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## Experimental investigation on the use of highly charged nanoparticles to improve the stability of weakly charged colloidal system

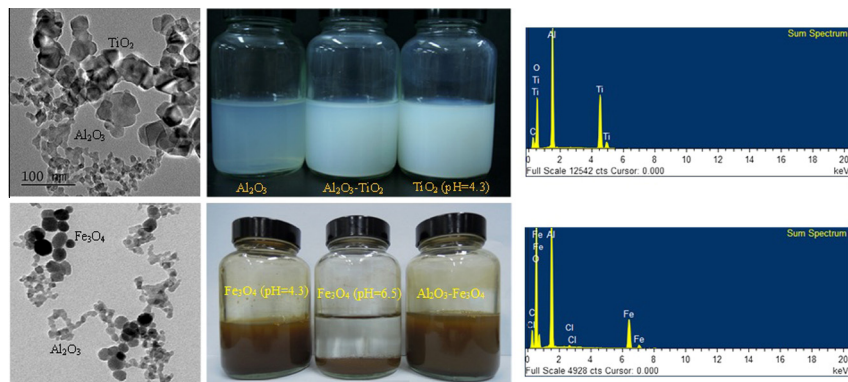


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### GRAPHICAL ABSTRACT



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### ABSTRACT

The present work highlighted on the implementation of a unique concept for stabilizing colloids at their incipiently low charge potential. A highly charged nanoparticle was introduced within a coagulated prone colloidal system, serving as stabilizer to resist otherwise rapid flocculation and sedimentation process. A low size asymmetry of nanoparticle/colloid serves as the new topic of investigation in addition to the well-established large size ratio nanoparticle/microparticle study. Highly charged Al<sub>2</sub>O<sub>3</sub> nanoparticles were used within the present research context to stabilize TiO<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub> based colloids via the formation of composite structures. It was believed, based on the experimental evidence, that Al<sub>2</sub>O<sub>3</sub> nanoparticle interact with the weakly charged TiO<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub> colloids within the binary system via absorption and/or haloing modes to increase the overall charge potential of the respective colloids, thus preventing further surface contact via van der Waal's attraction. Series of experimental results strongly suggest the presence of weakly charged colloids in the studied bimodal system where, in the absence of highly charged nanoparticle, experience rapid instability. Absorbance measurement indicated that the colloidal stability drops in accordance to the highly charged nanoparticle sedimentation rate, suggesting the dominant influence of nanoparticles to attain a well-dispersed binary system. Further, it was found that the level of colloidal stability was enhanced with increasing nanoparticle fraction within the mixture.

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Rheological observation revealed that each hybrid complexes demonstrated behavior reminiscent to water with negligible increase in viscosity which serves as highly favorable condition particularly in thermal transport applications.

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## 1. Introduction

It is evident that bottom-up engineering approach for fabrication and processing has grown tremendously since the inspiring lecture by Nobel laureate, Feynman [1] who highlighted on the possibility of manipulating molecules and atoms to gain unlimited authority over scale and functionality of the final product. As such, he also underlined the persisting challenges in working within sub-micron scale which include dealing with new jargon of physics such as gravity, surface tension and particle interaction, far dominant relative to macro-scale behavior. Thus, from the technological perspective, the key toward successful implementation of this concept strongly lies on the ability to produce and control the particles to achieve specific task.

Within the aspect of colloidal science, one of the underlying disciplines is associated to the study colloidal stability which plays a central role in numerous physicochemical processes. It is evident that the demand to attain highly dispersed colloidal suspension has spread to numerous technological applications ranging from coating, printing, drug carriers, catalysis, etc. [2–4]. A well-known approach to mitigate flocculation between suspended colloids is by tailoring the pH of the solvent away from the colloids isoelectric point to give columbic repulsive force as well as via steric route with the help dispersant and polymeric binder. Nanoparticles have also been used in unconventional way to stabilize colloids via electrostatic self-assembly mode [5,6].

The concept of using nanoparticles for colloidal stabilization has long been pursued for both emulsion and solid particles [7–9]. In solid particle, the nanoparticles absorbed onto the colloidal surface via charge asymmetry which promote better dispersion and less aggregation as highlighted earlier by Homola and co-workers [10]. The key toward successful application of nanoparticles in emulsions stabilization lies on the surface modification of the nanoparticles to contain both hydrophilic and hydrophobic segments which will interact at oil water interface, further preventing phase separation [11].

In recent years, hybridized colloids (i.e. colloidal system consisting of two or more particles of different type and morphology) have been used as additive in heat transfer media for thermal application [12,13]. The hybridization also leads to the formation of anisotropic colloidal structure which is favorable to suit specific application [14,15]. It was also proven that the addition of magnetic particles onto carbon nanotube via a simple two step mixing method with the help of surfactant enabled direct control of the thermal conductivity of the hybrid mixture in magnetic field environment [12,16].

While absorbing colloid–nanoparticle interaction has been widely studied, a new research paradigm on the subject of nanoparticle mediated colloidal assembly has been laid by Tohver et al. [17] which is based on weakly repulsive nature of colloidal interaction. Its fundamental theory lies on self-manipulating of highly charged nanoparticles on negligibly charged microparticles of the same polarity to form a halo structure. Nanoparticle haloing concept has been explored for various types of colloidal entities namely ceramic based microparticles, phospholipid, emulsions and carbon nanotubes [17–22]. Interestingly, recent work by Tian and Synder [23,24] highlighted the formation of thermodynamically stable silica nanoparticles close to their isoelectric point.

The stability of the colloid produced using a sol–gel method was attributed to the residual lysine linked to the silica surface.

Much works based on experimental [25–27], theoretical [28–30] and numerical [31–35] approaches have been pursued since the first reported finding on this newly exciting phenomenon. Several prominent progresses of research findings in nanoparticle stabilized colloids via haloing mechanism are worth mentioning here. Tohver et al. [17] elucidated on the ability of the stabilized binary system to generate a much ordered crystal structure under gravitational sedimentation colloidal assembly. Further, a dramatic transition of flow signature manifested from shear thinning associated with the nanoparticle free colloidal system toward Newtonian behavior in stable bi-dispersed system, indicating tremendous drop of viscosity. Chan and Lewis [27] laid an insight on the interaction between binary system at three different conditions namely strongly attractive, weakly interacting and strongly repulsive which give rise to bridging, haloing or depleting effects.

Research by Luijten and co-workers has been dedicated toward validating the phenomena via numerical approach [31–33]. They discovered that the interaction between nanoparticle–microparticle in haloing mode is dominated by electrostatic attraction which gives relatively close agreement to the experimental results. In contrast, Karanikas and Louis numerically highlighted the role of electrostatic repulsion between nanoparticles in solution as dominating factor in the halo formation [28]. However, this condition requires much higher Debye screening length to match the existing experimental results.

Martinez et al. [36] and Lee et al. [37] used a confocal laser scanning microscopy technique to obtain imagery proof on the structural evolution and formation of colloidal crystal in a system containing particles with size and charge asymmetry. Lee et al. [37] further highlighted that the use of nanoparticle haloing technique enables microparticles to deposit on the substrate at much lower separation distance relative to the traditional approach of tailoring the pH away from the isoelectric points. Further, this gap can be regulated by simply changing the nanoparticle size and concentration within the binary system to give much broader range of physical property of the final product. By using ultrasmall-angle X-ray scattering technique, Zhang et al. [38] conducted quantitative validation on the nanoparticle halo formation by measuring the anterior and lateral separation distances between microsphere–nanoparticle and between segregated nanoparticles as well as the spatial distribution and concentration of nanoparticles within the halo structure.

The prediction on the halo formation has also been put forward via elementary analysis on the interaction potential within binary mixture which is based on the classical Derjaguin–Landau–Verwey–Overbeek (DLVO) theory [39,40]. The analysis highlighted the existence of weak repulsive barrier that forms a basis to further quantifying the strength of micro/nanoparticle interaction in experimental, numerical and theoretical approaches [17,26,30].

Ji et al. [30], McKee et al. [41], Hong and Willing [42] and Huang and Ruckenstein [34] investigated the aspect of interactive forces in binary system at different pH which represent different colloidal strengths and highlighted that nanoparticles absorption remained an underlying challenge to achieve ideally reversible colloidal stability. In corroboration to the results obtained by Ji et al. [30], Xing et al. [43] discovered that the haloing phenomena could also

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