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Research Article

An experimental study on resuspension, thermostability and migration phenomenon of nanoparticles in pool boiling

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ABSTRACT

Nanoparticles have proven to be effective in sensible and latent heat exchanges alike. Applications of nanoparticles in phase change processes are associated with migration and resuspension of nanoparticles upon which our existing knowledge is very limited. This work experimentally investigates the migration ratio, stability and resuspension of nanoparticles during phase change. Knowledge on migration ratio is essential to gauge the thermal and lubricative enhancements in the subsequent processes. Al_2O_3/Water & CuO/Water nanofluids were prepared in four mass fractions (0.05, 0.1, 0.2, 0.4) using ultrasonic agitation technique. Nanofluids with mass fraction higher than 0.5% displayed poor stability over time also, agglomeration and sedimentation were pronounced and inevitable. Nanofluid destabilises and agglomerates rapidly at temperatures closer to saturation temperature. Resuspension of agglomerated chunks were observed during nucleate boiling where the test fluid became extremely nonhomogeneous. Migration ratio was found to commensurate with volume fraction where CuO/water nanofluid exhibited 23% lesser migration ratio than Al_2O_3/water nanofluid. Maximum migration ratio of 17.8% was observed for Al_2O_3/water with 0.05 wt%. Maximum migration was found when the molecular dimensions of nanoparticles and the base fluid are of similar magnitudes. It is inadvisable to involve nanoparticles in phase change systems.

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INTRODUCTION

Nanoparticles have been the standard prescription for years now, for thermal enhancement in heat exchangers, refrigerators and power producing systems[1–8].The phenomenon of migration of nanoparticles is a concomitant of phase change process, boiling and condensation alike. A lot of work has been carried out in understanding the thermophysical properties of nanofluids [9–17]. As the nanofluid is heated, it eventually reaches the state of nucleate boiling. The nanoparticles suspended in the base fluid move rapidly due to the higher energy state. Bubbles form and rise from the nucleation sites and when nanoparticles strike the bubble, they stick to the bubbles surface. As the bubble rises and collapses near the interface, the nanoparticles are lodged into the vapour stream and they leave the fluid medium. Migration ratio (MR) is the ratio between the mass of nanoparticles leaving the fluid media during boiling to the mass of nanoparticles initially present in the nanofluid. This phenomenon strongly depends on the size, shape and mass fraction of nanoparticle, vessel dimensions and properties of the base fluid.

Migration studies have been predominantly centered on the domain of refrigeration; heat exchangers, chemical processing industries, etc. especially vapour compression refrigeration systems(VCRS)[18–23]. Understanding the migration ratio is absolutely essential to estimate the actual impact of nanoparticles on the improvement of performance. Also, theorizing the particle-fluid interaction is important to better utilization of nanofluids in heat transfer devices. Kleinstreuer et al. [24] extensively reviewed on the thermal enhancement and thermophysical properties of nanorefrigerants. It was clearly evident from the literature that studies on migration of nanoparticles were absent and disregarded.

Ding et al. [19] were the pioneers in the domain of migration during pool boiling. Experiments were conducted presuming the boiling in the evaporator of VCR system as pool boiling. Experimental setup comprises of a vented beaker with thermocouples affixed at various positions. An electric heater is kept at the base of the beaker and the whole setup is punctiliously placed on an electronic precision balance. MR decreased with increase in volume fraction of nanoparticles. A rather counterintuitive result was presented and the impact of various parameters, such as, liquid column height, geometry of the evaporator, heat flux and intrinsic properties of the base fluid were not experimented. Later, very similar experiments with more data points were carried out by the same group of researchers. Peng et al. [19,25] studied the effects of viscosity, surface tension and thermal diffusivity on migration ratio. Usage of lubricant oil and increase in heat flux reduces the migration of nanoparticles. Increase in liquid column height improves the MR due to a large dwell time inside the nanoparticle abundant fluid phase. Peng et al. [26]carried out experiments using MWCNT with varying aspect ratio (100, 666.7, 18.8, 125). Results revealed an optimal aspect ratio of 125 at which MR is nearly 27%. From the existing literature a small notice was taken on the measurement technique, all the researchers have used weighing technique to gauge MR.

Stability of nanofluids is the only concern inhibiting the widespread usage in commercial and domestic applications. Wen et al. [27] conducted experiments to understand the impact of stability on the heat transfer rate. It was concluded that nanoparticles deteriorate the heat transfer by depositing on the heat transfer surface and by acting as a resistive layer. Resuspension of nanoparticles in the base fluid is essential to improve the stability of nanofluid. Senthilkumar et al. [28] discussed the effects of silicon carbide nanoparticles in a domestic refrigerator. R134a refrigerant is used with SiC of 0.325 g (0.2 vol.%) of nanoparticles. The average COP of 1.24, 2.0 and 1.81 was observed for the test case of conventional R134a, R134a-cryo-SiC refrigerant and R134a-SiC nanorefrigerant respectively. Recently, Lin et al. [29] experimentally studied the resuspension phenomenon of deposited copper nanoparticles during pool boiling. Experiments recorded with high-speed camera revealed the subtle recirculation of nanoparticles due to rigorous bubble formation. Ghadimi et al. [30] reviewed on the stability and preparation techniques of nanofluid. It was reported that the existing literature is ambiguous about the stability of nanofluid. One dilemma which was prevalently observed was on the saturation mass fraction. Many researchers have reported that mass fraction of 1% and above exhibits drastic agglomeration and sedimentation[30–33] and another group of authors describing the high stability of nanoparticles till 5% volume fraction [34–37]. Chung et al. [38] reported that ZnO/water mixture of 0.02% volume fraction exhibited stability for more than 10000-hrs. Ghadimi et al. [39] & Khooshechin et al. [40] studied the influence of preparation time on extent of stability where the ultra-sonication time was varied for different mass fractions. Usage of 0.1% of TiO_2 nanoparticle with 3-hrs of ultra-sonication provided the most stable mixture. Kumar et al. [41] studied the stability of SiO_2-TiO_2/water/Polyacrylamide at temperatures closer to the boiling point of water. The fluid displayed good thermo-stability till 90°C.

The authors of this work have extensively reviewed the boiling, condensation and tribological properties of nanorefrigerants [7]. The authors suggested few future works in the review, such as experiments concerning the safety and environmental impacts of nanoparticles. Similarly, experimental studies concerning the wear and tear in the compressors were suggested. Another key point mentioned was regarding the migration of nanoparticles in evaporator and condenser which is one of the most important parameters to understand the sustainability of the performance enhancement due to nanoparticles. In this paper experiments have been carried out with CuO and Al_2O_3 nanoparticles. Distilled water was used as the basefluid due to its non-flammable, non-toxic nature, ease of handling and its high boiling point. CuO/water & Al_2O_3/water nanofluids were prepared for four volume fractions (0.05, 0.1, 0.2, 0.4). Boiling and weighing technique was adopted to gauge the MR. Experiments were visually monitored to take notice of the resuspension and thermostability of the ebullient mixture. Also, a novel technique involving viscosity was tested to indirectly correlate the value of migration ratio.

THEORY

Nano Particle Preparation

CuO and Al_2O_3 nanoparticles of 30-50 nm size was weighed using a precision balance (Citizon_CY_204) with a least count of 0.1 mg. Nanoparticles were dispersed in the base fluid, water by ultrasonic agitation for four mass fractions of 0.05%, 0.1%, 0.2% and 0.4%. CuO nanoparticle has a true density of 6.4 g/cm^3 and Al_2O_3 has a true density of 3.97 g/cm^3. The time period of agitation was optimally selected for each volume fraction as reported in Afzal et al. [39] and Ghadimi et al. [42] . A considerable mass of base fluid was lost due evaporation corollary of high temperatures during ultra-sonication. Figure 1 shows the ultrasonicator device (Pci_analytics_DP-120, 20Khz, 750 W) with unagitated concoction.

The mass fraction of the prepared mixture might exhibit slight deviation from the intended mass fraction due to the aforementioned loss. For mass fractions higher than 0.4%, sedimentation was rapid and instantaneous. Also, the temperature rose to saturation temperature which led to rapid evaporation and rigorous cavitation. It must be noted that CuO was hard to prepare due to is poor stability even at lower mass fractions but also due to its rapid temperature change. Hence a stop-start method was adopted to avoid huge deviation from the expected mass fraction. Figure 2 shows the freshly prepared CuO and Al_2O_3 nanofluids.

In this work, two different test setups were used to study migration and resuspension of nanoparticles independently. The setup used to study resuspension consists of an oil bath, magnetic stirrer-hot plate (Remi 2MLH, 500W, 1200 rpm) and a custom-made lid for holding six test tubes. The test tubes were filled with the prepared nanofluids then immersed into the hot oil bath placed on the hot plate. This arrangement was adopted to have a deeper vessel to encourage the resuspension and rigorous bubble formation with a motive to visualize resuspension. But this setup was infeasible to study migration characteristics since, the condensation of the vapors along the walls of the test tube induced errors in the value of migration ratio. Also, the condensation drastically increased the time taken for complete boil off. This was antithetical to the primary purpose of the build in the first place. To circumvent the aforementioned constrains, another test setup was proposed. This consists of a petri dish (7.5 cm dia.) and the magnetic stirrer-hot plate. In this setup the dish was filled with nanofluid (10

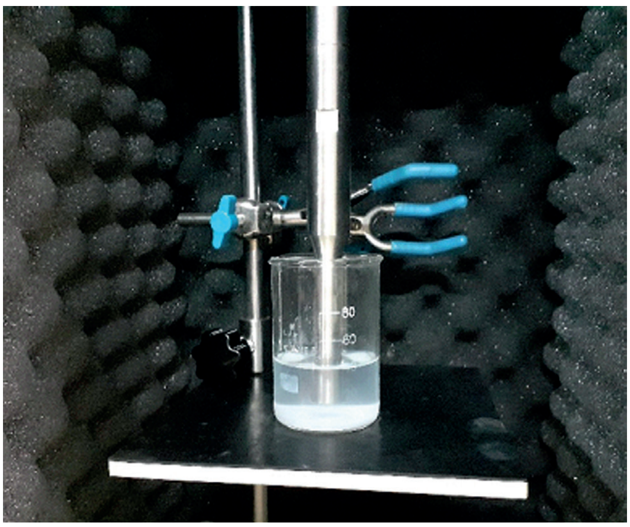

Figure 1. Unprepared Al_2O_3/Water concoction.

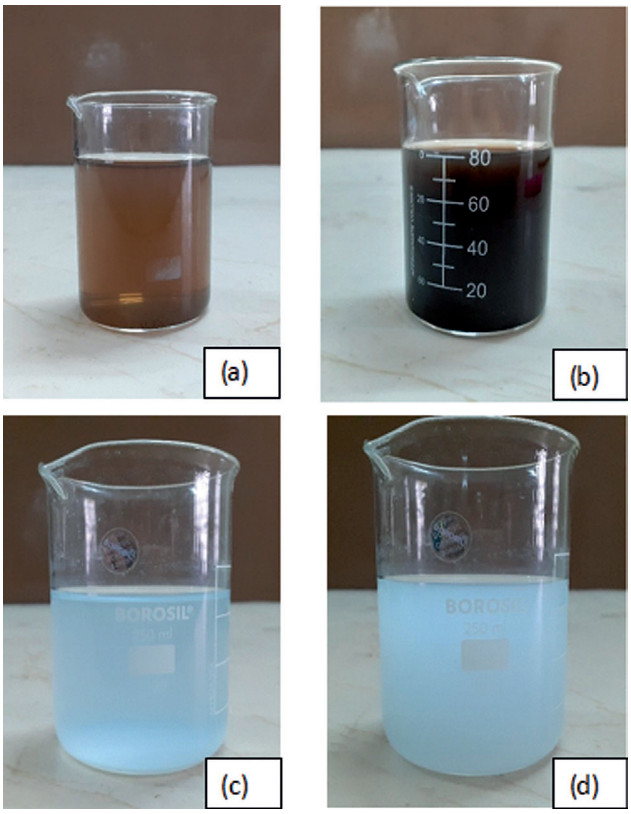

Figure 2. Freshly prepared CuO/water nanofluid of mass fractions a) 0.05% b) 0.4% Al_2O_3/water nanofluid of mass fractions c) 0.05% d) 0.4%.

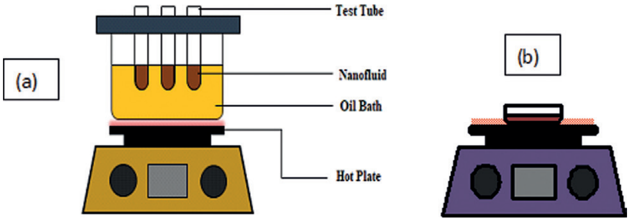

Figure 3. Line sketch of the setup for studying a) Resuspension b) Migration ratio.

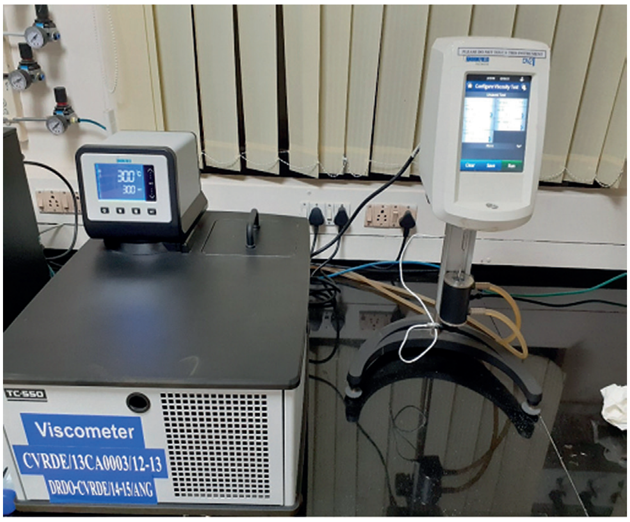

Figure 4. Brookfield viscometer with isothermal heat source system used to test viscosity.

ml, 30 ml and 50 ml) and using the hot plate the fluid was boiled and dried. The petri dish is shallow compared to the test tube hence there was no recondensation on the walls hence this test setup was adopted to study the MR. Figure 3 is a line diagram of the experimental setups used for studying resuspension and migration.

Viscosity of all the samples was measured using a Brookfield viscometer (Ametek_DV2T, accuracy of 1% and repeatability of 0.2%) at 80 rpm and 29.9 ± 0.2°C. The values were measured at three data points and averaged to obtain an accurate result. Viscosity of fresh samples and re-sonicated residual samples were measured and checked with existing correlations for the respective nanoparticle fluid combination. Figure 4 shows the Brookfield viscometer used for this experiment.

Weighing technique is a simple, yet effective method which is prevalently used to measure the migration ratio. Another technique which is used in the recent times is spectroscopy [43]. A new technique which can indirectly measure the nanoparticle concentration was also tested. The residual nanoparticles in the glassware after complete

Table 1. shows the measured values of viscosity of the original and prepared mixtures.

Nanoparticles	Mass fraction (%)	Viscosity of Original fluid (Centi Poise)	Viscosity of residual (Centi Poise)
CuO	0.05	1.190	1.391
	0.1	1.507	1.718
	0.2	2.160	2.410
	0.4	3.620	3.724
Al_2O_3	0.05	1.080	1.176
	0.1	1.377	1.660
	0.2	1.993	2.103
	0.4	3.285	3.125

boiling were ultrasonicated again. This new fluid has a concentration lesser than that of the original sample for which the mass fraction is known. The viscosity of both the samples was tested, similarly for all the mass fractions. The concept was to arrive at the mass fraction by using the existing correlations of the viscosity. Nguyen et al. [44] described clearly on this hysteresis effect in Al2O3/water nanofluid. A similar aberration was observed in this study, irrespective of the nanoparticles.

Methodology

To study migration ratio, the prepared nanofluids were filled in the petri dish in three volumes (10 ml, 30 ml and 50 ml). Initial mass of the petri dish (mi) was weighed on the precision balance thrice and averaged. The filled petri dishes were placed on the hot plate to boil the nanofluid. The petri dish was weighed upon the completion of boiling; consequently the petri dish was heated for another 10 mins and then weighed to ensure that no moisture remains on the petri dish. This process continues till two consequent values of the measured mass concur. Residual mass (mr) can be gauged from the difference between mass after boiling (mf) and initial mass of the petri dish (mi). Migrated mass (mmig) is the difference between residual mass and the original mass of nanoparticles (mn). Once the experiment is completed the residual nanoparticles are re-sonicated with the same volume of base fluid. This fluid is used in the indirect testing technique which measures the mass fraction through viscosity.

RESULTS

Migration of Nanoparticles

Migration ratio is the fraction of nanoparticles adherent with the vapour phase from the liquid phase during boiling and it is the key parameter to understand the sustainability

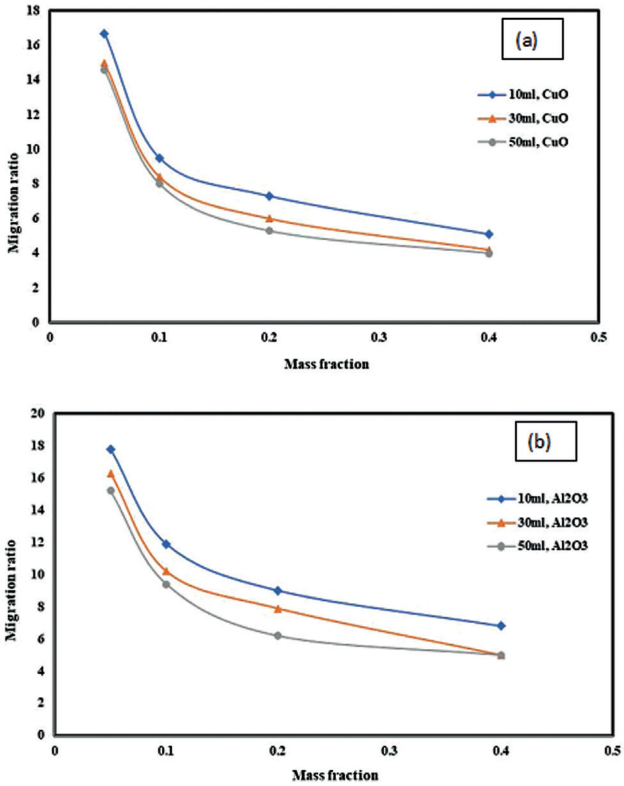

Figure 5. Migration ratio against the total liquid volume for a) CuO b) Al_2O_3.

of the coveted enhancement achieved by the addition of nanoparticles. Higher migration ensures the continuity of nanoparticles in the working cycle, lower migration ratio is undesired in most of the applications. The test petri-dishes were weighed using the precision balance the mass migrated was noted by equation the existing and prior masses. Migrated mass decreased as the mass fraction was increased from 0.05% to 0.4%. It was observed that Al_2O_3 nanoparticles displayed better migration when compared to CuO nanoparticles of the same mass fraction. A maximum of 17.8% migration was observed for Al_2O_3 of 0.05% mass fraction. Migration is also a function of total liquid volume. As the liquid volume was increased from 10 ml to 50 ml, the migration ratio decreased from 11.9% to 9.4% for Al_2O_3 of 0.1%.

Migration of nanoparticle is more for lower liquid volume because when the liquid level is low for the same nanoparticle mass fraction, it takes lesser time to totally boil off and hence, the nanoparticles remain for a shorter period in the fluid media. This means lesser chances of agglomeration and better migration. Similarly, lower volume fraction means the nanoparticles are sparsely suspended which in turn accounts for lesser agglomeration. It must be noted that the absolute migrated mass increases with mass

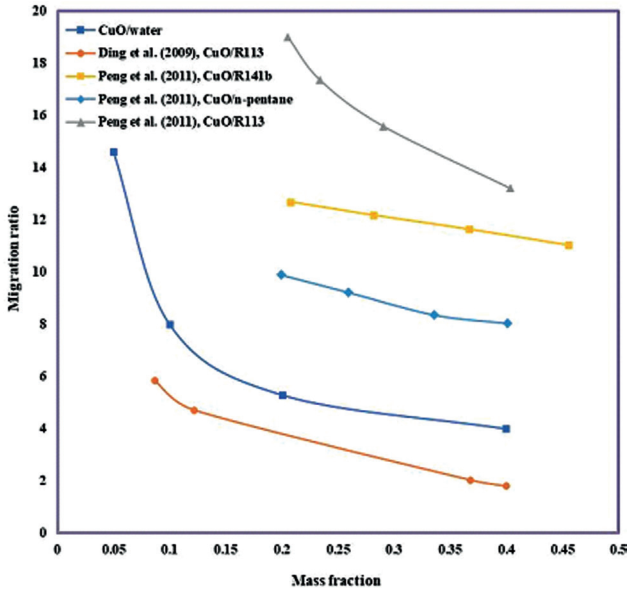

Figure 6. Impact of basefluid on the migration of nanoparticles.

fraction but the percentage of migration decreases. Figure 5 shows the migration ratio against the total liquid volume for CuO and Al_2O_3 nanoparticles.

Impact of Nanoparticle and Base fluid properties on Migration

The importance of base fluid is often neglected while the properties of nanoparticles are given more significance. There exists a great sense of ambiguity in the relationship between properties of base fluid and migration ratio. A necessary attempt was made by consolidating the existing literature and the present work to comprehend the impact of base fluid on migration ratio. Data points at similar working conditions were obtained and a comparison was made. Figure 6 shows the relationship between base fluid and migration for the same nanoparticle (CuO). It is clear from the graph that the molecular mass of refrigerant has a positive impact on migration ratio. Heavier base fluids are observed to exhibit more migration where R113 > R141b > n-pentane > water. More specific research is required to vividly correlate the other thermophysical factors like viscosity, thermal diffusivity, surface tension and thermal conductivity.

Figure 7 shows the impact of nanoparticle on the migration ratio. It can be observed that CuO nanoparticles migrate poorly independent of their size and base fluid combination. Size of nanoparticles plays a significant role on the migration behaviour. From Figure 8, it can be inferred that particles with smaller dimension migrates better when compared to their larger counterpart. Al_2O_3

exhibits the highest migration ratio of 31.33% at a mass fraction of 0.2%. All the nanoparticles seem to obey a similar relation with mass fraction where the curves show a similar decadence.

It is well known that effective momentum transfer happens between particles of similar dimensions. In this scenario molecules of heavier base fluid approaches the dimensions of the suspended nanoparticles and any collision will provide maximum momentum transfer. In order for migration to occur the nanoparticle trying to exit the liquid domain must have enough momentum to break through the liquid-air interface's surface tension. For particles of similar dimensions, it's easier to escape the liquid medium. For maximum migration to occur the dimensions of the molecules of base fluid must be large (R113 > R141b > n-pentane > water) and the dimensions of nanoparticles must be as small as possible (CuO – 20 nm> CuO – 30 nm> CuO – 40 nm).

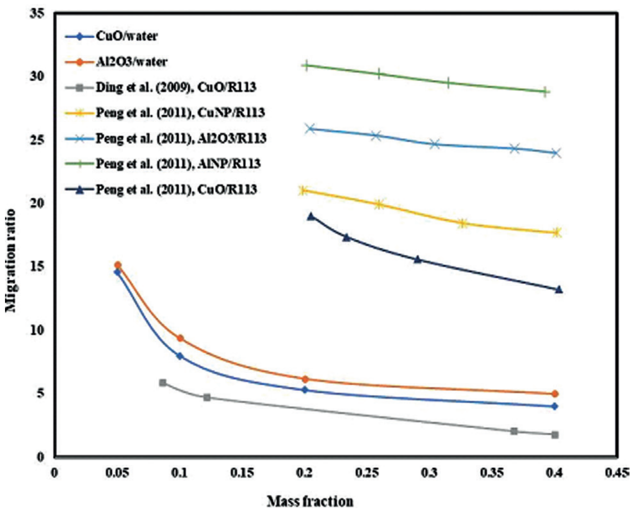

Figure 7. Impact of basefluid on the migration of nanoparticles.

Stability of Nanofluid

The stability of nanoparticles is of paramount importance for any application. In this study, stability of the solution was gauged by visual inspection. The samples were kept undisturbed in a vibration free environment and the samples were periodically inspected. High resolution images were captured after every 15 mins until complete sedimentation. Figure 8 shows the stability of nanofluid against time at room conditions (34°C, 1 atm.). From the image it can be clearly observed that CuO and Al_2O_3 sediments within 12-hrs of preparation. Al_2O_3/water nanofluid mixture exhibited stability for a longer period of time when compared with CuO/water mixture of same the mass fraction. Al_2O_3/water nanofluid of 0.05% mass fraction displayed remarkable stability for more than 8-hrs whereas, CuO particles sediment rapidly. It was also observed that mass fractions above 0.4% sedimented rapidly, independent of the type of nanoparticle and hence, the experiments were limited to a mass fraction of 0.4%.

Thermostability is more complex and less understood than the stability of nanofluid at room temperature. Thermostability can be defined as the state where nanoparticles are being suspended during evaporation or boiling. Unlike stability at room condition which is expected to

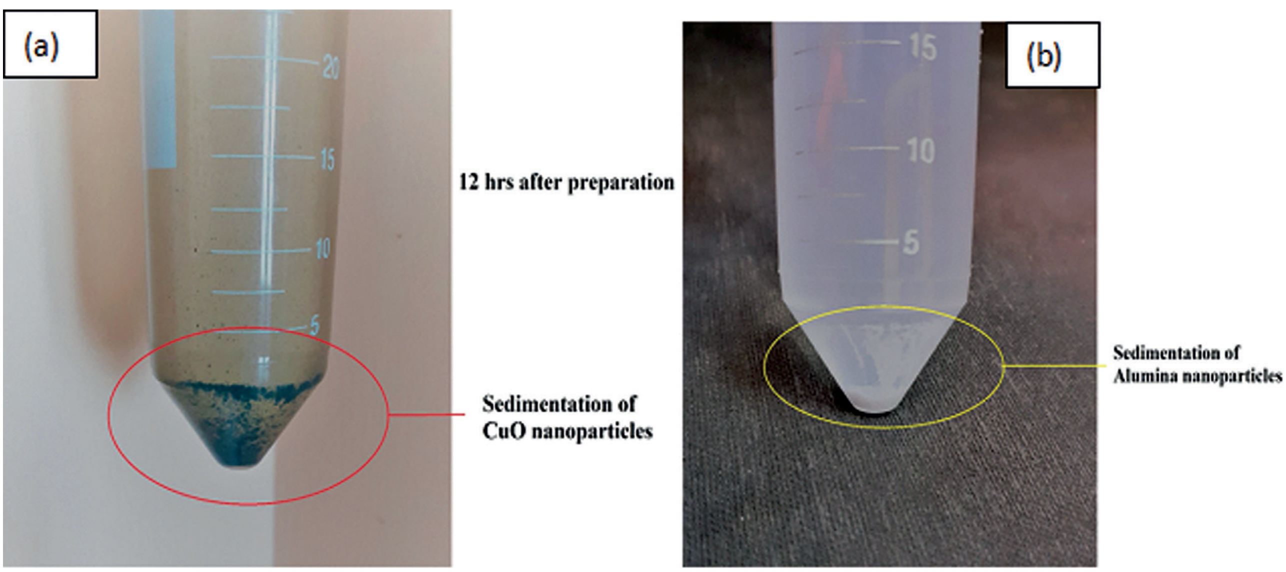

Figure 8. Test samples of (a) CuO and (b) Al_2O_3 after 12-hrs of preparation of 0.4% mass fraction of nanofluid.

last for hours this phenomenon is expected to last until boiling is completed. In the case of both CuO and Al_2O_3 nanoparticles, poor thermostability was observed at higher wall temperatures. From Figure 9, it can be seen that the nanoparticles sediment at the bottom of the test tube which was heated using an oil bath and hot plate. It shows the stability of nanofluid against time at elevated temperature. Both the mixtures completely agglomerated just before boiling. This lack of stability can be ascribed to the enhanced Brownian motion at elevated temperatures where the collision frequency of the suspended particles increases and the rate of agglomeration increases. Also, stabilizing agents such as surfactants are unstable at elevated temperatures. This unstable behaviour renders nanofluid incompetent for high temperature applications.

CuO nanoparticles exhibited a strange change in colour upon complete boiling, where the nanoparticles changes from black colour before sonification to golden yellow after complete boiling. This could be a result of agglomeration because our existing knowledge says that the colour of nanoparticles depends on the nanostructure and size. A myriad of colours can be observed for the same nanoparticle by just varying its size and they are different from what is observed at macro level. The nanoparticles may have changed its shape and size as a result of agglomeration and so there was a distinct change in colour [45].

Resuspension of Nanoparticles

During nucleate boiling the sedimented and agglomerated nanoparticles are unsettled by the rigorous bubble formation. The bubbles; put the nanoparticles into the convection current there by stopping it from sedimentation. During the experiments, resuspension of nanoparticles was observed well before the achievement of saturation temperature. The currents in the fluid domain were strong enough to keep the agglomerated chunks in motion. When the fluid entered nucleate boiling regime the majority of the nanoparticles had already agglomerated and the resuspended chunks were visible to naked eye. Resuspension of those agglomerated chunks may not be as effective as a homogeneously spread nanoparticle phase. Even though the phenomenon of resuspension is quite strong, it is not strong enough to break the agglomerated particles. Lin et al. [29] conducted experimental studies on resuspension behaviours of R141b/Cu Nanoparticle. This study and the mentioned work agree on the results inferenced on the resuspension of nanoparticles. Yet, the impact of resuspension & agglomeration on heat transfer rate, rheological & thermophysical properties is still ambiguous. More research is need in evaluating those faucets of nanofluids.

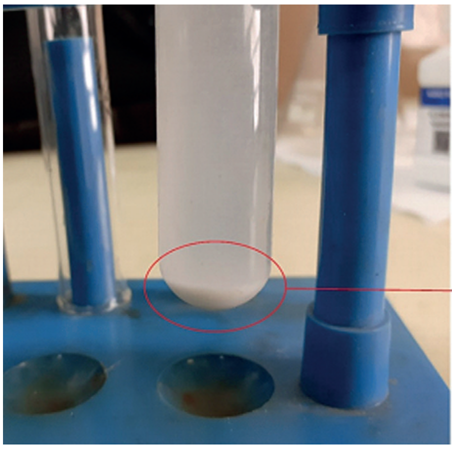

Figure 9. Stability of nanofluid against time at elevated temperature.

CONCLUSIONS

In this paper an experimental study was made to comprehend the three enigmatic phenomena observed in nanofluids namely, migration, resuspension and thermostability. During the investigation of this work the following results were concluded.

- CuO nanoparticles were less stable than Al_2O_3 nanoparticles for all the mass fractions. Similarly, CuO migrated 23% lesser than Al_2O_3.
- Al_2O_3 nanoparticles displayed better migration when compared to CuO nanoparticles of the same mass fraction. A maximum of 17.8% migration was observed for Al_2O_3 of 0.05% mass fraction
- Increase in both liquid volume and mass fraction negatively impacted the migration ratio. Al_2O_3 of 0.05% exhibited the maximum migration ratio of 17.8%.
- Heavier basefluid and smaller nanoparticles exhibit better migration due to effective momentum transfer.
- Migration ratio order R113> R141b> n-pentane> R718. Similarly, migration performance of CuO nanoparticles of average dimensions is in the order of 20 nm> 30 nm> 40 nm.
- A strange discolouration of the residual CuO nanoparticles was observed. The colour has changed from black to golden yellow over the course of boiling.
- Thermostability of CuO & Al_2O_3 nanoparticles is poor, complete sedimentation occurs within minutes. Nanofluids are ineffective in high temperature applications.
- Resuspension of nanoparticles exists well before the onset of pool boiling. Rigorous bubble formation during nucleate boiling is not strong enough to disintegrate the agglomerated chunks.

As a cumulation of all the results observed and measured, it can be concluded that application of nanoparticles in any phase change application is inadvisable. The desirable effects of nanofluid will deteriorate with time quickly at lower migration ratios. Lower migration also aids the agglomeration and sedimentation proclivities. In the future a lot of work in the domain of resuspension and thermostability is required. Currently, our knowledge on the stability and migration characteristics of nanoparticles is at a rudimentary level. More work is also need to understand the correlation between the basefluid and nanoparticle combination to better understand migration of nanoparticles.

NOMENCLATURE

Al_2O_3	Aluminium oxide
CuO	Copper II oxide
VCRS	Vapour compression refrigeration system
MR	Migration ratio
MWCNT	Multi walled carbon nanotubes
ZnO	Zinc oxide
TiO_2	Titanium IV oxide
SiO_2	Silicon IV oxide
RPM	Rotations per minute

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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