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## Investigation of alumina nanofluid stability using experimental and modified population balance methods



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#### ABSTRACT

In this paper, the stability of alumina-water nanofluid is studied both theoretically and experimentally. The theoretical study is accomplished using the two-phase Eulerian-Eulerian method and the population balance model. The model considers interactions between nanoclusters and their fractal structures to predict size distribution, average size, and settling rate of nanoclusters for different temperatures, fractal structures, and spatial concentrations. The experimental study is accomplished using the dynamic light scattering (DLS) method. Experiments are also performed to validate modeling results. It is found that increased concentration and temperature of the nanofluid lead to increasing aggregation and settling rate of nanoclusters. The results also show that the reducing fractal dimension at certain concentrations of primary nanoparticles leads to an increasing rate of nanocluster aggregation. Generally, the findings of the study indicate that the population balance model can be successfully employed to investigate the stability of alumina-water nanofluid when the interactions between the nanoparticles and the fractal structures of nanoclusters are duly considered.

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#### 1. Introduction

Nanofluids form improved environments favorable to heat transfer with applications in electronic, transportation, medical, and aerospace industries [1]. One of the major challenges in the technological development of nanofluids is the problem of nanofluid stability which is defined as the resistance of nanoparticles against clustering. Stability of nanofluid depends on several parameters such as the types of nanoparticles and base fluid as well as the size of primary nanoparticles and nanoclusters in the fluid. These parameters may cause alterations in nanofluid properties including thermal conductivity, viscosity, and specific heat. Although thermal properties of nanofluid, especially those containing Al<sub>2</sub>O<sub>3</sub> nanoparticles, have been studied extensively, no consistent results have been provided [2–9]. One reason for the discrepancies observed in the results is the different sizes of nanoclusters present in the nanofluid despite the identical size of primary nanoparticles [10]. Both experimental and theoretical methods have been employed to investigate the clustering process in nanofluids and their stability. Sedimentation process, dynamic

\* Corresponding author. *E-mail address:* haghshenas@cc.iut.ac.ir (M. Haghshenasfard). light scattering (DLS), and UV-vis spectrum methods are typically employed in experimental studies of nanofluid stability [11–16].

The literature on theoretical investigation of nanofluid stability is quite rare [17–19]. However, theoretical methods are more useful tools not only because they save time and cost but because they also provide more detailed information about the aggregation process. These methods commonly consider aggregation of nanoparticles, collision mechanisms, and interactions among them.

The Population Balance Model (PBM) is one such method that captures all these three parameters. Collision mechanisms consist of Brownian motion (Perikinetic aggregation), imposed gradient velocity (orthokinetic aggregation), and differences in the settling velocity of individual nanoparticles [20]. Perikinetic and orthokinetic aggregations were first modeled by Esmelocheski and later developed by others [21–28].

The PBM was used for modeling such processes as crystallization [29,30], wet granulation [31], liquid-liquid extraction [32,33], fluidized bed reactors [34,35], polymerization reactors [35–37], powder mixing [38,39], and bubble columns [40,41].

The same techniques were also used in the field of nanofluids to calculate the particle size distribution which depends on time and space coordinates.

Charagozloo [17] modeled the thermal diffusion and aggregation process in nanofluids using the Monte Carlo method. Their



results showed that the presence of nanoclusters leads to variations in the thermal conductivity and viscosity of nanofluids.

Jiang et al. [18] proposed a model for considering simultaneously the aggregation and sedimentation of nanoparticles. They showed that nanoparticle concentration is the most important parameter involved in the aggregation of nanoparticles.

Recently, Krishna et al. [42,43] performed a computational and experimental study to investigate the particle agglomeration in nanofluids. The effects of nanoparticle size, volume fraction, pH, and electrolyte concentration on the rate of agglomeration are investigated. Numerical methods base on population balance equations have been used in their work and the results showed that the computational models can be quite useful to evaluate the shelf life and useful life of nanofluids.

Our review of above literature shows that the well-known PBM has not yet been used for predicting nanofluid stability. As such, the objective of the present paper is to study the stability of  $Al_2O_3$  nanofluid using a modified version of PBM to predict the aggregation of nanoparticles in a base fluid. The applicability of the model has been already shown for different processes [29–41]. In the present work, however, a modified version of the common PBM is used to consider the effects of Initial nanocluster size, fractal dimension, settling of nanoparticles, and the zeta potential on nanocluster aggregation rate. To our knowledge, none of the previous theoretical studies of nanofluid stability considered the four above mentioned parameters simultaneously [21–28].

Nanofluids usually are made in two-step method in more experimental researches. In this way synthesized nanoparticles dispersed in the base fluid. Usually, the ultrasonic device is used to disperse the nanoparticles [10]. Sadeghi et al. [10] and Mahbubul et al. [44] showed that nanoclusters do not reach to the primary nanoparticles size even in high mixing times of ultrasonic. In the experimental research different powers and ultrasonic mixing times were used. Therefor different initial nanocluster sizes were used in the experimental researches. On the other hand stability of nanofluids depends on their size.

The nanoclusters which formed by the collision mechanisms have fractal structure. The number of nanoparticles in each nanocluster depends on fractal structures and affects the aggregation and settling rates [20] which cause both nanoparticle concentration and aggregation rate reduces. It is usually assumed that the settling rate of nanoclusters is so low that its effects on concentration and aggregation rate are insignificant. However, this assumption does not seem to be reasonable for long times. The fourth parameter is the zeta potential that is not only commonly used in investigating the stability of nanofluids but also directly included in modeling nanoparticle aggregation. It depends on a variety of factors such as nanoparticle and base fluid types, salt concentration, and nanoparticle and nanoclusters sizes [10,45–47]. Any increase in the absolute value of Zeta potential leads to a corresponding enhancement in the stability of nanofluids. It should be noted that while the model proposed by Jiang et al. [18] failed to consider the effect of variation in zeta potential on changes in cluster size, the results of our previous study of nanofluid stability [10] show that zeta potential varies strongly with the size of nanoclusters, which in turn affects the rate of nanoparticle and nanocluster aggregation.

The rest of the paper is organized as follows. Section 2 presents the results of dynamic light scattering (DLS) and small angel X-ray scattering (SAXS) for the Al<sub>2</sub>O<sub>3</sub> nanofluid. These experiments are performed to obtain the data on fractal dimension, average size, zeta potential, and size distribution of nanoclusters. The results are then used as model input data and later for validating the results obtained from the model. Section 3 provides a brief description of the modified PBM and its application for modeling aggregation of nanoclusters. In Section 4, the calculation procedure is described. Section 5 provides the model results and discusses the effects of different parameters such as zeta potential of nanoclusters, concentration of primary nanoparticles, temperature and fractal dimension nanoclusters on aggregation rate. Finally, conclusions are presented in Section 6.

#### 2. Experimental procedure

For the purposes of this study,  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> nanoparticles (Nanoamor Co.) with a diameter of 25 nm and a specific surface area of 180 m<sup>2</sup>/g were dispersed in de-ionized water. An ultrasonic vibrator (Hielsher Up200, 200 W, 24 kHz, Germany) was used to disperse the nanoparticles in the base fluid (de-ionized water). An isothermal bath (Wisecircu WCL-P8, Korea) was employed to stabilize the temperature within ±0.5 °C throughout the experiments. The nanofluids thus prepared were used to study their stability at different concentrations of 1, 2, and 3 V% (by volume) and different temperatures of 15, 25, 35, and 45 °C. In these studies, the size distribution, zeta potential, and average size of nanoclusters after ultrasonic mixing were measured by a zeta-sizer (Malvern, HS C1330-3000) device using the dynamic light scattering (DLS) method. Fig. 1a shows the volume size distribution of the nanoclusters over various ultrasonic mixing times. The average size and zeta potential of nanoclusters versus ultrasonic mixing time are shown in Fig. 1b [10]. As reported in our previous work [10], nanofluids are stable for zeta potentials greater than 50 mV. Based on Fig. 1b, therefore, the nanofluid becomes stable after 180 min of

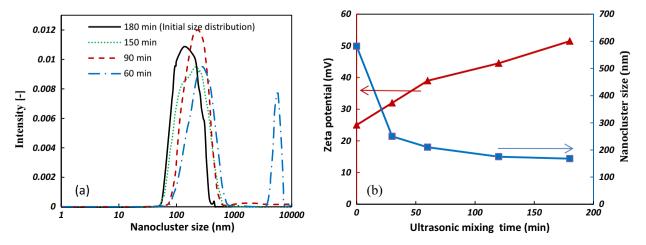


Fig. 1. DLS results, (a) normalized size distribution, (b) zeta potential and nanocluster size vs. ultrasonic mixing time (present study and Sadeghi et al. [10]).

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