

Review of nanoscale boiling enhancement techniques and proposed systematic testing strategy to ensure cooling reliability and repeatability



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HIGHLIGHTS

- This review explores merits and drawbacks of two-phase nanoscale enhancement techniques.
- Emphasis is placed on inconsistencies in reported mechanisms and trends.
- Review shows lack of long-term and repeatability experiments.
- New 'Uniform Testing Protocol' is proposed for future nanoscale enhancement studies.

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ABSTRACT

Boiling heat transfer enhancement using nanoscale techniques has received considerable attention in recent years. This paper provides a comprehensive review of published articles addressing such techniques for both pool boiling and flow boiling enhancement. Discussed are heat transfer merits or drawbacks (in regards to both heat transfer coefficient and critical heat flux) for each technique, with particular focus on reliability concerns stemming from any time-dependent variations in boiling performance. Despite the many studies reporting measurable improvement in heat transfer performance, significant inconsistencies exist regarding both dominant enhancement mechanism and heat transfer trends. And lacking most in the vast majority of studies are long-term and repeatability experiments. To tackle this critical shortcoming, the authors are proposing a step-by-step strategy for future nanoscale enhancement studies. Without this type of strategy, it is believed two-phase nanoscale research will continue to entail simplistic curiosity experiments, lacking conformance to testing norms learned from decades of two-phase research, or convincing evidence of viability of enhancement schemes to practical cooling applications.

1. Introduction

1.1. High-Heat-Flux cooling applications and solutions

High-heat-flux applications refer to a specific category of systems demanding removal of heat fluxes in the range of 25–1,000 W/cm² for safe operation [1,2]. They include high performance microprocessors, hybrid vehicle power electronics, x-ray medical devices, commercial and military avionics, fuel cells, geothermal systems, power plant boilers, and spacecraft thermal control systems (TCSs). But there are also far more demanding cooling situations, such as fusion reactor blankets, particle accelerator targets, laser and microwave directed energy

weapons, advanced radar, rocket nozzles, gas turbine engine blades and afterburners, which are categorized as ultra-high-heat-flux applications, demanding removal of heat fluxes well in excess of 1,000 W/cm² [1,2]. Fig. 1(a) shows examples of both high-heat-flux and ultra-high-heat-flux applications. In both heat flux categories, failure to remove such vast amounts of heat can result in permanent device or system damage via meltdown or burnout. Clearly, developing highly effective cooling solutions has become an integral part of system design in virtually all these applications.

Clearly, heat dissipation levels in both high-heat-flux and ultra-high-heat-flux applications have exceeded capabilities of traditional air cooling and single-phase liquid cooling schemes. This is evident from

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Nomenclature		Subscripts	
G	mass velocity	f	liquid
h	heat transfer coefficient	in	inlet
q''	surface heat flux	nf	nanofluid
q''_{CHF}	critical heat flux	$vol.$	volume
T	temperature	w	heated wall
T_{sat}	saturation temperature	$wt.$	weight
T_w	heated surface temperature		
ΔT_{sat}	surface superheat ($T_w - T_{sat}$)		

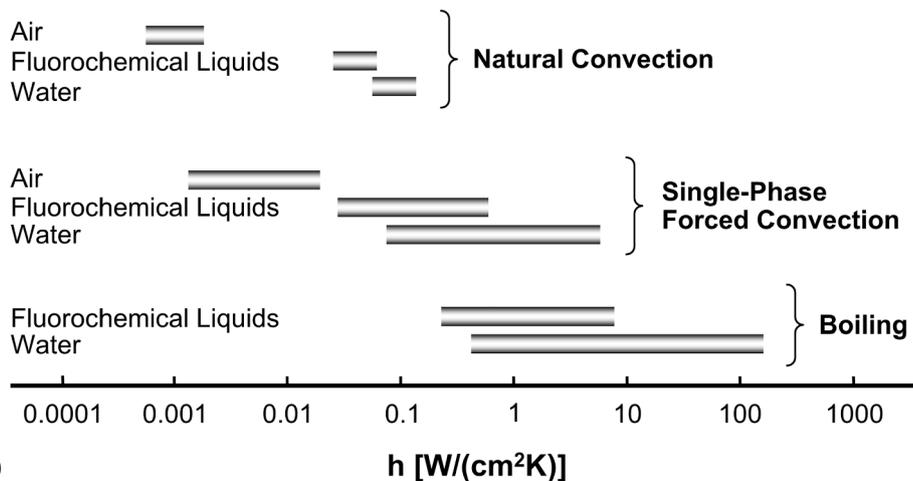
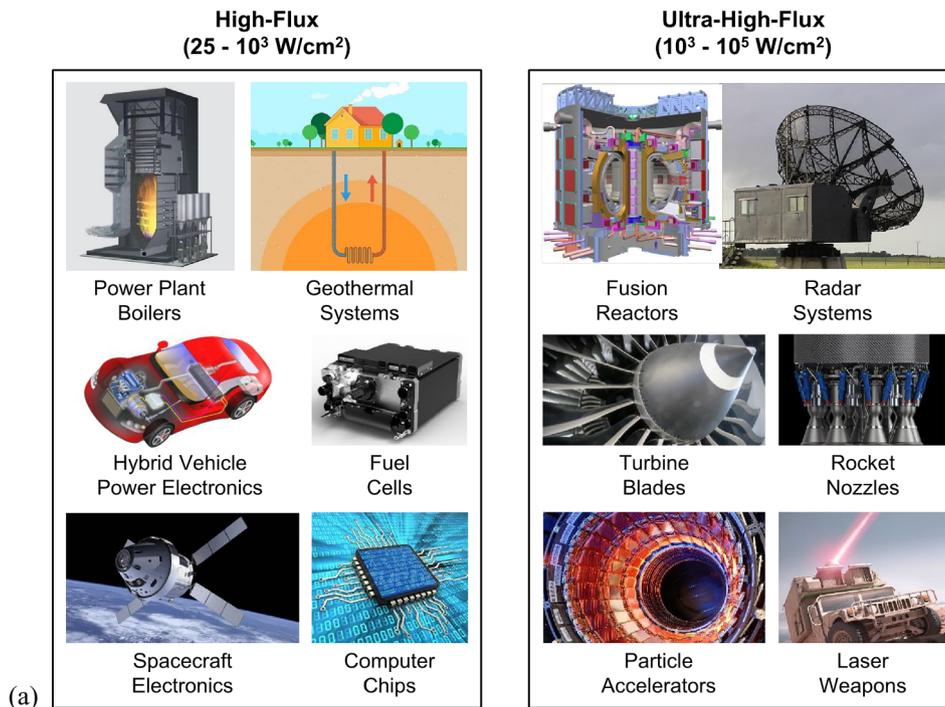


Fig. 1. (a) Examples of high-heat-flux and ultra-high-heat-flux applications. (b) Ranges of heat transfer coefficients attainable with different coolants using natural convection, single-phase forced convection, and boiling.

the recent shift to liquid-to-vapor phase-change cooling schemes. Unlike single-phase liquid cooling schemes, which rely entirely on heat removal by raising the coolant’s sensible heat, two-phase schemes capitalize on both sensible and latent heat of the coolant, thereby allowing far greater amounts of heat removal while simultaneously

maintaining relatively low device temperatures. This fact is reflected in the large heat transfer coefficients attainable with two-phase (boiling) schemes, Fig. 1(b), as compared to natural convection and single-phase forced convection.

Developing two-phase cooling schemes for both high-heat-flux and

ultra-high-heat flux applications has been a major impetus for research at the Purdue University Boiling and Two-Phase Flow Laboratory (PU-BTFFL) spanning well over three decades [1,2]. These schemes include pump-free cooling solutions using capillary-driven devices (heat pipes, capillary pumped loops, and loop heat pipes) [3] and pool boiling thermosyphons [4], as well as solutions involving use of a mechanical pump, which include falling film [5], channel flow boiling [6–8], mini/micro-channel flow boiling [9–12], jet-impingement [13] and spray [14–16], as well as hybrid cooling schemes combining the merits of mini/micro-channel flow boiling and jet impingement [17]. Other important works promoting two-phase cooling include nanofluid boiling by Wen and Ding's group [18] and Buongiorno's group [19], boiling on microporous surfaces by El-Genk's group [20], micro-pin-fin surfaces by Honda and Wei's group [21], micro and nanostructured surfaces by Joshi's group [22], hierarchically structured surfaces by Wang's group [23], porous surface by You's group [24] and Tang's group [25], and evaporation/boiling in capillary wicks by Peterson and Li's group [26].

1.2. Objectives of surface augmentation (Enhancement) in pool boiling and flow boiling

Given the recent sharp increases in heat dissipation in many modern applications, it is often difficult to achieve safe operation even when using the above-referenced two-phase cooling schemes. This has motivated many researchers to pursue surface augmentation as a means to ameliorate cooling performance of basic two-phase cooling schemes. And, while surface augmentation can be achieved using a variety of conventional macroscale techniques, recent research has been focused mostly on microscale and nanoscale methods, given the stringent volume and weight requirements in most modern applications. The present study is focused entirely on nanoscale surface augmentation methods.

To explore cooling benefits of surface augmentation, it is important to briefly present the dominant heat transfer regimes in both pool boiling and channel flow boiling. These regimes are clearly identifiable using a conventional boiling curve, Fig. 2, which depicts variation of heat flux from heating surface to coolant with wall superheat (wall temperature minus liquid saturation temperature). Shown are the four primary heat transfer regimes: single-phase liquid, nucleate boiling, transition boiling, and film boiling regime. These regimes are demarcated by three important transition points: onset of boiling (ONB) (incipient boiling), critical heat flux (CHF) (or Leidenfrost point) [27]. Most modern applications of interest involve high-flux dissipation and simultaneously maintaining low surface temperatures. These requirements are met within the nucleate boiling regime; the primary function of the cooling system is to maintain operation above ONB but safely below CHF. CHF is arguably the most important design parameter for a two-phase cooling scheme, as exceeding this limit would culminate in sharp escalation in wall temperature as operation transitions (for heat-flux controlled situations) from the highly cooling efficient nucleate boiling to the very cooling deficient film boiling.

One application where heat transfer enhancement is highly desirable is thermal management in space systems. Absence of gravity can compromise cooling performance by virtue of unusually low CHF [28–32]. A variety of enhancement methods are being sought to ameliorate CHF, including use of both nanofluids and surface augmentation.

Depending on application, there are several specific objectives for surface augmentation scheme, most important of which are (a) initiating ONB at lower wall heat flux and lower superheat, (b) reducing wall temperature excursion and sharp temperature drop sometimes encountered at ONB (especially for low contact angle fluids), (c) increasing nucleate boiling heat transfer coefficient, (d) ameliorating CHF, and (d) mitigating system instabilities.

But, aside from these well-established objectives for any surface augmentation scheme, recent findings point to an important emerging

concern, especially in conjunction with nanoscale schemes, stemming from serious time-dependent variations in cooling performance, which might endanger both reliability and durability of the entire cooling system. These performance variations are the key focus of the present study. The popularity of various nanoscale schemes in boiling enhancement can be ascribed to their great abilities to improve both wickability and wettability of the boiling surface, which point to the importance of new developments in nanomaterial science.

1.3. Objective of present review

The present review will provide an assessment of pool boiling and channel flow boiling heat transfer enhancement using nanoscale techniques. Key topics discussed are boiling heat transfer of nanofluids and enhanced surfaces with a variety of nanoscale structures. However, emphasis in the review will be placed on assessment of cooling reliability of nanofluids and nanoscale surface modifications based on recent findings showing time-dependent variations of boiling performance. Other useful information concerning heat transfer enhancement can be found elsewhere: pool boiling and flow boiling using nanofluids [33,34] and various scale surface modifications [27,35]. This review will be concluded with recommendations concerning future work that is needed to address these important issues.

2. Boiling enhancement with nanofluids

2.1. Nanofluid pool boiling

2.1.1. Description of nanofluids

Nanofluids are heat transfer fluids containing nanoparticles (typically 1–100 nm in size) that are uniformly and stably suspended in a liquid [36]. A large variety of nanoparticle materials have been proposed for inclusion in nanofluids, including chemically stable metals, metal oxides, and various forms of carbon. However, nanofluids were not examined in conjunction with boiling until 2003, before which efforts were focused on understanding their thermal properties and single-phase heat transfer characteristics.

Nanofluids are prepared using either one-step or two-step methods [36]. In a typical two-step method, nanoparticles are initially produced in the form of dry powder by physical or chemical treatment, followed by dispersion of the powder into the base liquid. A primary concern with the two-step method is potential aggregation of nanoparticles in

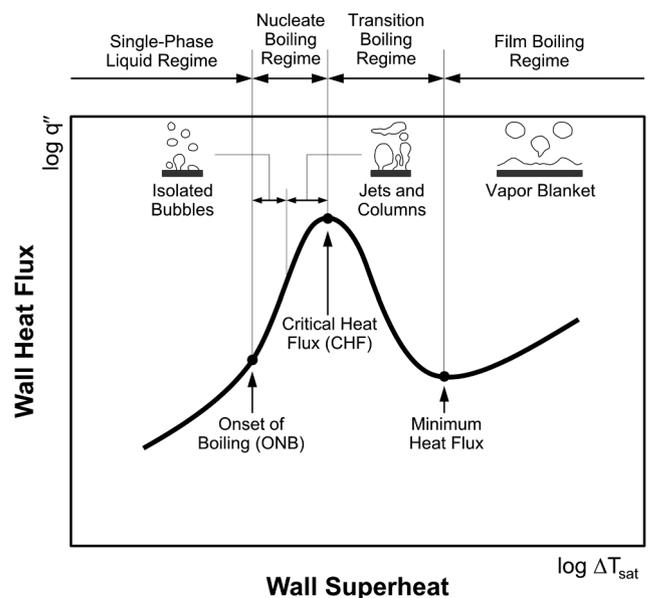


Fig. 2. Boiling curve and dominant heat transfer regimes and transition points.

the fluid. In contrast, synthesis and dispersion of nanoparticles into the base liquid in the one-step method are achieved simultaneously, providing better dispersability and stability than two-step methods. Fig. 3 shows TEM images of water-based carbonic nanofluid using one-step method and alumina nanofluid using two-step method [37].

In general, improper preparation of the nanofluid may lead to nanoparticle sedimentation and creation of undesirable zones of high thermal resistance. Stability and good dispersion are two key requirements for nanofluids that can be achieved by (a) changing solution pH with acid addition (to keep nanoparticles away from their isoelectric point), (b) adding surfactants and/or dispersants, and (c) ultrasonic vibration or electrostatic stabilization.

2.1.2. Boiling heat transfer coefficient trends

Different trends have been reported regarding nanofluid pool boiling heat transfer. For example, Wen and Ding [38] showed water-based Al_2O_3 nanofluids enhance nucleate boiling heat transfer coefficient, Fig. 4(a), and the enhancement improves monotonically with increased nanoparticle concentration. Similar enhancement was reported by Ali *et al.* [39] using water-based TiO_2 nanofluids. Soltani *et al.* [40] and Karimzadehkhoei *et al.* [41] found the degree of enhancement varies greatly with both nanoparticle material and concentration.

In contrast, You *et al.* [42] found no appreciable difference in pool boiling heat transfer performance between water-based 0.001–0.05 g/l Al_2O_3 nanofluid and pure water, as shown in Fig. 4(b). And many investigators even showed degradation in nucleate boiling performance resulting from addition of nanoparticles into base liquid. Examples include works by Bang and Chang [43] and Sarafraz *et al.* [44], who both reported water-based Al_2O_3 nanofluids decrease nucleate boiling heat transfer coefficient compared to pure water, as shown in Fig. 4(c). This decrease has been attributed to combined effects of nucleation site blockage and surface thermal resistance created by the nanoparticle deposition layer [45], and to reduced surface roughness brought about by nanoparticle trapping in the surface [46].

These serious inconsistencies in nanofluid impact on pool boiling heat transfer have been attributed to a number of factors, including (a) instability caused by sedimentation of non-homogenous nanofluid [38], (b) layering of agglomerates on the heating surface [47], (c) dispersants and/or surfactants used to stabilize nanoparticle suspension [48], (d) relative scale between initial surface roughness and nanoparticle size [43], and (e) thermal measurement technique as well as both orientation and size of heating surface [49].

2.1.3. Critical heat flux trends

Despite the conflicting results concerning nanofluid pool boiling heat transfer coefficient, almost all investigations point to enhancement of pool boiling CHF with nanofluids. You *et al.* [42] were the first to report significant improvement in CHF (up to 200%) using water-based 0.005 g/l Al_2O_3 nanofluid compared to pure water, but no enhancement was achieved when concentration exceeded 0.01 g/l, as shown in Fig. 5(a). Sulaiman *et al.* [50] showed CHF enhancement is virtually independent of particle material for water-based Al_2O_3 , TiO_2 and SiO_2 nanoparticles and concentrations of 0.04–1 g/l. However, Ham *et al.* [51], Dadjoo *et al.* [49], and Suriyawong and Wongwises [52] all showed best CHF enhancement is achieved at a particular optimum nanoparticle concentration.

Several mechanisms have been proposed regarding CHF enhancement with nanofluids. Kim *et al.* [53] suggested the enhancement is closely associated with modification to surface topography resulting from the nanoparticle coating. They confirmed this hypothesis by superior CHF measured with pure water boiling on NiCr surface initially coated by nanoparticles compared to nanofluid boiling on bare NiCr surface, as shown in Fig. 5(b). Another widely accepted mechanism for the CHF enhancement is improved wettability during boiling [54]. Yet another mechanism is one proposed by Hegde *et al.* [55], who reported CHF enhancement with water-based Al_2O_3 nanofluid is the outcome of

formation of porous deposition layer with abundant microcavities. And Sefiane [56] reported that presence of nanoparticles in the liquid promoted pinning of meniscus contact line on the surface, which serves to enhance CHF by hindering the spread of dry spots. Kim and Kim [57] observed CHF enhancement with nanofluids was accompanied by a decrease in contact angle, but CHF continued to increase after contact angle reached a smaller threshold value; they attributed the enhancement to improved capillary wicking. Kim *et al.* [58] proposed the nanoparticle layer improves stability of the evaporating microlayer or meniscus beneath a growing bubble, thus inhibiting irreversible growth of hot/dry spots even at high wall superheat, and therefore improves CHF.

Benefits of CHF enhancement using nanofluids might be of value to in-vessel retention (IVR) as one-off severe accident management strategy for light-water reactors. However, nanofluids are difficult to use in heat-flux-controlled applications (e.g., electronics cooling) due mainly to time-dependent changes to boiling behavior, as well as to several other practical concerns that will be addressed in later sections. Moreover, despite the widely accepted mechanism of improved surface wettability by the nanoparticle deposition layer as primary cause for CHF enhancement, there are uncertainties and contradictory findings concerning other possible mechanisms and influences of relevant parameters. For example, nanoparticle material and size have been shown to impact surface micro/nanostructure.

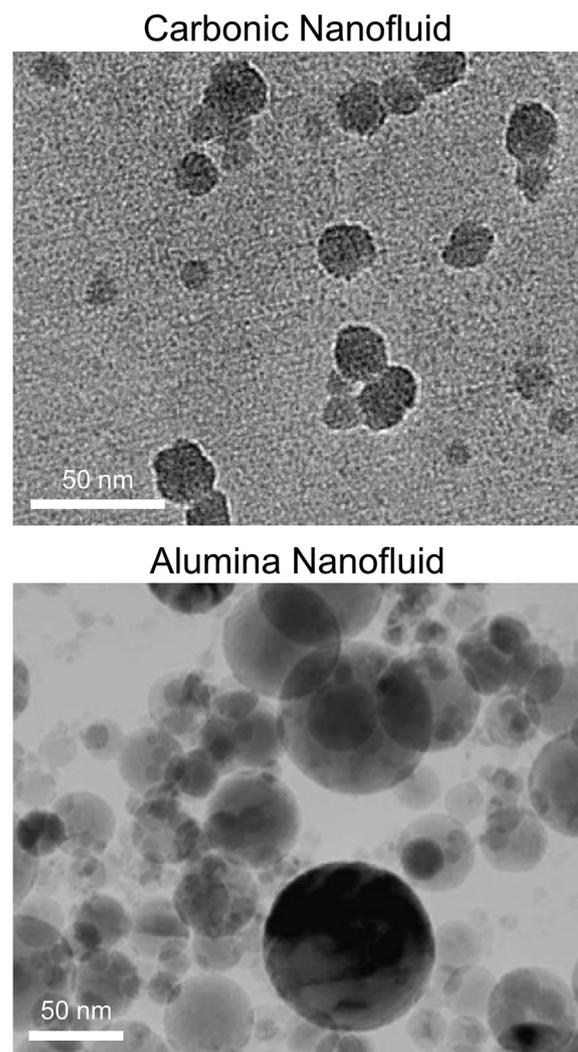
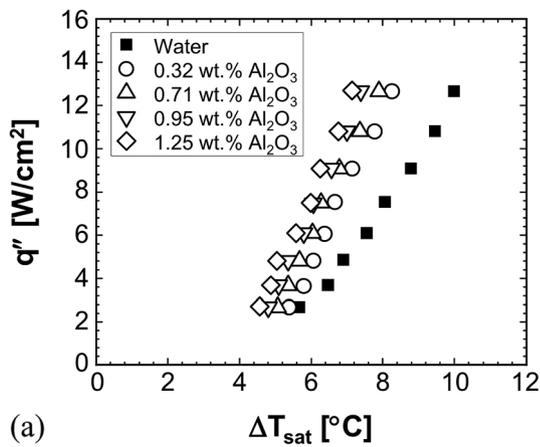
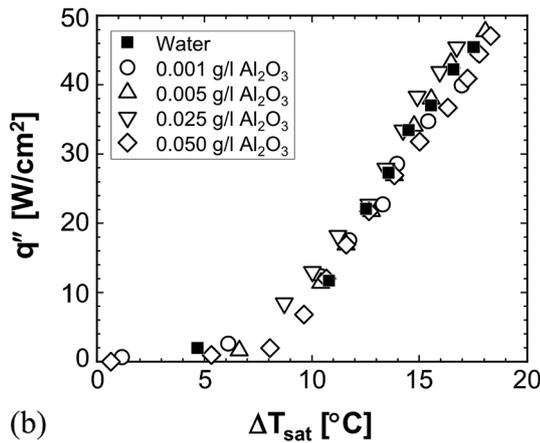


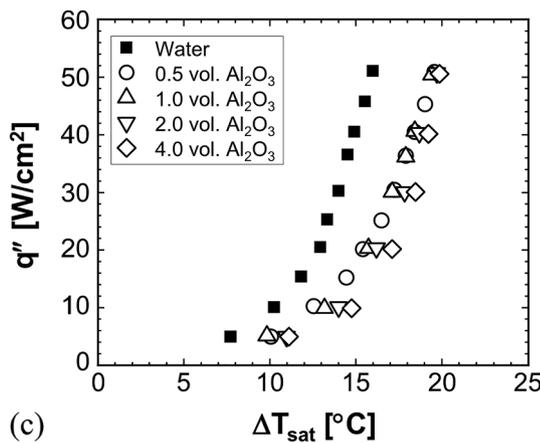
Fig. 3. TEM images of water-based carbonic nanofluid formulated using one-step method and alumina nanofluid using two-step method, adapted from Kim *et al.* [37].



(a)



(b)



(c)

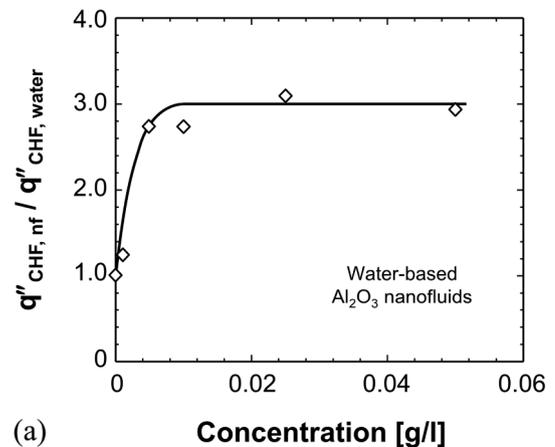
Fig. 4. Comparison of pool boiling heat transfer data for water-based Al_2O_3 nanofluids and water, showing (a) appreciable enhancement (adapted from Wen and Ding [38]), (b) no measurable change (adapted from You *et al.* [42]), and (c) appreciable deterioration in heat transfer coefficient (adapted from Bang and Chang [43]).

2.2. Nanofluid channel flow boiling

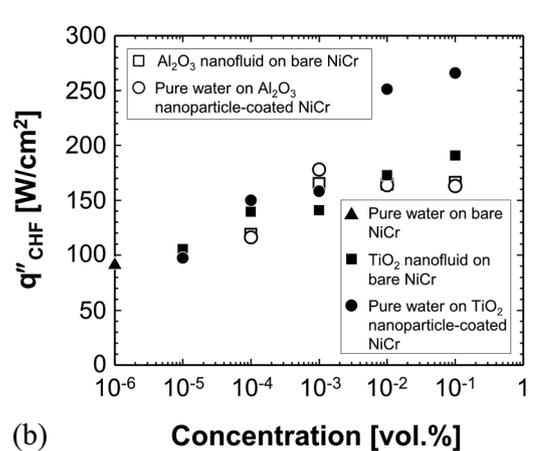
2.2.1. Boiling heat transfer coefficient trends

Nanofluid flow boiling is fundamentally different from nanofluid pool boiling in that, with the former, moving fluid might wash away nanoparticles deposited on the heating surface. Unfortunately, most of studies on nanofluid channel flow have been focused on single-phase heat transfer. One major reason for shying away from channel flow boiling is risk of blockage by nanoparticle agglomeration, especially in micro-channels.

Among the relatively few studies addressing nanofluid channel flow boiling, there is consensus that nanofluids do enhance nucleate boiling



(a)



(b)

Fig. 5. (a) Ratio of CHF for water-based Al_2O_3 nanofluid to that for pure water, adapted from You *et al.* [42]. (b) Comparison of CHF enhancement for nanofluids on bare surface and for pure water on nanoparticle-coated surface, adapted from Kim *et al.* [53].

heat transfer, however inconsistencies exist regarding data trends. Wang and Su [59] found that the enhancement with water-based Al_2O_3 nanofluid in a 6-mm diameter tube is highly dependent on both nanoparticle concentration and mass velocity. On the other hand, Zhang *et al.* [60] showed a dependence on vapor quality and mass velocity for R123-based carbon nanotube (CNT) nanofluid in a 9-mm diameter tube; which implies the enhancement is also a function of axial location along the tube [61], as shown in Fig. 6(a). Diao *et al.* [62] examined flow boiling of R141b-based Al_2O_3 nanofluid along a rectangular micro-channel with $0.25 \times 0.5 \text{ mm}^2$ cross-section, and showed existence of an optimal nanoparticle concentration below which heat transfer is improved because of increased nucleation density on the porous coating, but above which heat transfer is compromised because of increased thermal resistance.

One important drawback often cited in use of nanofluids is increased pressure drop [64]. Furthermore, Duursma *et al.* [65] showed that two-phase pressure drop for ethanol-based Al_2O_3 nanofluid flow in a rectangular channel is unstable and fluctuates with frequency and amplitude that are concentration dependent.

Contrary to the majority of studies reporting positive impact of nanofluids on heat transfer enhancement, several studies reported no enhancement, or even deterioration, in heat transfer performance. For example, Kim *et al.* [66] showed no appreciable change in heat transfer coefficient for flow boiling of water-based Al_2O_3 , ZnO, and diamond nanofluids along a 5.53-mm diameter tube. And Afrand *et al.* [67] reported deterioration in heat transfer coefficient along a 10-mm diameter tube for water-based TiO_2 nanofluid compared to that for pure water. Sarafraz and Hormozi [68] investigated water-based CuO

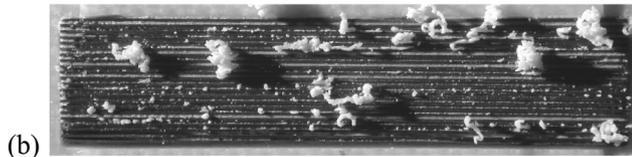
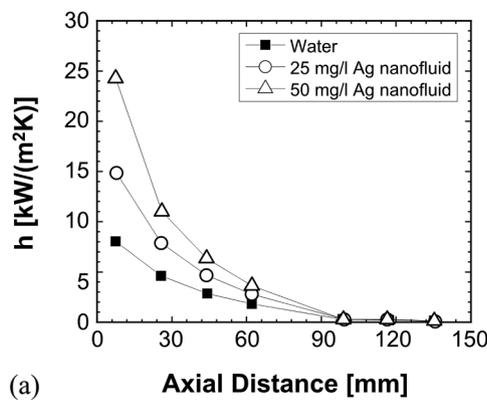


Fig. 6. (a) Heat transfer coefficient for water-based Ag nanofluid flow along a rectangular channel with 800- μm hydraulic diameter, adapted from Chehade *et al.* [61]. (b) Images capturing deposition and adhesion of Al_2O_3 nanoparticles in $215 \times 821 \mu\text{m}^2$ rectangular channels, adapted from Lee and Mudawar *et al.* [63].

nanofluid flow boiling along an annulus with 30-mm hydraulic diameter, and showed fouling rate increases with increasing nanoparticle concentration, a possible reason for observed deterioration in heat transfer performance.

Lee and Mudawar [63] showed severe wall deposition of nanoparticles once boiling commences in water-based Al_2O_3 nanofluid along $215 \times 821 \mu\text{m}^2$ rectangular channels. This phenomenon is clearly manifest in Fig. 6(b), and points to rapid deposition of nanoparticles into relatively large clusters along the channel walls. From a practical point of view, the clustering effect would prevent operation beyond onset of boiling, and might result in catastrophic failure of the cooling system. On the other hand, Xu and Xu [69] showed flow boiling of 0.2 wt% Al_2O_3 water-based nanofluid along a single $0.1 \times 0.25 \text{mm}^2$ rectangular channel is not accompanied by nanoparticle deposition. They also reported the nanoparticles contribute stability to the boiling process, reduce pressure drop, and enhance heat transfer.

Overall, it is difficult to come out with definitive assessment of any enhancement or deterioration in nanofluid flow boiling performance in channels (especially micro-channels), given the differences in criteria adopted in the performance assessment. As noted by Liang and Mudawar [34], enhancement inferences from experimental studies are often based on specific criteria, such as equal pumping power, equal flow velocity, equal flow rate, and equal Reynolds number, which may lead to conflicting findings concerning enhancement trends. For example, while the criterion of equal Reynolds number often points to better performance with nanofluid compared to base fluid, the inferred enhancement might actually be the outcome of increased flow velocity at higher Reynolds number, rather than due to nanofluid use.

2.2.2. Critical heat flux trends

Given the afore-mentioned blockage of small channels by nanoparticles during flow boiling, especially at high heat fluxes, investigators have limited their experiments to relatively low heat fluxes, well below CHF. This explains why CHF data for nanofluid flow boiling are quite sparse compared to those for nanofluid pool boiling. Among studies that did yield flow boiling CHF data, Kim *et al.* [70] and Ahn *et al.* [71] showed CHF enhancement with water-based Al_2O_3 , ZnO, and diamond nanofluids in 5.53-mm and 8.7-mm diameter tubes, respectively, which was attributed to increased surface wettability. Lee *et al.*

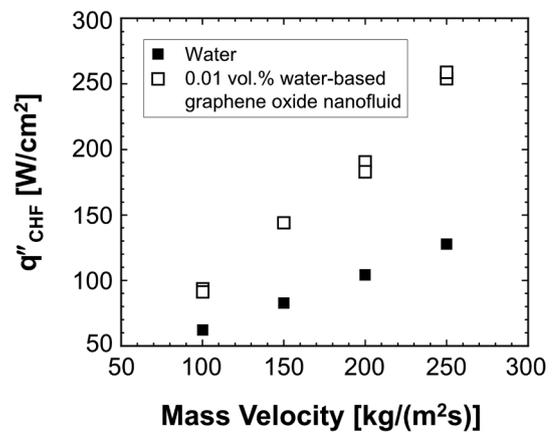


Fig. 7. Variation of CHF versus mass velocity for flow boiling of pure water and water-based 0.01 vol.% graphene oxide nanofluid versus mass flux along a 1.27-mm diameter tube, adapted from Lee *et al.* [72].

[72] arrived at a similar conclusion for water-based 0.01 vol.% graphene oxide nanofluid flow in a 1.27-mm diameter tube, as shown in Fig. 7. The capacity of nanofluids to improve CHF along a single 510- μm diameter channel was observed by Vafaei and Wen [73], who pointed out that thickness of the nanoparticle deposition layer is quite small (few hundred nanometers) compared to the hydraulic diameter, implying the deposition layer might not cause appreciable flow obstruction.

Kim *et al.* [74] compared flow boiling performances of water-based Al_2O_3 nanofluid and pure water in an Al_2O_3 nanoparticle-coated 7.745-mm diameter tube. The two cases yielded virtually equal CHF values, but about 80% higher than for pure water flowing in an untreated tube, which led them to conclude that the CHF enhancement with nanofluids was the result of nanoparticle deposition along the tube's inner wall. Park and Bang [75] found no improvement in surface wettability from nanoparticle deposition, which contradicts findings by many previous investigators. Instead, they suggested CHF enhancement in nanofluid flow boiling is the outcome of improved thermal activity resulting from delayed ability of bubbles to form hot spots.

Overall, the afore-mentioned studies provide very contradictory findings concerning impact of nanofluids on both pool boiling and flow boiling heat transfer coefficient. Some of these contradictions might be rooted in dependence of potential enhancement on a broad number of influences, such as those of nanoparticle material, size, shape (or aspect ratio), and concentration, let alone those of other parameters important to flow boiling of even pure liquids, such as vapor quality, channel shape and dimensions, mass velocity, and subcooling. And despite inferences by some concerning positive impact of nanofluids on both pool boiling and flow boiling CHF, buildup of the nanoparticle deposition layer on the heating surface after prolonged boiling and/or repeated boiling tests is inevitable, which may lead to catastrophic failure of the cooling device (especially for micro-channels) or even entire cooling system.

2.3. Time-dependent variations of nanofluid boiling performance

The dependence of nanofluid boiling heat transfer coefficient and CHF on boiling time as well as lack of repeatability of heat transfer performance have been reported for at least the past ten years, yet received far less attention than short term enhancement merits. Major relevant studies are summarized in Table 1. Clearly, special attention must be paid to findings from the few studies pointing to time-dependent variations of both repeated and long duration nanofluid boiling performance.

Kwark *et al.* [76], Wen [77] and Mourgues *et al.* [78] all reported continuous growth of nanoparticle deposition layer on the heating

Table 1
Literature related to time-dependent boiling performance of nanofluids.

Authors	Nanofluid type	Heating cycles or boiling duration	Surface change after repeated tests or boiling experiment	Time dependency of boiling performance
Kwark <i>et al.</i> [76]	Al ₂ O ₃ nanofluid	240 min	Nanoparticle layer grew with an upper thickness limit	Boiling heat transfer coefficient deteriorated but CHF remained constant
Mourgues <i>et al.</i> [78]	ZnO nanofluid	5 heating cycles	Nanoparticles deposition	Both surface temperature and CHF increased with repeated boiling cycles
Minakov <i>et al.</i> [79]	SiO ₂ nanofluid	25 min	Nanoparticles deposition	Initially CHF increased rapidly before subsiding thereafter
Hegde <i>et al.</i> [81]	CuO nanofluid	3 heating cycles	Increased surface roughness	CHF first deteriorated but was then enhanced
Zhang & Jacobi [83]	Al ₂ O ₃ nanofluid	30 min	Increased surface wettability	
Dareh <i>et al.</i> [84]	Al ₂ O ₃ nanofluid	10 heating cycles	Increased surface roughness	Boiling heat transfer coefficient deteriorated but CHF was enhanced within 4 runs. Further increase in number of boiling cycles had minor impact.
Park <i>et al.</i> [85]	Al ₂ O ₃ nanofluid	180 min	Nanoparticles deposition	CHF enhancement had a threshold of boiling duration
Sharma <i>et al.</i> [87]	ZnO nanofluid	100 s	Formation of porous and hydrophilic nanoparticle layer	CHF was enhanced with 10- and 100-s durations, but 1-s duration yielded no enhancement
Salari <i>et al.</i> [88]	Fe ₃ O ₄ nanofluid	1000 min	Fouling	No impact on nucleate boiling heat transfer coefficient
Chopkar <i>et al.</i> [89]	ZrO ₂ nanofluid	3 heating cycles	Smoothing of surface	Heat transfer coefficient deteriorated

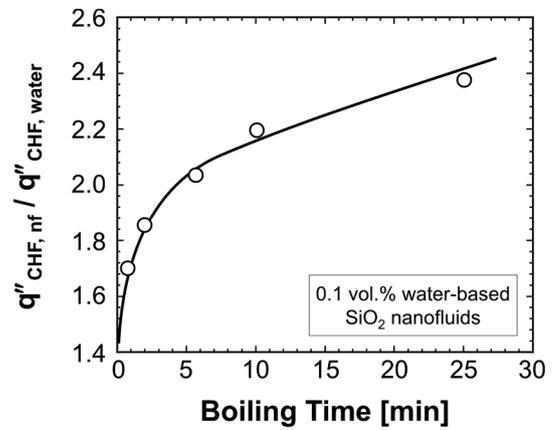


Fig. 8. Ratio of CHF for water-based SiO₂ nanofluid to that for pure water versus boiling time, adapted from Minakov *et al.* [79].

surface during pool boiling experiments renders nanofluid pool boiling highly time-dependent, influenced by both duration of experiment and heat flux, which is in sharp contrast with pool boiling of pure liquids. As the deposition layer has often been reported as a major contributor to the boiling enhancement, the highly transitory growth of the same layer constitutes an important drawback for all applications demanding high repeatability and predictability of cooling performance. And, while Kwark *et al.* did suggest existence of an upper limit for nanoparticle deposition thickness, Wen suggested unless active methods are employed to control the nanoparticle deposition, it is difficult to envision future application of nanofluid boiling as cooling solution in modern high-heat-flux applications.

Minakov *et al.* [79] also addressed lack of repeatability in CHF for nanofluid pool boiling. They showed that, aside from dependence on nanoparticle size and material and heater size, CHF exhibited significant dependence on boiling duration, Fig. 8, first increasing rapidly before subsiding thereafter. However, their experiments were limited to relatively short durations (no more than 25 min) and did not address long-term repeatability concerns. As improved wettability and/or wickability in the porous deposition layer are responsible for the enhanced CHF, Tetreault-Friend *et al.* [80] found that maximum CHF was dictated by both pore size and layer thickness.

Hegde *et al.* [81] investigated changes in surface topography resulting from two different types of pool boiling experiments: (a) exposing heating surface to many heating cycles in water-based CuO nanofluids with fixed volume concentration, and (b) exposing heating surface to a single heating cycle in CuO nanofluids with different volume concentrations. They observed changes in surface roughness resulting not only from concentration changes, but heating time as well.

Wen *et al.* [82] reported surface modification by nanoparticle deposition is an inherent phenomenon occurring after every pool boiling or flow boiling test. Consequently, any enhancement or deterioration in nanofluid boiling heat transfer performance must be a function of the number and frequency of usage of the heating surface, and this might explain contradictory findings concerning nucleate boiling heat transfer coefficient trends for nanofluids. In another study, Zhang and Jacobi [83] studied effects of pool boiling duration on surface wettability for water-based Al₂O₃ nanofluids. They showed increasing boiling time has a positive influence on wettability, the first 10 min of which being the most effective in terms of nanoparticle deposition.

Dareh *et al.* [84] studied effects of boiling time and repeated boiling cycles on pool boiling performance of water-based Al₂O₃ nanofluids. After four test cycles, they measured a 13% reduction in heat transfer coefficient and 50% enhancement in CHF compared to pure water boiling on a clean polished surface. But further increases in number of boiling cycles had minor impact on boiling performance. And, by increasing boiling time in a single test from 5 to 10 min, surface

roughness increased by about 150%, compared to only an additional 8% as boiling time was increased to 15 min. These results point to the majority of the nanoparticles being deposited during the first 10 min of boiling, yet they do not provide guidance on very long duration effects.

Park *et al.* [85] noted existence of a threshold pool boiling duration exceeding which CHF enhancement gradually abated because of decreasing porosity of the deposition layer. As shown in Fig. 9(a), this duration threshold corresponds to the time contact angle on the heating surface being deposited by nanoparticles achieves constant value. Yang and Liu [86] compared pool boiling performance of ‘functionalized’ SiO₂ nanofluid, treated by grafting silanes to the nanoparticle surfaces, to that of conventional nanofluids without the treatment. The functionalization improved both dispersion and stability of the nanoparticles, evidenced by absence of sedimentation over a 12-month period even for concentrations as high as 10 wt%. Despite better stability and dispersion, the functionalized nanofluid achieved only slight enhancement in the nucleate boiling heat transfer coefficient, and CHF remained virtually unchanged. Sharma *et al.* [87] addressed effects of boiling time on pool boiling of water-based ZnO nanofluids. For 10- and 100-s durations, a porous and hydrophilic nanoparticle layer formed, which yielded measureable CHF enhancement, Fig. 9(b), while a 1-s test yielded no deposition or CHF enhancement. Salari *et al.* [88] reported nanoparticle deposition (which they termed ‘fouling’) increased appreciably with increasing pool boiling time but had no impact on nucleate boiling heat transfer coefficient, behavior they explained by bubble interactions causing partial detachment of the porous deposited layer.

Using water-based ZrO₂ nanofluids, Chopkar *et al.* [89] showed that, while even low nanoparticle concentrations do provide initial improvement in nucleate pool boiling performance, repeated boiling tests using the same heating surface gradually reduce the surface roughness, causing a commensurate decrease in heat transfer coefficient, even below that of base liquid (water).

Aside from boiling time, nanofluid storage time prior to the boiling tests is another important parameter influencing boiling performance. This is evident from experiments by Lee *et al.* [90], who used a two-step method to produce water-based Fe₃O₄ nanofluid, and showed that pool boiling CHF enhancement could be ensured for at least 12 months. However, this enhancement did decrease gradually with increasing nanofluid storage time, presumably because of gradual agglomeration of nanoparticles compromising stability of the nanofluid.

Flow boiling of nanofluids is far more concerning, given, other than the heating surface, the large number of flow loop components that could be adversely impacted by the nanoparticles. This is especially the case for micro-channels, where studies [63,91] have shown severe wall deposition of nanoparticles within the cooling module once boiling commences, even within a short duration from boiling test initiation. There are also long term concerns even when flow boiling in macro-channels, which are brought about by clustering, sedimentation, and precipitation of nanoparticles. These effects are expected to promote clogging of flow passages (especially at high temperatures) within the cooling module as well as cooling loop’s pump, filter, flow regulators, *etc.*, eventually triggering catastrophic failure of the entire cooling system. Yet another concern is gradual damage (and ultimate failure) of temperature-sensitive heating surfaces (such as electronic and power devices) by erosion caused by the nanoparticles or acidic stabilizer, or by surface buildup of nanoparticle deposition layer. Other concerns (for both pool boiling and flow boiling) are difficulty preparing the nanofluid and associated high cost, as well as lack of quality assurance in nanofluid experiments. Detailed information related to these issues can be found in the authors’ previous review [33].

Overall, future studies must adopt more consistent practices when exploring potential benefits of nanofluids to nucleate boiling heat transfer coefficient and CHF. Key among those practices is assessment of the effects of boiling time and repeated boiling tests, and ensuing micro/nanostructure of the nanoparticle deposition layer. This would

both enable better capture of dominant enhancement mechanism(s) of improved wettability and/or wickability and provide a convincing case for or against use of nanofluids in practical cooling situations. It is proposed boiling tests be performed over varying durations, from minutes to days or even months, in order to gauge gradual changes in boiling performance and relate those changes to observable changes in both surface deposition layer morphology and nanofluid itself. Equally important is the need to repeat boiling experiments using the same batch of nanofluid and same heating surface. These repeatability experiments serve to capture any aging effects, which are inherent to boiling even certain pure liquids and surface materials [92]. These important testing practices are integral to the authors’ proposed testing strategy, which is presented in the conclusions section of this study. Besides, two important effects often overlooked in nanofluid boiling studies are those of substrate material and surface finish, as nanoparticle deposition is influenced by surface chemistry, at least at the beginning of the deposition. These issues warrant further investigation in future studies. It is also proposed that future studies employ non-intrusive, high-resolution diagnostics to elucidate the mechanisms that lead to enhancement or impairment of boiling heat transfer and CHF.

3. Boiling enhancement by nanoscale surface modification

3.1. Pool boiling enhancement

3.1.1. Nanotube and nanowire

Modifying surface texture is a well-established strategy to enhancing heat transfer performance including not only convective heat transfer but radiation as well [93]. Earlier surface texture enhancement schemes involved use of macro-, mini- and micro-scale modifications. However, with evolution of effective nano-scale fabrication techniques, interest in recent years has shifted to nano-scale surface modification strategies. These include use of nanotubes, nanowires, and nano-coating.

Nanotube is defined as extremely thin tube of carbon or TiO₂ with outer diameter ranging from 1 to 100 nm and 1 to 50 μ m length, and having extraordinarily high thermal conductivity and mechanical properties. And nanowire is defined as a nanoscale rod having a diameter of tens of nanometers and large length to diameter ratio [35].

Ujreh *et al.* [94] investigated pool boiling performance of FC-72 on a heating surface coated with carbon nanotubes (CNTs), Fig. 10(a). As

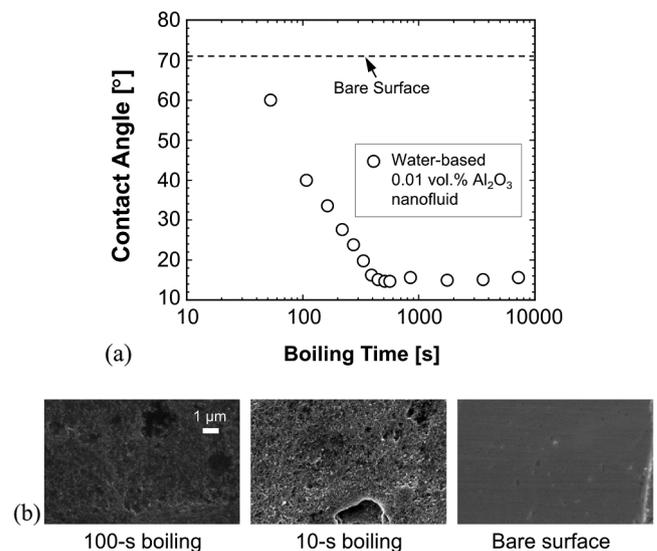


Fig. 9. (a) Variation of contact angle of nanoparticle deposited surface with boiling time for water-based Al₂O₃ nanofluid, adapted from Park *et al.* [85]. (b) SEM images of boiling surface after different boiling times for water-based ZnO nanofluid, adapted from Sharma *et al.* [87].

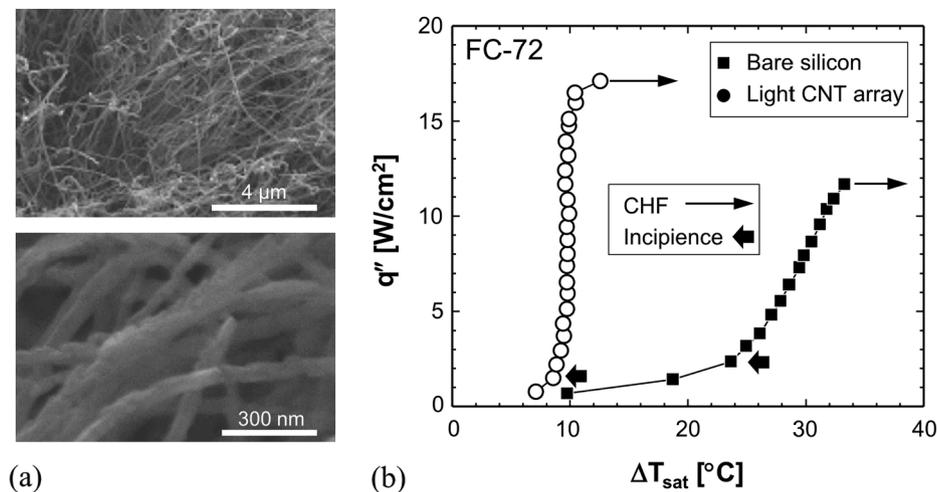


Fig. 10. (a) SEM images of silicon substrate coated with light CNT arrays, and (b) comparison of boiling curves of FC-72 for bare and fully CNT-coated silicon surfaces. Adapted from Ujereh *et al.* [94].

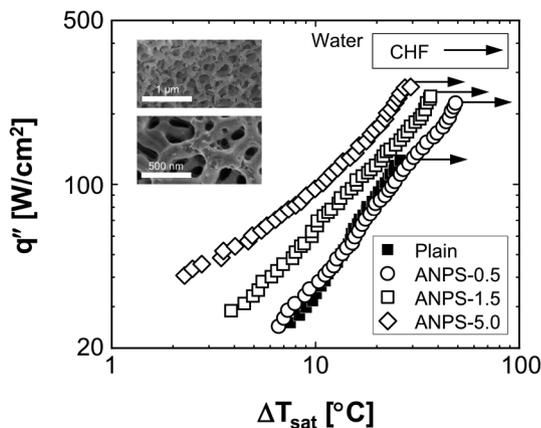


Fig. 11. Water pool boiling curves of Al_2O_3 nanoporous surface (ANPS) with SEM images inserted. Adapted from Zhang and Kim [103].

shown in Fig. 10(b), their tests showed all desired enhancement benefits of surface modification: earlier ONB, greatly enhanced nucleate boiling heat transfer coefficient, and improved CHF. They attributed these benefits to a large number of submicron cavities created by intertwining CNT structures. Sathyamurthi *et al.* [95] found boiling performance for PF-5060 was independent of CNT coating height, but Bertossi *et al.* [96] observed enhancement in water nucleate boiling heat transfer coefficient as CNT length was increased from 3 to 10 μm . On the other hand, using a vacuum filtration process to form randomly-oriented CNT coating, Seo *et al.* [97] reported deterioration in nucleate boiling heat transfer coefficient for water, which they attributed to decreased surface roughness on the CNT-coated surface, but CHF did increase compared to an uncoated surface.

Chen *et al.* [98] reported over 100% enhancement in both nucleate boiling heat transfer coefficient and CHF for water pool boiling on silicon surfaces coated with arrays of silicon nanowire and copper nanowire, which they attributed to increased nucleation site density, superhydrophilicity, and enhanced capillary pumping. Superior ability to increase water CHF for the surface with silicon nanowire arrays was reported by Shim *et al.* [99], who attributed the enhancement to improved liquid spreadability and increased volumetric wicking, respectively. Using water and FC-72, respectively, Shi *et al.* [100] and Kumar *et al.* [101] reported dependence of pool boiling enhancement on nanowire height and diameter, and Im *et al.* [102] demonstrated 2- μm nanowire height yielded the highest CHF enhancement for PF-5060.

Coating the heating surface with nanotubes or nanowires may

increase number of nucleation cavities and therefore improve nucleate boiling heat transfer coefficient. But, depending on nanotube material, diameter and length, and coating thickness, surface roughness may even decrease, adversely influencing nucleate boiling performance. Any enhancement or deterioration of heat transfer coefficient is the outcome of competition between initial surface roughness and coating properties. Overall, nanotube or nanowire coatings have been proven effective at improving capillary wicking, thereby increasing ability to replenish liquid beneath vapor bubbles and therefore enhance CHF.

3.1.2. Nano-coating

Nano-coating techniques used to improve pool boiling heat transfer can be divided into two major groups: nanoporous and nanofilm. The main purposes of nanoporous coating are to regulate bubble dynamics during the boiling process and enhance capillary wicking, while nanofilm coating aims to modify surface wettability.

Using anodic oxidation technique, Zhang and Kim [103] developed a 3-D interconnected Al_2O_3 nanoporous surface (ANPS) on aluminum alloy substrate, and demonstrated significant enhancement in both nucleate boiling heat transfer coefficient and CHF for water, as shown in Fig. 11, where ANPS-0.5, -1.5 and -5.0 represent nanoporous structures after 0.5, 1.5 and 5 h of anodization, respectively. Vemuri and Kim [104] showed a 30% reduction in incipience superheat for FC-72 with nanoporous alumina coating on an aluminum substrate. Lu *et al.* [105] produced nanoporous copper surfaces having 50–200 nm pores on copper substrate, and were able to achieve 63.3% reduction in incipience superheat and 172.7% enhancement in nucleate boiling heat transfer coefficient for water. Hendricks *et al.* [106] formed flower-like ZnO nanostructures on aluminum and copper substrates, and showed significant reduction (25 to 38 $^\circ\text{C}$) in nucleate boiling wall superheat, and up to 4-fold enhancement in CHF for water.

Forrest *et al.* [107] fabricated a polymer/silica nanoparticle film coating on nickel substrate using a layer-by-layer technique to improve nucleation and surface wettability, which showed over 100% enhancement in both nucleate boiling heat transfer coefficient and CHF for water. Feng *et al.* [108] formed alumina coating on a platinum substrate using atomic layer deposition. CHF for water increased with increasing coating thickness up to 20 nm because of improved wettability, but no further enhancement was observed above 20 nm. Seo *et al.* [109] used DC sputtering to form FeCrAl coating on stainless steel, and showed enhancement in pool boiling CHF for water resulting from increased surface roughness.

Aside from nanoporous and nanofilm, coating by boiling induced nanoparticle deposition has attracted considerable attention in recent

years, the goals of which being to increase surface wettability and modify surface micro/nanoscale morphology. Results for this type of coating for pure liquids often coincide with those of nanofluid boiling on bare surfaces. Kwark *et al.* [110] found enhancement of water boiling performance was independent of Al_2O_3 nanoparticle size, contradicting observations by Souza *et al.* [111], who demonstrated deposition of 10-nm Fe_2O_3 nanoparticles enhanced nucleate boiling heat transfer coefficient for HFE-7100, while 80-nm nanoparticles degraded performance. Sakashita [112] showed little difference in liquid–vapor interfacial behavior during water pool boiling between TiO_2 -coated and uncoated copper surfaces, but the former did enhance CHF by virtue of increased thickness of macrolayer beneath large bubbles. However, particle deposition strength and surface homogeneity issues in long duration boiling pose questions regarding viability of this coating scheme.

One important drawback of virtually all nano-coating studies is absence of findings on time-dependent aging, and potential eventual deterioration of both nucleate boiling heat transfer coefficient and CHF.

3.2. Flow boiling enhancement

3.2.1. Nanotube and nanowire

Kousalya *et al.* [113] enhanced water flow boiling in a $5.5 \times 17.5 \text{ mm}^2$ rectangular channel using CNT-coated copper surface that was exposed to low-intensity ultraviolet–visible excitation. Compared to non-illuminated results, average boiling incipience decreased by 4.6°C and heat transfer coefficient improved by 41.5% with light exposure, apparently the result of augmented hydrophilicity and nanoscale optothermal effects. Singh *et al.* [114] achieved better heat flux enhancement for water flow boiling from a CNT-coated horizontal heating surface in a $10 \times 10 \text{ cm}^2$ channel at lower flow rates and lower subcoolings.

Li *et al.* [115] showed experimentally that silicon nanowire coating in rectangular $250 \times 200 \mu\text{m}^2$ micro-channels enhances flow boiling heat transfer for water by initiating boiling at lower surface temperature and enhancing nucleate boiling heat transfer coefficient, as shown in Fig. 12. Yang *et al.* [116] examined HFE-7000 flow boiling in a $250 \times 220 \mu\text{m}^2$ rectangular micro-channel having both side and bottom walls coated with silicon nanowires. They achieved up to 344% enhancement in heat transfer coefficient for mass velocities ranging from 1018 to 2206 $\text{kg/m}^2\text{s}$, which they attributed to enhanced evaporation and nucleate boiling. Shin *et al.* [117] explored the impact of silicon nanowire coating on surface tension for flow boiling along a $8 \times 8 \text{ mm}^2$ rectangular channel. The surface tension changes were found highly fluid dependent. For example, contact angle for water decreased from 43.6° (for plain surface) to 4.5° , whereas that of FC-72 increased from 9.8° to 16.1° ; the latter led to CHF deterioration. Alam *et al.* [118] examined heat transfer attributes of water flow boiling on nanowire-coated surface along a 10-mm long $250 \times 220 \mu\text{m}^2$ rectangular channel. They reported CHF enhancement from improved wetting caused by relatively high surface tension force along the liquid–vapor interface.

It is concluded nanotube/nanowire-coated surfaces do provide multiple performance advantages, including earlier onset of boiling, enhanced nucleate boiling heat transfer coefficient, ameliorated CHF, and some suppression of flow instabilities. However, here too, no attention has been paid to time-dependent performance. Additionally, future work needs to address the import influence of inlet subcooling [119] when implementing these enhancement schemes.

3.2.2. Nano-coating

Ahn *et al.* [120] investigated water flow boiling in a 5-mm high rectangular channel along an Al_2O_3 nanoparticle deposited surface produced initially by nanofluid boiling. While improved wettability did enhance CHF, this surface both delayed onset of boiling and reduced flow boiling heat transfer coefficient. Kumar *et al.* [121] devised a spray

pyrolyzed Fe-doped Al_2O_3 - TiO_2 composite coating, which they used to explore potential heat transfer merits for water flow boiling along a $20 \times 0.4 \text{ mm}^2$ rectangular channel. They reported maximum enhancement levels of 52.39% and 44.11% for CHF and nucleate boiling heat transfer coefficient, respectively, at a mass velocity of 88 $\text{kg/m}^2\text{s}$. Çikim *et al.* [122] studied flow boiling in circular 249–908- μm tubes having inner walls modified by cross-linked pHEMA coatings with different thicknesses in the range of 50 to 150 nm. Using water as coolant, flow boiling heat transfer coefficient and CHF were enhanced by up to 126.2% and 29.7%, respectively, with increasing coating thickness providing monotonic improvements in both, as shown in Fig. 13. Nedaee *et al.* [123] employed initiated chemical vapor deposition to coat pPFDA on inner walls of circular 600- and 889- μm diameter micro-tubes with thicknesses ranging from 50 to 160 nm. This scheme produced appreciable enhancement in flow boiling heat transfer coefficient for water, which they attributed to increased number of active nucleation sites brought about by enhanced hydrophobicity and nanoporous surface morphology.

Despite the above-mentioned merits, it remains unclear whether nano-coatings can withstand violent bubble detachment at high mass velocities during long duration operation.

3.3. Time-dependent variations of boiling performance

Studies relating to repeatability and time-dependence of boiling performance are summarized in Table 2. It should be emphasized that initial surface coating using nanoparticle deposition by nanofluid boiling is highly prone to risk of nanoparticle detachment, which brings into question long-term sustainability of flow boiling performance. Evidence of this problem comes from works by Ahn *et al.* [71] and Lin *et al.* [124], who reported nanoparticle detachment becoming more severe with increasing heat flux in flow boiling and pool boiling, respectively. Choi *et al.* [125] performed subcooled water flow boiling experiments in a 10.92-mm diameter tube having Fe_3O_4 nanoparticle-deposited inner wall, which was formed by initial nanofluid boiling. Tests at relatively high mass velocities from 1000 to 5000 $\text{kg/m}^2\text{s}$ and 20–60 $^\circ\text{C}$ subcoolings yielded CHF enhancement by up to 40%, brought about by increased surface wettability. But, following the boiling experiments, Choi *et al.* [125] reported observing partial detachment of the deposited nanoparticles from the surface. Okawa *et al.* [126] performed experiments involving water pool boiling on TiO_2 nanoparticle coated surface in which CHF increased asymptotically with increasing boiling time. However, partial detachment of the nanoparticle layer was clearly observed in several boiling runs, which was the outcome of weak adhesion of nanoparticles to the surface, causing appreciable subsequent deterioration in CHF. Stutz *et al.* [127] showed weak

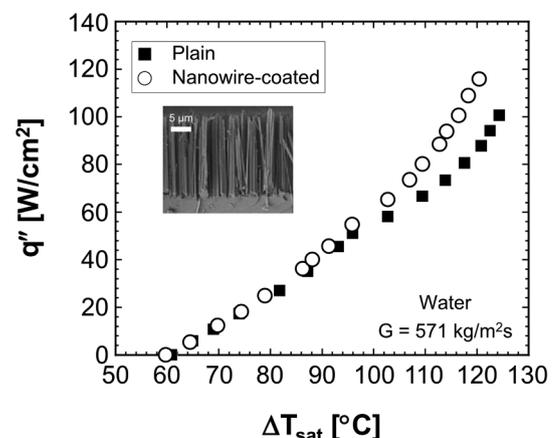


Fig. 12. Water flow boiling curve for nanowire-coated $250 \times 200 \mu\text{m}^2$ micro-channels, adapted from Li *et al.* [115].

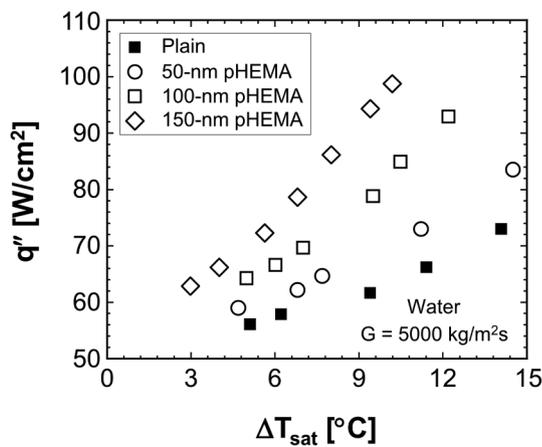


Fig. 13. Water flow boiling curves for pHEMA-coated 507- μm diameter microtube corresponding to different coating thicknesses, adapted from Çıkım *et al.* [122].

reproducibility of pentane pool boiling on a Fe_2O_3 nanoparticle layer formed initially by vigorous nanofluid pool boiling. As shown in Fig. 14, not only were the boiling data highly time dependent, but also non-monotonic.

Morshed *et al.* [128] coated inner walls of a copper 672- μm hydraulic diameter rectangular channel with Al_2O_3 nanoparticles using initial ethanol-based Al_2O_3 nanofluid boiling. They reported up to 39% in water flow boiling CHF, but at the expense of reduction in heat transfer coefficient compared to untreated channel walls. Perhaps the most important finding from their study is wash out of most of the loosely attached nanoparticles by the flowing liquid, with only a very thin layer of coating remaining on the surface; this residual layer was sustained for a finite number of flow boiling tests.

Lee *et al.* [129] used an anodizing technique to fabricate nanoporous structure on aluminum alloy surface upon which the heat transfer coefficient for water remained virtually unchanged following 500 h of pool boiling, but deteriorated after 900 h. They attributed the deterioration to the nanoporous layer wearing off from the surface after prolonged boiling. Follow-up work by Lee *et al.* [130] and Zhang *et al.* [131] addressed surface aging effects resulting from oxidation following prolonged pool boiling in water, which changed morphology of the nanoporous surface by formation of aluminum hydroxide surface deposits; these deposits resulted from filling of pores between nanostructures, which decreased the number of active cavities and therefore reduced the nucleate boiling heat transfer coefficient.

Using facile hot-dip galvanizing/dealloying process, Tang *et al.* [25] formed nanoporous copper surfaces having pores smaller than 200 nm on copper substrate. However, the 3-D nanostructure and porosity changed slightly over 100 h of boiling [105], and average porosity length scale changed from 20.0 to 43.8 nm after 160 h of boiling. Das *et al.* [132] formed TiO_2 nanostructures on copper substrate using electron beam evaporation. Following the boiling experiment, the surface was greatly altered, from one having highly ordered nanostructure to 'blunt' structure. And, with increasing number of test runs, nanostructures became more densely populated, and pores among nanostructures more filled and smoothed.

Demir *et al.* [133] used metal-assisted chemical etching to fabricate nanorods of various lengths on silicon substrate. While Demir *et al.* did not encounter fracture or failure of individual nanorods, the tops of nanorods became rounded following the experiments.

Hunter *et al.* [134] used chemical deposition technique to form microscale and nanoscale copper structures on copper substrate, which proved effective at enhancing water pool boiling CHF appreciably. However, boiling performance was only repeatable at lower heat fluxes, as high fluxes caused partial or full removal of the micro/

Table 2
Literature related to time-dependent boiling performance by surface modification.

Authors	Technique or material	Heating cycles or boiling duration	Surface change after repeated tests or boiling experiment	Time dependency of boiling performance
Okawa <i>et al.</i> [126]	TiO_2 nanoparticle coating	60 min	Particles detach partially	CHF deteriorated
Morshed <i>et al.</i> [128]	Al_2O_3 nanoparticle coating	40 h	Particles detach partially	Single-phase heat transfer coefficient enhanced slightly
Lee <i>et al.</i> [129]	Nanoporous structure by anodization	900 h	Nanostructures wear off	Heat transfer coefficient unchanged following 500 hrs but deteriorated after 900 hrs
Lee <i>et al.</i> [130]	Nanoporous structure by oxidation	5 heating cycles	Aluminum hydroxide forms	Boiling heat transfer coefficient deteriorated
Lu <i>et al.</i> [105]	Nanoporous structure by galvanizing/ dealloying	100 h	Porosity becomes coarser.	—
Das <i>et al.</i> [132]	Nanostructures by electron beam evaporation	3 heating cycles	Nanostructures blunted	Heat transfer coefficient unchanged after three test runs
Hunter <i>et al.</i> [134]	Nanostructures by chemical deposition	6 heating cycles	Nanostructures delaminated	CHF and heat transfer coefficient increased before removal of nanostructures
Khamikar <i>et al.</i> [135]	GNTs	5 heating cycles	Initially near-vertical CNTs bent down	CHF degraded
Ho <i>et al.</i> [136]	GNTs	4 heating cycles	Appreciable aging	No variation in boiling performance
Rahman <i>et al.</i> [137]	Nanostructured coating by self-assembly of Tobacco mosaic virus technique	24 h	No observable physical degradation	—
Liu <i>et al.</i> [138]	Nano layer by liquid phase deposition	27 h	No fouling	No variation in heat transfer coefficient
Sahu <i>et al.</i> [139]	Nanofibers	5–6 h	No delamination	No appreciable degradation in heat transfer enhancement
Morshed <i>et al.</i> [141]	Nanocomposite coating by electroco-deposition	100 h	No noticeable morphological changes	—
Morshed <i>et al.</i> [142]	Copper nanowires	144 h	No loss of nanowires	No observable degradation

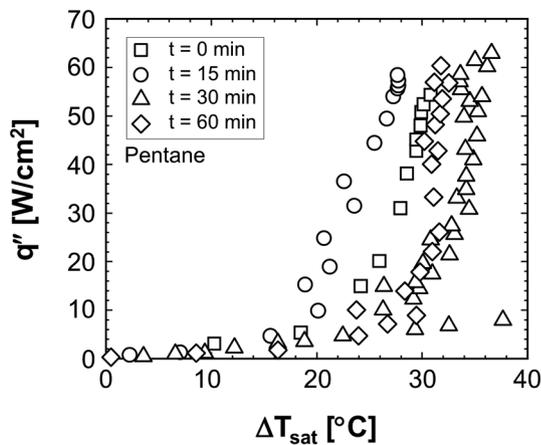


Fig. 14. Effect of boiling time on pentane pool boiling from Fe_2O_3 nanoparticle layer formed initially by vigorous nanofluid boiling, adapted from Stutz *et al.* [127].

nanostructures from the surface.

Khanikar *et al.* [135] explored potential heat transfer benefits of coating the bottom wall of a shallow $0.371 \times 10 \text{ mm}^2$ rectangular micro-channel with CNTs. Water flow boiling CHF data were repeatable only at low mass velocities but degraded following repeated tests at high mass velocities because of appreciable changes to morphology of the CNTs-coated surface. They observed that initially near-vertical CNTs were bent upon the heating surface at high mass fluxes to form a fish scale pattern, Fig. 15(a). Voids between the fish scales provided near-zero-angle cavities, which aided bubble nucleation in the nucleate boiling regime, while CHF was enhanced by increased heat transfer area associated with the CNT coating. However, the CHF enhancement degraded with repeated tests, Fig. 15(b), as CNT fin effect was compromised by the bending effect.

Ho *et al.* [136] fabricated a fully coated CNT surface and another interlace patterned CNT surface with 215- μm coating thickness using

CVD technique. While results from two consecutive tests with the CNT surface (initially immersed in FC-72 for more than 10 h) were consistent and within limits of experimental uncertainty, there were appreciable aging effects following the two tests, evidenced by noticeable changes in both wall temperature and CHF.

Rahman *et al.* [137] used self-assembly of Tobacco mosaic virus technique to fabricate nanostructured coatings on gold, copper, aluminum and stainless steel substrates. They showed water pool boiling heat transfer coefficient and CHF could be enhanced by up to 200%, with no observable physical degradation following 24 h of boiling. Liu *et al.* [138] employed a liquid phase deposition method to coat nanometer TiO_2 layer on the heating surface, and demonstrated enhancement in water flow boiling heat transfer coefficient along a 38-mm diameter tube, which they attributed to reduced wettability; they also reported their coating resisted fouling over 27 h of boiling. Sahu *et al.* [139] used electrically-assisted supersonic solution blowing technique to fabricate copper-plated polymer nanofibers on copper substrate. The nanofibers showed relatively strong adhesion to substrate, allowing them to withstand repeated 5–6 h pool boiling cycles in ethanol, water, and ethanol–water mixture without delamination or appreciable degradation in heat transfer enhancement, as shown in Fig. 16. Gao *et al.* [140] fabricated nanoporous dendritic structures on copper substrate using electrodeposition and annealing to enhance water nucleate pool boiling. They suggested binding force between nanostructures and substrate with this technique was strengthened by optimal control of both annealing temperature and time.

Using electroco-deposition, Morshed *et al.* [141] fabricated $\text{Cu-Al}_2\text{O}_3$ nanocomposite coating on the bottom wall of a rectangular 672- μm hydraulic diameter micro-channel. They reported no noticeable morphological changes to the surface after about 100 h of boiling. Morshed *et al.* [142] performed similar experiments with the channel's bottom wall coated with copper nanowires. No observable degradation was found in performance following 144 h of boiling.

Overall, despite the few studies reporting no appreciable changes to nanoscale surface morphology or boiling performance following relatively short-term boiling operation, most studies do point to numerous

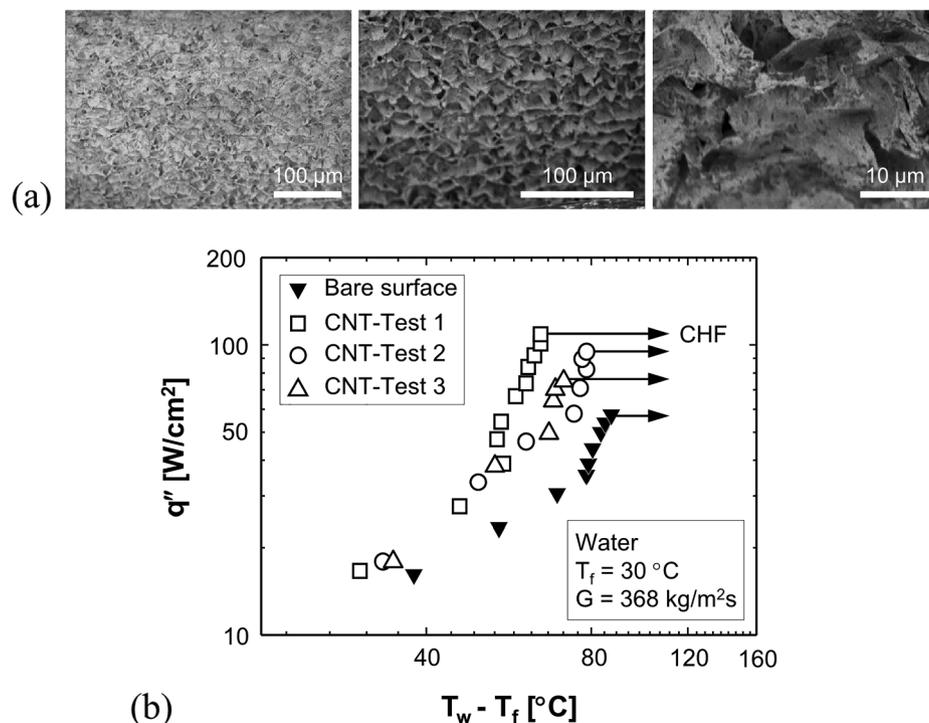


Fig. 15. (a) SEM images of dominant fish-scale pattern observed over CNT-coated surface after five boiling tests, and (b) subcooled water flow boiling curves measured at $G = 368 \text{ kg/m}^2\text{s}$ and $T_{in} = 30 \text{ }^\circ\text{C}$, adapted from Khanikar *et al.* [135].

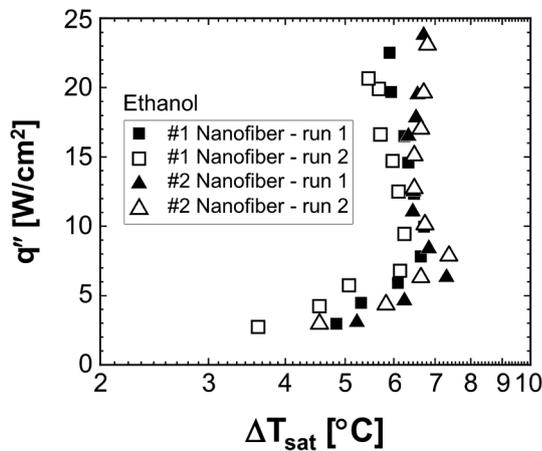


Fig. 16. Ethanol pool boiling curves from two different copper-plated nanofiber mats for repeated runs, adapted from Sahu *et al.* [139].

problems resulting from prolonged boiling. These problems include (a) partial or complete detachment of nanostructures, (b) appreciable changes to surface morphology, and (c) degradation of boiling performance, all of which may be exacerbated by fluid motion in flow boiling situations, especially in micro-channels. Aside from these problems, aluminum-based surfaces also suffer continuous oxidation in water boiling, resulting in weak reproducibility of boiling data. As mechanical failure of nano-engineered coatings often occurs at the coating/substrate interface, surface preparation and coating techniques need to be paid more attention. Also, one must make certain the repeated or long-term testing be performed at the same operating conditions, and follow a consistent protocol to allow fair comparison of results among different

groups [143].

These findings point to an urgent need for a systematic testing strategy (this topic will be detailed in the next section) for all nanoscale schemes intended to enhance pool or flow boiling heat transfer. Another noteworthy shortcoming of nanoscale research is absence of robust computational [144], theoretical [145,146] or universal empirical correlations [147,148] that are crucial for cooling device and system design. Notice that flow boiling enhancement differs significantly from that for pool boiling even when employing the same nanoscale surface modification technique. While, in both situations, surface topography will be altered because of vigorous bubble nucleation and detachment, surface nanostructures in flow boiling are susceptible to additional deformation caused by the fluid motion. This is expected to further reduce the durability of nanostructures and compromise the repeatability of boiling performance in flow boiling, especially at high flow velocities. This points to the need to balance the advantages of employing high mass velocities or high Reynolds numbers when aiming for superior performance against the higher likelihood of damaging the surface nanostructures.

4. Proposed systematic testing strategy

Ironically, time-dependent changes to cooling performance are prevalent even when using pure heat transfer liquids without surface enhancement. This is evident from common practices in the heat exchanger industry, where periodic maintenance is required to combat fouling problems and maintain desired heat transfer performance long term.

Clearly, the time-dependent performance issues associated with nanoscale enhancement at a certain heat flux under steady state are far more concerning than those for macro-scale heat exchangers and therefore demand adopting a systematic testing methodology to make

Proposed Systematic Testing Strategy for Nanoscale Boiling Enhancement

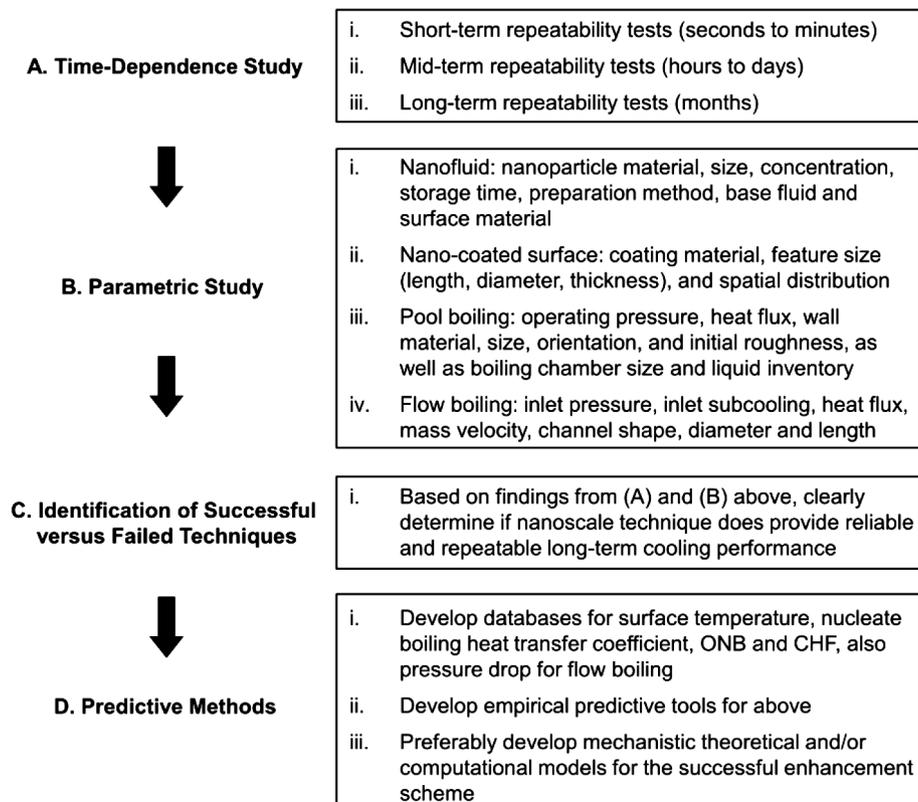


Fig. 17. Systematic testing strategy for nanoscale boiling enhancement.

certain any reported enhancement mechanisms remain prevalent and performance trends consistent long term.

To tackle this critical shortcoming, the authors of the present review recommend adopting a systematic testing strategy in future studies addressing nanoscale enhancement of pool boiling and flow boiling, as shown in Fig. 17. Following are key components of this strategy, which are based on observations drawn from the articles reviewed earlier:

A. Time-dependence study: to check both mechanical and chemical durability and repeatability of a certain technique at the same conditions in a consistent manner.

- i. Short-term repeatability tests: repeat tests over periods ranging from a few seconds to several minutes (to capture any 'rapid' changes to boiling behavior), starting each time with a fresh heating surface without contaminants (to decouple contamination and aging effects), including before and after SEM imaging of the surface, careful characterization of surface morphology and wettability, and, for nanofluids, characterization of nanoparticle suspension in base fluid.
- ii. Mid-term repeatability tests: repeat tests separated by several hours to several days (to assess ability of enhancement scheme to achieve mid-term 'steady state' operation), again starting each time with a fresh heating surface, and including before and after SEM imaging of the surface, careful characterization of surface morphology and wettability, and, for nanofluids, characterization of nanoparticle suspension in base fluid.
- iii. Long-term repeatability tests: repeat tests separated by several months to preclude any additional long-term changes to heat transfer performance. Like short-term and mid-term tests, long-term repeatability tests must include before and after SEM imaging of the surface, careful characterization of surface morphology and wettability, and, for nanofluids, characterization of nanoparticle suspension in base fluid.

B. Parametric study: perform steps in (A) above subject to the following parametric influences to determine major parameters affecting boiling performance.

- i. Nanofluid: vary nanoparticle material, size, concentration, storage time, and preparation method, as well as base fluid and surface material.
- ii. Nano-coated surface: vary nano-coating material, nano feature size (length, diameter, thickness), and spatial distribution of nanostructures on heating surface.
- iii. Pool boiling: vary operating pressure, heat flux, heating wall material, size, orientation, and initial roughness, as well as boiling chamber size and liquid inventory.
- iv. Flow boiling: vary inlet pressure, inlet subcooling, heat flux, mass velocity, and channel shape, hydraulic diameter and length. Special attention here should be paid to distinguishing between macro-, mini-, and micro-channels.

C. Identification of successful versus failed techniques: based on findings from (A) and (B) above, clearly determine if the nanoscale technique does provide reliable and repeatable long-term cooling performance to satisfy practical engineering requirements, by monitoring cumulative damages coming from delamination, detachment, wear-out and erosion, aside from repeatable heat transfer coefficient and CHF.

D. Predictive methods: for those techniques deemed successful in (C) above to provide useful predictive tools for engineering design.

- i. Develop a database for surface temperature, ONB, nucleate boiling heat transfer coefficient, and CHF, also pressure drop for flow boiling.
- ii. Develop empirical predictive tools for above.
- iii. Preferably develop mechanistic theoretical and/or computational

models for the successful enhancement scheme.

In fact, the engineering judgment depends on the working timescale of the surface in the applications where the enhancing technique is applied. However, despite differences in surface durability for different applications, investigators proposing a new enhancement technique must at a bare minimum clearly state duration of their boiling experiments to avoid misleading potential users.

Without this type of testing strategy, the authors believe two-phase nanoscale research will continue to entail simplistic curiosity experiments, lacking conformance to testing norms learned from decades of two-phase research or convincing evidence of viability of enhancement scheme to practical cooling applications.

5. Concluding remarks

This paper provided a comprehensive review of published articles addressing nanoscale techniques for pool boiling and flow boiling enhancement. Discussed were both heat transfer merits and drawbacks for each technique, with particular focus on reliability concerns stemming from time-dependent variations in boiling performance. Key observations from the review can be summarized as follows:

- (1) Significant inconsistencies surrounding influence of nanofluids on heat transfer coefficient in both pool boiling and flow boiling have been reported. These inconsistencies point to a need to perform nanofluid experiments in a far more comprehensive and systematic manner, and to cover broad ranges of operating parameters, in order to provide convincing assessment of dominant enhancement mechanisms. And, while most investigators point to nanofluids yielding measurable enhancement in CHF, resulting from improved surface wettability by the nanoparticle deposition layer, many questions remain concerning long-term sustainability of this enhancement. Absent systematic long-term boiling experiments and repeatability tests, it is expected phenomena such as nanoparticle clustering, sedimentation, erosion to heating surface, clogging of flow passages, high cost, and lack of quality assurance in nanofluid production and use, all cast serious doubts over viability of nanofluids for high-flux cooling applications.
- (2) As to nanotube/nanowire-coatings, enhancement or degradation of heat transfer coefficient for pool and flow boiling appear to be the outcome of competition between initial surface roughness and coating morphology. Studies point to decreased boiling incipience superheat and suppression of flow instabilities with these coating techniques, as well as enhanced CHF because of improved capillary wicking. But, here too, in the absence of systematic aging and repeatability tests, many important questions remain concerning long-term sustainability of the heat transfer enhancement, brought about mostly by partial or complete detachment of nanostructures and ensuing changes to surface morphology, which, like for nanofluids, bring into question viability of nanoscale coatings for high-flux cooling situations.
- (3) To address the above practical issues, a step-by-step strategy for future nanoscale enhancement work is proposed to ensure repeatability and reliability of cooling performance. Without this type of strategy, it is believed two-phase nanoscale research will continue to entail simplistic curiosity experiments, lacking conformance to testing norms learned from decades of two-phase research, or convincing evidence of viability of enhancement schemes to practical cooling applications.

CRedit authorship contribution statement

Gangtao Liang: Conceptualization, Writing - review & editing, Investigation, Funding acquisition. **Issam Mudawar:** Writing - review & editing, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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