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Effect of nanomaterial and media physicochemical properties on nanomaterial aggregation kinetics

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ABSTRACT

Understanding nanomaterial (NM) stability is required for an adequate interpretation of ecotoxicological test outcomes, fate and behavior studies, to generate parameters (such as critical coagulation concentration, CCC; and attachment efficiency, α) for environmental fate models, and for comparison among different studies. Numerous studies measured CCC and α for different types of NMs with a major focus on investigating the effect of ionic strength, ion valency and natural organic matter, with fewer studies investigating the effect of NM and other medium properties. Consequently, wide discrepancies can be found in the literature among the reported CCC and α values, even for NMs of the same composition and properties. In this context, the aim of this review is to investigate the dependence of NM aggregation kinetic parameters (*e.g.* CCC and α) on NM and medium physicochemical properties and to rationalize the differences observed among different studies, where possible. We found that various material and medium physicochemical properties need to be considered to predict NM aggregation behavior. Some trends were observed and rationalized based on theoretical studies and data available in the literature. For charge stabilized NMs with constant zeta potential, NM stability (CCC) decreases with the increase in Hamaker constant, increase in NM size, increase in buffer (carbonate and phosphate) concentration, increase in temperature, and light irradiation. The CCC increase with counterion complexation. For sterically stabilized NMs, the CCC increases with the increased surface coverage by the capping agent molecules and completely coated NMs do not aggregate even in high ions strength medium (*e.g.* seawater). These results highlight the significant role of NM and medium properties in influencing the environmental stability and fate of NMs, and will help refine NM fate models and improve our understanding of NM uptake and toxicity.

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1. Introduction

The stability of engineered nanomaterials (NMs) has been studied over the past two decades, since the evolution of nanotechnology. However, the stability of colloidal dispersions has been studied for over a century (Verwey and Overbeek, 1948; Derjaguin and Landau, 1941). A substantial progress in the understanding of the stability phenomena has been made since the introduction of the DLVO theory, which attributed the interaction between two identical particles to van der Waals attraction and the electrical double layer repulsion (Verwey and Overbeek, 1948; Derjaguin and Landau, 1941). Additionally, numerous studies have shown the importance of other interaction forces such as steric forces, and hydration forces on NM aggregation and aggregation kinetics (Baalousha et al., 2011a).

Quantitative assessment of NM stability can be achieved by estimating the attachment efficiency (α) - the inverse of the stability ratio (W) - and the critical coagulation concentration (CCC), which is the minimum counterion concentration required to fully destabilize the dispersion (see details in Section 2) (Tadros, 2007). The aggregation kinetics parameters (α, W, and CCC) of a specific counterion can be calculated theoretically using the classical DLVO theory on the basis of a static force balance. The aggregation kinetic parameters also can be measured experimentally from a kinetic point of view by studying the process of colloidal aggregation (Liu et al., 2009a). This can be achieved by monitoring the growth in NM size, or the loss of UV–vis absorbance for plasmonic NMs (*e.g.* Au and Ag NMs) at early stage of aggregation (Baalousha et al., 2013).

Numerous aggregation kinetics' experimental studies have focused on investigating the effect of ionic strength, ion valency, pH and natural organic matter (NOM) concentration on NM kinetic stability (CCC and α) (Baalousha et al., 2013; Hotze et al., 2010; Zhou and Keller, 2010; Baalousha et al., 2008; Baalousha, 2009). However, wide discrepancies can be found in the literature among the reported CCC and α values. These discrepancies are likely to be attributed to differences in the experimental conditions among the different studies such as: **1)** method employed to investigate NM aggregation kinetics (*e.g.* DLS, and UV–vis) (Baalousha et al., 2013), **2)** physicochemical properties of the NMs (*e.g.* size, shape, surface coating, *etc.*) (Penners and Koopal, 1987; Zhou et al., 2013), and **3)** media chemistry (*e.g.* buffer type and concentration, and ligand type and concentration) (Stemig et al., 2014), and **4)** environmental factors (*e.g.* temperature, and irradiation) (Hotze et al., 2010; Zhou and Keller, 2010; Zhou et al., 2013).

There is currently a limited knowledge on the effect of intrinsic properties of NMs (*e.g.* particle size, morphology, crystal structure, surface heterogeneity, and dopants) and intrinsic properties of the medium (*e.g.* buffer type and concentration, interaction of NM with medium constituents, and dissolved oxygen) and environmental factors (*e.g.* light, and temperature) on NM aggregation (Hotze et al., 2010; Zhou and Keller, 2010; Zhou et al., 2013). For instance, recent studies reported contradictory results on the dependence of CCC on NM size including a decrease with the decrease in NM size (*e.g.* hematite (He et al., 2008), TiO_2 (Zhou et al., 2013)), an increase with the decrease in NM size (*e.g.* CdSe (Mulvihill et al., 2010)) or independence of CCC of NM size (*e.g.* Au and Ag NMs (Liu et al., 2012; Afshinnia et al., 2016a)). In addition, whereas some studies reported a linear correlation between the CCC and NM primary particle size (*e.g.* anatase TiO_2 (Zhou et al., 2013)), other found that the CCC correlated better with NM specific surface area (*e.g.* anatase TiO_2 (Zhou et al., 2013), CdSe (Mulvihill et al., 2010)). Other studies reported an important role of stabilizing agent (Mulvihill et al., 2010), impurities introduced during synthesis process (Liu et al., 2011), *etc.* on NM stability. These studies suggest that material properties such as particle size, capping ligand, and impurities are important parameters affecting NMs' aqueous stability. However, a systematic review on the role of NM, and medium intrinsic properties on NM aggregation kinetics is lacking.

Several studies also reported deviations in NM aggregation behavior from classical DLVO behavior. Such deviations have been attributed to a wide selection of causes including discreetness of surface charge (Schudel et al., 1997), steric and relaxation effects (Ortega-Vinuesa et al., 1996), the presence of nanobubbles on particles (Mahnke et al., 1999; Parker et al., 1994; Yakubov et al., 2000), and the inherent surface roughness of the particles themselves (Shulepov and Frens, 1996; Bhattacharjee et al., 1998; Kostoglou and Karabelas, 1995; Sun and Walz, 2001). The deviation of NM aggregation behavior from the classical DLVO theory and the corrections proposed in the literature to the DLVO theory is discussed where relevant in the manuscript.

In this context, this article aims to investigate the relationship between the CCC values and the parameters affecting NM aggregation behavior and kinetics, in particular NM and medium properties. This review focuses on studies reporting NM aggregation kinetics measurement and is organized as follows: **1)** a brief review of the key concepts in NM stability (*e.g.* DLVO theory, Schulze-Hardy rule, α and CCC), **2)** a brief discussion of the methods used to measure NM aggregation kinetics (*e.g.* DLS and UV–vis), **3)** a critical discussion on the factors affecting NM aggregation kinetics, together with a rationale for the variability in the reported CCC values for the same type of NM based on NM and media physicochemical properties, and **4)** critical discussion on deviations from DLVO theory due to NM specific properties, which is integrated throughout the manuscript.

Understanding of the fate and behavior of NMs in environmental and toxicological media is crucial to allow for comprehensive environmental risk assessment of NMs (Klaine et al., 2008). Aggregation is

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