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Role of coating agent in iron oxide nanoparticle formation in an aqueous dispersion: Experiments and simulation

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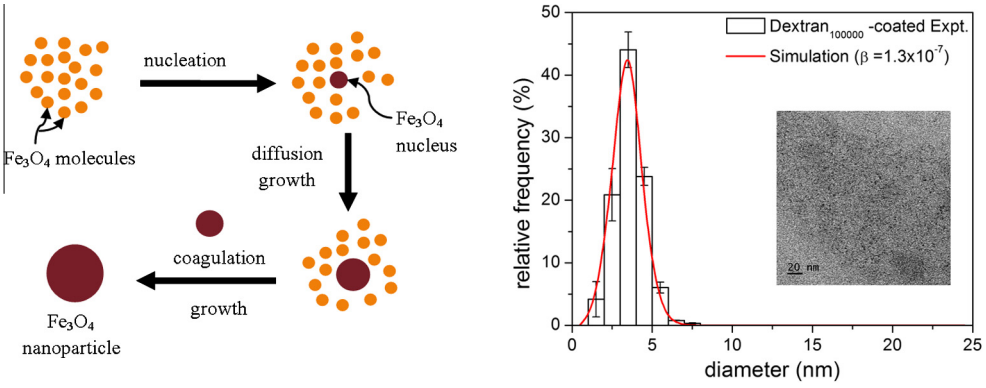
GRAPHICAL ABSTRACT



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ABSTRACT

Iron oxide (Fe_3O_4) nanoparticle was synthesized by coprecipitation and was modeled and solved using a hybrid (discrete–continuous) model, based on a kinetic Monte Carlo (kMC) simulation scheme. The latter was combined with the constant number MC method, to improve both speed and accuracy of the simulation. Complete particle size distribution (PSD) from simulation matches very well with PSD of both uncoated and coated (with either polyacrylic acid or dextran) Fe_3O_4 nanoparticles, obtained from our experiments. The model is general, as the time scales of various processes (nucleation, diffusion-growth and coagulation-growth) are incorporated in rate equations, while, input simulation parameters are experimentally measured quantities. With the help of the validated model, effect of coating agent on coagulation-growth was estimated by a single, fitted, *coagulation-efficiency* parameter. Our simulation shows that, logarithm of coagulation-efficiency scales linearly with logarithm of inverse of the molecular weight of the coating agent. With this scaling law, our model is able to a priori predict the experimental PSD of Fe_3O_4 nanoparticles, synthesized with an even higher molecular weight of dextran.

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1. Introduction

Iron oxide, specifically magnetite (Fe_3O_4), shows superparamagnetism in the size-range <20 nm. This property of Fe_3O_4 can be used in many biomedical applications like, magnetic resonance imaging (MRI) [1], hyperthermia and magnetic cell sorting [2]. Fe_3O_4 nanoparticles in this size range can be made by various different processes, like chemical coprecipitation [3], thermal decomposition [4] and microemulsion [5]. Each individual, separate nanoparticle in dispersion is referred to as the primary particle. Depending on the presence or absence of a coating agent, primary

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Nomenclature

English symbols

A	pre-exponential factor for nucleation, $m^{-3}\ s^{-1}$
$C(l)$	concentration of Fe_3O_4 molecule, at $t = 0$, mol m^{-3}
$C_i(l)$	concentration of Fe_3O_4 molecule, at the solid particle and liquid interface, mol m^{-3}
D_m	diffusivity of Fe_3O_4 molecule in water, $m^2\ s^{-1}$
$D_{p,P2.1k}$	diffusivity of PAA ($M_w \sim 2100$ g mol^{-1}) molecule in water, $m^2\ s^{-1}$
$D_{p,P5.1k}$	diffusivity of PAA ($M_w \sim 5100$ g mol^{-1}) molecule in water, $m^2\ s^{-1}$
$D_{p,D40k}$	diffusivity of dextran ($M_w \sim 40{,}000$ g mol^{-1}) molecule in water, $m^2\ s^{-1}$
$D_{p,D60k}$	diffusivity of dextran ($M_w \sim 60{,}000$ g mol^{-1}) molecule in water, $m^2\ s^{-1}$
$D_{p,D100k}$	diffusivity of dextran ($M_w \sim 1{,}00{,}000$ g mol^{-1}) molecule in water, $m^2\ s^{-1}$
d	diameter of nanoparticle, m
$\bar{d}_p$	mean diameter of nanoparticle, m
$\bar{d}$	nondimensionalized diameter of any particle
f_c	coagulation frequency, s^{-1}
f_n	nucleation frequency, s^{-1}
f_t	$f_n + f_c$, total frequency of both nucleation and coagulation, s^{-1}
K	constant coagulation frequency, for constant coagulation regime, s^{-1}
k_B	Boltzmann constant, 1.38×10^{-23} J K^{-1}
l_c	number of Fe_3O_4 molecule in nuclei
M_w	molecular weight, g mol^{-1}
N	number density of particles, m^{-3}
N_0	initial number of particle at $t = 0$, for constant coagulation regime
N_A	Avogadro number, 6.023×10^{23} mol^{-1}
N_p	total number of solid Fe_3O_4 particles in the simulation box at the end of simulation
$N(t)$	total number of particles at any time t
n_i	number of particle constituted by i molecules at any time
$n_{m,0}$	initial number of Fe_3O_4 molecules (in liquid phase) in simulation box at $t = 0$
$n(v,t)$	number density of particles having volume v at time t, m^{-3}
$\bar{n}(\bar{v},\tau)$	nondimensionalize number density of particles having nondimensionalize, volume $\bar{v}$ at time τ
$\bar{n}(\bar{d},\tau)$	nondimensionalize number density of particles having nondimensionalize, diameter $\bar{d}$ at time τ
p_c	probability of coagulation
$p_i(t)$	probability of ith event, where i is nucleation or coagulation
$p(i)$	probability of finding a particles constituted by i molecules in a particle size distribution (PSD)
$p_t(i)$	probability of adsorption of one molecule onto a particles constituted by i molecules in a PSD
$p_q(i)$	probability of finding a particles, which will adsorb q_i molecules during τ_Q
$p_d(d)$	probability of finding a particles of diameter d in a PSD
q_p	Brownian collision frequency, $m^{-3}\ s^{-1}$
q_i	number of Fe_3O_4 molecules diffusing onto a solid Fe_3O_4 particle (constituted by i molecules) during τ_Q
$\langle R_g^2 \rangle^{1/2}$	root-mean square radius of gyration of polymer molecule to estimate its diffusivity
S	solubility of Fe_3O_4 in water, mol m^{-3}
T	temperature, K
t	time, s
U	uniformly distributed random variable in the range $[0,1)$
V	volume of liquid dispersion, m^3
V_m	volume of a Fe_3O_4 molecule, m^{-3}
$\bar{v}$	nondimensionalized volume of any particle
x	mean-square displacement of molecule in a given time

Greek symbols

β	coagulation efficiency
λ_l	degree of supersaturation
μ	viscosity, kg $m^{-1}\ s^{-1}$
σ	interfacial energy between Fe_3O_4 nanoparticle and water, J m^{-2}
τ	nondimensionalized time
τ_c	coagulation timescale, s
τ_n	nucleation timescale, s
τ_g	diffusion timescale of one molecule to cross the diffusion layer, s
τ_p	timescale of polymer adsorption, s
τ_Q	interval of quiescence, s

Abbreviations

D40k	dextran, $M_w = 40{,}000$ g mol^{-1}
D60k	dextran, $M_w = 60{,}000$ g mol^{-1}
D100k	dextran, $M_w = 1{,}00{,}000$ g mol^{-1}
kMC	kinetic Monte Carlo simulation
PBE	population balance equation
PSD	particle size distribution
PAA	polyacrylic acid
P2.1k	polyacrylic acid, $M_w = 2100$ g mol^{-1}
P5.1k	polyacrylic acid, $M_w = 5100$ g mol^{-1}
TEM	transmission electron microscope
U	uncoated nanoparticle

nanoparticles can aggregate to weak, secondary aggregate structures. It is therefore important to model particle size distribution (PSD) of the primary particle, for an in-depth understanding of the mechanism of the coprecipitation route and the role of the coating agent.

For using nanoparticle dispersions in real applications, control of mean diameter of the primary particle is very important, yet difficult to achieve. Not only that, stability of the colloidal dispersion of these nanoparticles (resulting from the issue of whether secondary aggregates form and if so, the aggregate-size) is also another important issue for in vivo use. Use of polymeric coating agent is one of the common ways to control mean particle diameter and the entire PSD of the primary particle. In this work, we show how to quantify the effect of different polymeric coating agents on primary particle growth and hence control the PSD. We do not model possibility of formation of secondary aggregates, which has been addressed by Kumar et al. [6].

In the bulk coprecipitation method, the complete range of PSD of uncoated, primary Fe_3O_4 nanoparticles obtained are approximately 3–20 nm. However, the hydrodynamic diameter of secondary aggregates comprised of primary particles (referred to as aggregate size and measured by dynamic light scattering technique), can be as large as 50–100 nm [7]. On addition of a coating agent, *e.g.*, polyacrylic acid (PAA), the diameter-range of primary particles goes down to 2–15 nm and the secondary aggregate size also reduces to 8–30 nm (since presence of polymeric coating

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