frequency range, enhancing the practicality of the findings.
- Establishing a hierarchy of ionic influences on DMS, combined with the effect of incubation time, offers critical insights for optimizing SPION stability and functionality in various physiological conditions.

These contributions are expected to significantly advance the design and application of SPION-based biosensors in medical diagnostics, where precision in biomolecule detection is essential.