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Effective ultrasonication process for better colloidal dispersion of nanofluid

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ABSTRACT

Improving dispersion stability of nanofluids through ultrasonication has been shown to be effective. Determining specific conditions of ultrasonication for a certain nanofluid is necessary. For this purpose, nanofluids of varying nanoparticle concentrations were prepared and studied to find out a suitable and rather mono-dispersed concentration (i.e., 0.5 vol.%, determined through transmission electron microscopy (TEM) analyses). This study aims to report applicable ultrasonication conditions for the dispersion of Al_2O_3 nanoparticles within H_2O through the two-step production method. The prepared samples were ultrasonicated via an ultrasonic horn for 1–5 h at two different amplitudes (25% and 50%). The microstructure, particle size distribution (PSD), and zeta potentials were analyzed to investigate the dispersion characteristics. Better particle dispersion, smaller aggregate sizes, and higher zeta potentials were observed at 3 and 5 h of ultrasonication duration for the 50% and 25% of sonicator power amplitudes, respectively.

1. Introduction

Stability is a critical and necessary condition for most of the materials used in industry, since it implies a fairly predictable and controllable condition of their behavior. In this regard, nanofluids are desired to have thermodynamic, kinetic, chemical, and dispersion stabilities [1]. Since nanofluids have been considered as advantageous in heat transfer applications due to their improved thermophysical properties, their stability in heat transfer experiments needs to be investigated. Due to the inter-particle adhesion forces, nanoparticles become agglomerated and their settlement can be observed due to the gravity forces. In order to start with a stable and usable condition of nanofluids, it is desired to have an aggregate- and sediment-free structure where all the nanoparticles contribute to the dispersion, which will give the maximum benefit from the nanoparticles, in terms of their thermophysical properties [2]. In this regard, a nanofluid with the stable dispersion can be defined in which the nanoparticles are mono-dispersed. Due to the presence of nanoparticle aggregates, the dispersion stability may decay with time [1]. Elcioglu and Okutucu-Ozyurt [2] indicate the requirement of performing stability measurements in a frequent and periodic manner. To increase the stable lifetime of nanofluids, ultrasonication has been widely utilized, and has been accepted as an essential step in the production of nanofluids through two-step method [3]. However, no standard has been established to prepare nanofluids especially on how long should a nanofluid have to be homogenized, how much sonicator power amplitude is needed, and what type or durations of pulse mode should be used. Nevertheless, the National Institute of Standards and Technology (NIST, Gaithersburg, MD) with the Center for the Environmental Implications of Nanotechnology (CEINT of Duke University) has started to develop some standardized and validated protocols for the dispersion of nanoparticles [4]. Use of cooling bath, pulse mode operation, and cylindrical shaped flat-bottom beakers are some proposed guidelines. They urged that, the optimal ultrasonication parameters should be determined by considering different parameters of the ultrasound process. It could be noted that ultrasonication is a complicated physicochemical process, which can break down the agglomeration as well as create further aggregation, and many other effects together with chemical reactions [4].

There are contradictory results among the researchers about the effect of ultrasonication duration on colloidal dispersion of nanoparticles. Some researchers pointed out that, higher ultrasonication duration is better for proper dispersion of nanoparticles. Among them, Yang et al. [5] studied the effect of ultrasonication on agglomeration size for nanotube-in-oil dispersions. They characterized the samples by TEM, and found that the cluster size

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decreased with increasing sonication time/energy. Amrollahi et al. [6] studied the effects of ultrasonication parameters on the settling time, and TEM microstructure of carbon nanotube (CNT)–ethylene glycol (EG) nanofluids. Their results showed that, for lower ultrasonication times, the settling in the most concentrated nanofluid (2.5 vol.%) was less than that of the most diluted nanofluid (0.5 vol.%), and for the longer ultrasonication durations, the phenomenon was reversed. The precipitation measured by human eye is not a precise method even though the author claimed that the precision was ±10 min. Again, the author carried measurements with TEM only after three durations as 15 min, 5 h, and 20 h of ultrasonication and for only 2.5 vol.% concentration of particles. Ruan and Jacobi [7] applied 5, 40, 140, 520, and 1355 min of ultrasonication duration to homogenize multi-walled carbon nanotube (MWCNT) in EG. The nanofluids were prepared by using both continuous and pulsed mode of ultrasonication. Microstructure, agglomerate size, nanotube length, and aspect ratio were determined through TEM to study the effect of ultrasonication. They observed that, average cluster size, length, and aspect ratio of nanotubes decreased with increasing sonication time or energy.

Most other researchers report that, there are specific optimal ultrasonication durations available based on different conditions/properties of nanofluids, e.g., particle concentration and type, and amount of and type of base fluid [8]. Chen et al. [9] ultrasonicated TiO_2-EG suspension up to 40 h to find out the optimum sonication duration. Their characterization with light scattering for agglomeration size showed that, 20 h of homogenization gave the best result that was 140 nm size and for longer durations no further size reduction was achieved. Garg et al. [10] investigated the effect of sonication time on the dispersion behaviors of nanofluids. They prepared four samples of 1 wt.% MWCNT in DIW with GA as additives and subjected the samples to ultrasonication for 20, 40, 60, and 80 min. They performed analyses with TEM and found that the optimum ultrasonication time for homogenization was 40 min, using a 130 W and 20 kHz ultrasonicator. Zhu et al. [11] determined the influence of ultrasonication time on average cluster size. They analyzed the dispersions of $CaCO_3$–water, which were ultrasonicated for 1–45 min and found that, the cluster size rapidly decreased within 20 min of ultrasonication, after that, it was slightly increased with ultrasonication duration. As their primary substance was in paste form, therefore, most of the aggregates were soft and they were broken up rapidly within 20 min. Nguyen et al. [12] studied the effect of ultrasonication duration, power, and pulse mode on de-agglomeration of alumina nanoparticles dispersed in water, where the maximum input power of the machine was 400 W with a frequency of 20 kHz. They used 10%, 30%, and 60% of vibration amplitude with different pulse modes and optimal break-up of agglomeration were found for 30% amplitude. In the case of 60% amplitude, the cluster size again increased after 300 s of ultrasonication. Hence, the authors point out that, higher power of ultrasonication could result in re-agglomeration of the particles. Nevertheless, for 10% and 30% amplitudes, the aggregate sizes were continuously decreased by the increase of sonication time. They used different modes of pulse as continuous and pulsed with long and short durations; however, no difference and similar outcomes were observed. Chakraborty et al. [13] analyzed the influence of ultrasonication durations on TiO_2 nanofluid. They added 0.1, 0.2, and 0.4 wt.% of silver (Ag) nanoparticles and ultrasonicated for 10, 20, and 30 min of durations. They observed the settling time and report that for lower concentration of particles, ultrasonication did not have a significant role. Kole and Dey [14] ultrasonicated ZnO nanoparticles in EG up to 100 h and characterized the PSD and microstructure. They reported that, the lowest cluster size was obtained for 60 h of sonication and after that, cluster size again increased. Mahbubul et al. [15] investigated the effect of the ultrasonication duration (0–180 min, 50% amplitude, 2 s ON and 2 s OFF pulses) on the colloidal structure of 0.5 vol.% alumina–water nanofluid. The authors observed a decrease in the aggregate size for ultrasonication up to 90 min. For longer durations (i.e., for 120 and 150 min) particles formed aggregates, again. Further ultrasonication until 180 min yielded more homogeneous dispersion of nanoparticles.

It can be inferred from the literature that the studies about the effect of ultrasonication on the aggregation tendency of nanoparticles within nanofluids are still immature. Some of the researchers recommend higher sonication time for better dispersion, while some other researchers claim that the agglomeration could be minimized after certain duration of ultrasonication. Nevertheless, there is no specific or common duration of ultrasonication suggested by the researchers that could be followed for better dispersion. Moreover, most of the literature studied only the sonication period and most are concerned with CNT nanofluids. Hence, the present study aims to evaluate the effective ultrasonication conditions (sonicator amplitudes and sonication duration) on dispersion characteristics to prepare an alumina–water nanofluid through two-step method. The research is the extension of our previous study [15] that was fixed with 50% amplitudes and until 180 min of duration only. Here a prolonged ultrasonication duration until 5 h is considered for analysis. Moreover, two different amplitudes as 25% and 50% of sonicator power were used for the analysis with the hope that this study will give more guidelines for the researchers regarding ultrasound sonication.

2. Experimental method

2.1. Nanofluid preparation

The Al_2O_3 nanoparticles in powder form (manufactured by Sigma–Aldrich, USA) with the manufacturer defined average particle size of 13 nm and a purity of 99.5% was dispersed in distilled water, to prepare the nanofluids. The nanofluids were prepared via the two-step method, i.e., the nanoparticles were primarily arranged and then mixed with the base fluid using ultrasound [16]. Four volume concentrations (0.01, 0.1, 0.5, and 1 vol.%) of Al_2O_3–water nanofluids have been prepared using 50% ultrasonication amplitude with 2 s ON and 2 s OFF pulses for 1 h of ultrasonication. Then the microstructures of these four samples were analyzed by a TEM (Model LIBRA 120, Zeiss, Germany). The TEM results are provided in Fig. 1.

Based on the TEM analyses, the dispersion characteristics of the samples with varying nanoparticle concentrations can be observed in Fig. 1. It is revealed from the TEM micrographs that, the particles were in a rather involved and overlapping condition for 1 vol.% nanofluid compared to the 0.01, 0.1 and 0.5 vol.% samples. Such an observation of the sample microstructure can give preliminary conclusions on the nanoparticle-clustering tendency, which is inevitable in the long term. In order not to render the possible improvements in thermophysical properties coming with the increased nanoparticle concentration, 0.5 vol.% nanofluid is selected for further investigation as it appeared to be the preferable one among the concentrations studied, in terms of the nanoparticle dispersion. The sample of 1 vol.% was found to be the most concentrated nanofluid. However, 0.01 vol.% was observed to have the most diluted concentration. Hence, 0.5 vol.% of Al_2O_3–H_2O nanofluids have been further investigated for the effective ultrasonication parameters.

First, the nanoparticles were suspended in the base fluid, and a very narrow (3 mm diameter) glass tube was used to stir the mixture for 1 min to enable the nanoparticles to be mixed with the base fluid completely. Then, the nanofluids were ultrasonicated