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Self-consistent magnetic properties of magnetite tracers optimized for magnetic particle imaging measured by ac susceptometry, magnetorelaxometry and magnetic particle spectroscopy

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ABSTRACT

Sensitivity and spatial resolution in magnetic particle imaging are affected by magnetic properties of the nanoparticle tracers used during imaging. Here, we have carried out a comprehensive magnetic characterization of single-core iron oxide nanoparticles that were designed for MPI. We used ac susceptometry, fluxgate magnetorelaxometry, and magnetic particle spectroscopy to evaluate the tracer's magnetic core size, hydrodynamic size, and magnetic anisotropy. Our results present a self-consistent set of magnetic and structural parameters for the tracers that is consistent with direct measurements of size using transmission electron microscopy and dynamic light scattering and that can be used to better understand their MPI performance.

1. Introduction

Both sensitivity and spatial resolution of magnetic particle imaging (MPI) – a new modality for the fast imaging of the spatial distribution of magnetic markers [1] – are critically determined by the availability of optimal magnetic nanoparticle tracers. So far, most of the MPI experiments utilize Resovist® – a clinically approved contrast agent originally developed for magnetic resonance imaging (MRI). In addition, MPI performance, comparable to that of Resovist, was demonstrated with other tracers optimized for MRI such as FeraSpin R™ from nanoPET Pharma GmbH [2]. It is, however, known that only a small fraction of the nanoparticles of Resovist and FeraSpin R contribute to the MPI signal. In fact, by using optimum fractions of the original suspensions of these MRI contrast agents, the MPI performance can be enhanced by about a factor of 2 as presented by Ludwig et al. [3] and Löwa et al. [4] (Table 1).

Whereas both Resovist and FeraSpin R particles are multicore ones, consisting of elementary crystallites with sizes between 5 and 7 nm, it has been shown that single-core iron-oxide nanoparticles with typical core diameters of (20–25) nm exhibit a MPI performance superior to that of Resovist and FeraSpin R. Moreover, both sensitivity and spatial resolution in MPI for single-core nanoparticle tracers is strongly coupled to nanoparticle size, with monosized dispersions providing superior performance [5–8]. In addition, use of a single-core particle tracer in MPI is also simpler to interpret with appropriate magnetization and relaxation models.

In this paper, we present a comprehensive magnetic characterization of these single-core nanoparticles for use in MPI. We include a self-consistent set of measurements of the effective magnetic anisotropy, K, which influences the magnetic reversal. Such measurements enable accurate assessment and improvement of MPI tracers and the magnetization models used to interpret MPI imaging. A deeper understanding of tracer behavior in dynamic magnetic fields is also critical to the continued development and optimization of MPI scanner design and related image reconstruction algorithms.

2. Samples

Oleic acid coated iron oxide nanoparticles (NPs) were synthesized by thermal decomposition of iron oleate in the presence of oleic acid, following a method reported earlier [9]. The hydrophobic NPs were then transferred to water using a co-polymer of poly(maleic anhydride-alt-1-octadecene) and polyethylene glycol (PMAO–PEG) [10]. The median core size of the NPs was calculated to be 20 nm, with a standard deviation of 0.245, by fitting $m(H)$ data to a log-normal distribution of particle sizes and assuming a

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Langevin response [11] (Fig. 1(a)). Magnetic size determined in this way matched with size measured by transmission electron microscope analysis (Fig. 1(b)). The hydrodynamic size of the polymer coated NPs in water was measured to be 62 nm (harmonic mean of the intensity distribution) with a polydispersity index of 0.195, using dynamic light scattering (DLS) (Fig. 1(c)).

Table 1
Nanoparticle characterization.

Method	μ_C [nm]	σ_C	μ_H [nm]	σ_H	M_s [A/m]	K [J/m^3]
ACMS	22.2*	0.2*	59	0.3	4.8×10^5*	3200
MRX	20	0.23	n/a	n/a	4.8×10^5*	6265
MPS	19	0.3*	59	0.3*	4.8×10^5*	6000*

* Indicates fixed parameter.

3. Characterizations

3.1. AC susceptibility

The complex ac susceptibility (ACS) measurement is a standard technique for the characterization of magnetic nanoparticles since it is rather simple and it covers a large range of relaxation times. ACS measurements for frequencies up to 10 MHz on suspensions of single-core magnetite particles which are similar to the ones studied here were recently presented [12]. In contrast to the model discussed below, the authors only consider a distribution of core diameters assuming a constant organic shell thickness. The ACS spectra discussed here were recorded with our 1 MHz ac susceptometer having a field amplitude of 95 $\mu T/\mu_0$ [13] and Fig. 2 shows the measured real and imaginary parts as a function of frequency. Clearly, the spectra cannot be modeled with a simple Debye model. To fit the measured spectra, we applied a generalization of the Debye model as

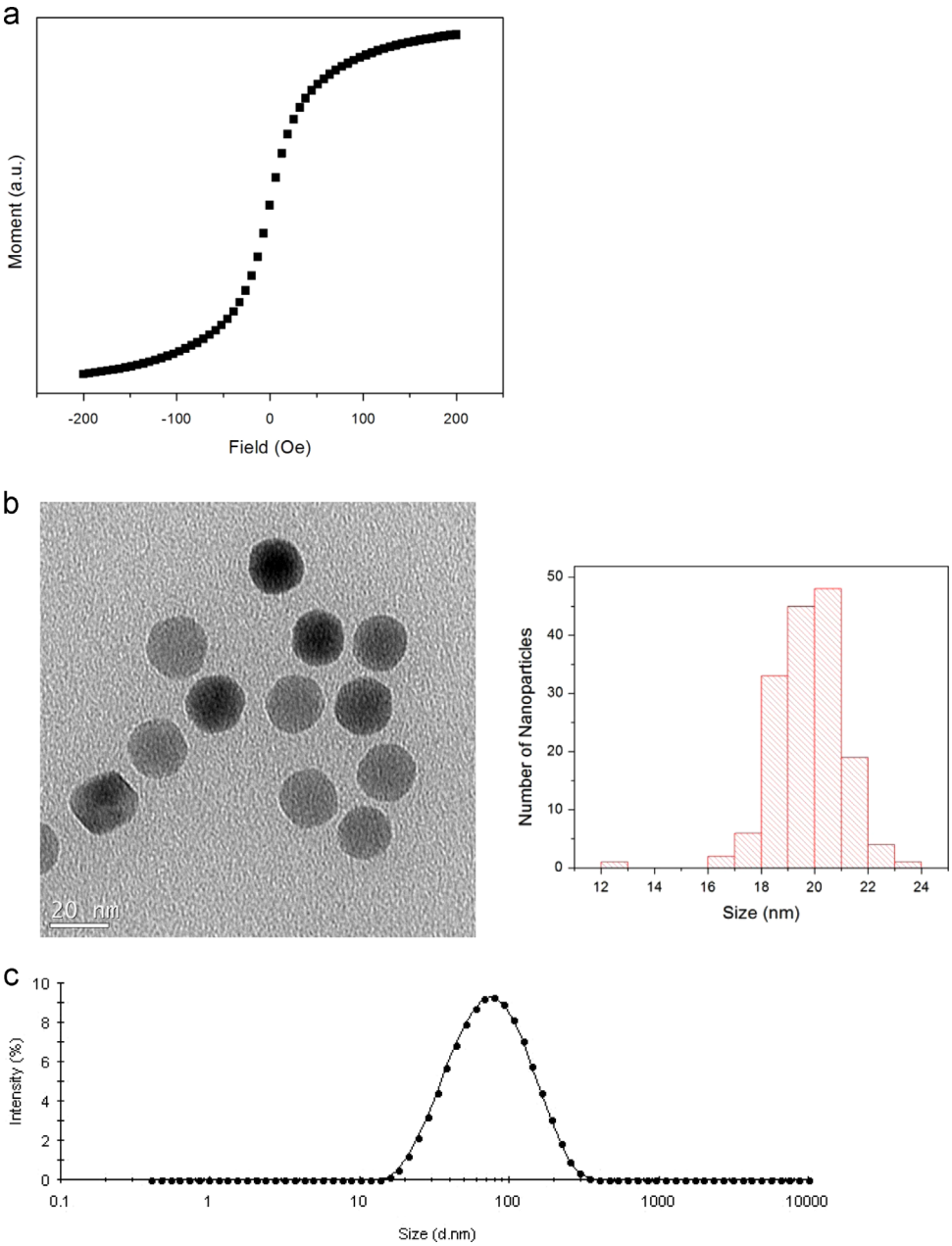

Fig. 1. (a) The $m(H)$ curve of the NPs after their phase transfer to water. (b) The TEM image and size distribution of the NPs. (c) The intensity-weighted hydrodynamic size distribution of the NPs.