



Experiments on Single-Phase Nanofluid Heat Transfer Mechanisms in Microchannel Heat Sinks: A Review

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Abstract: For more than 20 years, the use of nanofluids to enhance heat transfer in microchannel heat sinks (MCHSs) has been the subject of a large number of scientific articles. Despite the great potentialities reported in several works, the presence of controversial results and the lack of understanding of heat transfer enhancement mechanisms prevent further advancement in the use of nanofluids as coolants. This article reviews the scientific literature focused on several aspects of nanofluids that have a role in the heat transfer enhancement within the MCHSs: nanofluid stability, thermal conductivity, and particle clustering, as well as the particle–surface interactions, i.e., abrasion, erosion, and corrosion. We also include the most relevant works on the convective heat transfer and MCHSs operated with nanofluids in our review.

Keywords: nanofluids; nanofluid stability; convective heat transfer; microchannel heat sink; particle clustering; thermal conductivity; abrasion; erosion; corrosion



Citation: Eneren, P.; Aksoy, Y.T.; Vetrano, M.R. Experiments on Single-Phase Nanofluid Heat Transfer Mechanisms in Microchannel Heat Sinks: A Review. *Energies* 2022, *15*, 2525. https:// doi.org/10.3390/en15072525

Academic Editor: Patrick Phelan

Received: 13 February 2022 Accepted: 27 March 2022 Published: 30 March 2022

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1. Introduction

The design of compact heat exchangers is a crucial task in major industrial environments, such as microelectronics, nuclear energy, biotechnology, automotive, and aerospace engineering [1]. For example, electronic devices comprise tiny components, such as transistors, microcontrollers, and microprocessor chips, which are heat-sensitive and can easily be damaged by overheating. The growing demand in increasing their potentiality, together with the rapid decrease in their component size, has led to an essential need for thermal management and packaging [2].

Both active (jet and spray cooling, mechanical mixing, vibration) and passive methods (surface coating, turbulence and mixing intensification, engineered fluid, etc.) are employed to remove the excess heat. However, with the continuous increase in power density, current heat dissipation methods will lack the required efficiency, leading to a technological bottleneck. Hence, understanding the transport phenomena involved in the micro-heat transfer will allow further miniaturization and optimization in cooling systems.

Single-phase cooling is one of the most promising and practical mechanisms for highpower electronics when assessing the combined effect of heat removal, operating pressure losses, operational stability, leakage, and device fabrication efforts [3]. As a drawback, the fully developed thermal boundary layer limits the single-phase cooling in terms of high surface temperature gradients. For instance, the highest Nusselt number is obtained at the inlet, and it monotonically reduces in the flow direction due to thermal boundary layer development, which eventually plateaus to a constant in the fully developed region [4]. Although chip cooling via two-phase flow exploits thermo-dynamic advantages, such as a minimal presence of hot-spots and surface temperature gradients [5], single-phase water cooling is considered as the opted near-term solution. This is mainly due to the overall high hydro-thermal performance and reduced operating cost and system complexity.

Different strategies can be employed to enhance the heat transfer in single-phase cooling. One of them is to optimize the microchannel geometry, but the increase in the

pressure drop and manufacturing constraints may limit this solution. Another possibility is the use of fluids with enhanced thermal conductivity (*nanofluids*), and there has been a popular trend to apply nanofluids in miscellaneous applications [6,7] (see Figure 1).



Figure 1. Scopus search: *nanofluid* as the keyword and the narrowed search with *nanofluid* + *mi*-*crochannel heat sink*. The search illustrates an exponential growth in the number of published works.

Although the research about nanofluids has attracted an enormous attention worldwide and a great number of published works exist in the literature [8], nanofluids have not been applied yet in practice for heat transfer applications. This gap between the research community and the industry is partially due to the reported controversial and inconsistent experimental results [9], which can even strongly scatter under the similar experimental conditions.

2. Nanofluids and Heat Transfer Mechanisms

There are critical length scales for the physical mechanisms below whose materials' physical properties significantly alter. For example, nanoparticles with diameters of <100 nm possess features unlike those of conventional bulk solids [10]. With the advent of technology in material science, nanometer-sized solid substances are engineered on the atomic or molecular scale. This helps to tune novel or enhanced physical properties, which the conventional bulk solids do not exhibit. These noble functionalities of nanometer-sized materials result from the relatively high surface-area-to-volume ratio, due to the high proportion of constituent atoms residing at the grain boundaries. Consequently, the thermal, electrical, mechanical, optical, and magnetic properties of nano-phase materials become superior compared to those of conventional types with coarse grain structures.

For over a century, many researchers have endeavored to improve the inherently poor thermal performance of traditional heat transfer fluids, such as water, oil, or ethylene glycol (thermal conductivity; 0.6, 0.15, and 0.25 W/(m·K) respectively). Since the theoretical study of Maxwell, based on thermal diffusion [11], plenty of theoretical, numerical, and experimental works have been carried out, investigating the thermal conductivity of suspensions containing solid particles. However, those studies were limited to millimeter-or micrometer-sized particles. Their use has brought numerous problems, such as rapid particle deposition, high pressure drop, abrasion, and clogging, even though some heat transfer enhancement is observed [12]. Later on, the term *nanofluid* is coined by Choi and Eastman. In 1995, at the Argonne National Laboratory, they introduced a new class of engineered fluid [10]: a colloidal mixture of base fluid with nanometer-sized solid particles, with at least one dimension being less than 100 nm [13].

The nanoparticle materials are chosen regarding their chemical inertness with the base fluid. Hence, commonly used materials include oxides (alumina Al_2O_3 , silica SiO_2 , titania TiO_2 , copper oxide CuO), metals (Cu, Au, Ag), graphene flakes, and carbon nanotubes

(CNTs). Nanofluids are prepared via the following methods, the one-step and the twostep method.

Among the different ways to enhance heat transfer, the use of nanofluids seems to be very promising [14]. As heat transfer is a surface phenomenon and takes place at the surface of nanoparticles, an enhancement in heat transfer is expected for decreased particle size, i.e., increased surface-area-to-volume ratio [15,16]. Indeed, due to the addition of nanoparticles, the thermophysical properties of some conventional coolants are improved [17]. In particular, some nanofluids posses a higher thermal conductivity compared to that of the base fluid. The presence of nanoparticles not only modifies the thermo-physical properties of the base fluid, but also affects the heat transfer mechanism [18]. Figure 2 shows the postulated heat transfer mechanisms for nanofluids [19,20]. Four heat transfer modes can be identified as follows:

- Mode 1: the collision of base fluid molecules with each other;
- Mode 2: the thermal diffusion occurring within the nanoparticles;
- Mode 3: the nanoparticles' collisions with each other (Brownian motion);
- Mode 4: the thermal interactions of dynamic or dancing nanoparticles with the base fluid molecules.



Figure 2. Energy transport modes in nanofluids [19,20]. Adapted with permission from Ref. [19], Copyright 2004 AIP Publishing LLC.

Especially, the Brownian motion (mode 3) is influenced by the temperature, viscosity, and size of the nanoparticles [21]. Increased temperature in the system and smaller size of the nanoparticles augment the Brownian motion, leading to higher thermal conductivity, whereas the increase in the fluid viscosity suppresses the Brownian motion. The particle-particle interactions and micromovements of particles enhance the thermal conductivity of nanofluids [22]. In addition, the microconvection streams due to the microscopic movement of particles contribute to the convective heat transfer enhancement. Keblinski et al. [18] try to explain the enhancement in the thermal conductivity due to smaller grain size with the following further possible causes:

- Brownian motion enabling direct heat transfer from one solid to another;
- Liquid layering between the liquid-particle interface;
- Nature of heat transport in nanoparticles;
- Clusters of nanoparticles by creating lower thermal resistance paths.

When the nanofluid flows in a microchannel, the interactions between the nanoparticles and the walls also play an essential role in the convective heat transfer performance of nanofluids. The thermal boundary layer distortion by the nanoparticles is considered one of the reasons for the improved heat transfer mechanism [23]. Since the nanoparticles frequently hit the channel walls, they distort the boundary layer and move across the bulk fluid. That is, the presence of nanoparticles promotes mixing in the thermal boundary layer due to the strong cross-flow velocities, which further enhance the heat exchange within the boundary layer [24]. Moreover, these interactions intensify with the increased nanoparticle concentration, yielding a larger heat transfer coefficient and Nusselt number [25].

Though CNTs and graphene flakes are exceptionally good thermal conductors, their orientation in the flow field can remarkably change the thermal performance of the nanofluids [26]. The ballistic conduction of the heat-carrying phonons along the tubes axis and the flake faces facilitates the heat transfer, yet in the perpendicular direction, the thermal performance can drastically drop [14].

Nevertheless, some studies also report the deterioration of thermal performance of nanofluids [27]. The contradictions demonstrate that there might be an optimal nanoparticle concentration, beyond which particles start to cluster and deposit, worsening the heat transfer [21,28]. In addition, Hung et al. conclude that the thermal resistance of a system containing nanofluids first drops and then increases for the increased particle concentration [29]. Bergman [30] theoretically expresses that nanofluids' thermal performance may not be the same when used in another application, in which the geometry and the mass flow rate are different because of the competition between the increased thermal conductivity and the reduced specific heat of nanofluids. As an optimal particle concentration, <2 vol.% is recommended so that the the overall thermal performance of the nanofluid does not decrease due to the increased pressure drop [31].

3. Stability of Nanofluids

The larger relative surface area of nanoparticles not only improves the heat transfer capabilities but also enhances the suspension stability [32]. Both uniform dispersion and stable suspension of nanoparticles in the base fluid can have a substantial influence on the final thermo-physical properties of nanofluids [33]. They act as key factors in most nanofluidic applications since nanofluids may not be stable in time, i.e., nanoparticles may aggregate and sediment in the form of clusters.

The stability of nanofluids and colloids can be explained with the DLVO theory (Deryaguin–Landau–Verwey–Overbeek). It combines the van der Waals attraction and electrostatic repulsion to describe the tendency of the colloids to either cluster or separate [34,35]. That is, the competition between these forces determines the stability of the nanofluids (see Figure 3).



Figure 3. A schematic describing the DLVO theory.

As schematized in Figure 4, the nanoparticles in the base fluid are exposed to several intermolecular forces. Some of these forces can be neglected due to the minute size of the nanoparticles. In contrast, the dominant ones result from the interactions between the nanoparticles and the liquid molecules and the inertia of the adsorbed liquid layer [36]. All in all, the molecular momenta of liquid–nanoparticle collisions impact the energy transport and thermal diffusion at the nanoscale and on the thermal conductivity at the microscale.



Figure 4. Applied forces on a nanoparticle in the base fluid. Adapted from Ref. [36].

In a stable nanofluid, the particles should be small enough to be suspended by the Brownian motion and be preserved against clustering. This can be chemically or mechanically achieved by modifying the bulk medium or by using additives (see Figure 5).





Steric and electrostatic stabilization, as visualized in Figure 6, are the two traditional chemical mechanisms that help to improve the colloid stability by increasing the repulsion between the particles [37]. For instance, if the charging of the particles is near their isoelectric point (IEP), it is very likely to observe particle clustering. Even for the stable Al_2O_3 nanoparticles suspended in water operating close to IEP (pH \approx 9), the colloid can become unstable due to the low energy barrier, and the doublets are created after a while, forming very large clusters in the end [38]. The ultra-sonication, magnetic stirring, bead milling, and high-shear mixing are the main mechanical methods for the proper dispersion of nanoparticles in the base fluid [9].



Figure 6. A representation of the steric (**left**) and the electrostatic (**right**) stabilization on the particles [39]. Adapted with permission from Ref. [39], Copyright 2002 American Chemical Society.

The level of the stability can be evaluated by the zeta potential [2], the turbidity scan index (TSI) [40], and the dispersion quality index (DQI) [41].

Figure 7 presents different stability scenarios, for which the measurements (thermal conductivity, viscosity, etc.) would differ from each other.



Figure 7. Various dispersity levels of suspensions, i.e., stable and unstable nanofluids [42].

4. Thermal Conductivity Measurements

The effective viscosity and thermal conductivity of nanofluids are essential transport properties for applications of nanofluids as a new class of heat transfer fluids in thermal devices or systems, such as heat exchangers or cooling systems [33]. The formulations of the effective thermal conductivity work well for micrometer- or larger-sized particles, yet fail to explain the unusual characteristics of the nanoparticles [32]. For instance, theoretical models such as Maxwell [11] and Hamilton–Crosser [43] underestimate the thermal conductivity of nanofluids since those models do not take into account the Brownian motion of the particles, particle size, nanolayering, and nanoparticle clustering [44]. Various hypotheses are speculated to explain these unexpected and intriguing findings [45]:

- Particle Brownian motion agitating the fluid, and thus creating a micro-convection effect that increases the energy transport;
- Clusters of particles being formed within the nanofluid and local percolation of heat preferentially along such clusters;
- Base fluid molecules forming highly ordered high thermal-conductivity layer around the particles, which augments the effective volumetric fraction of the particles.

However, the confirmations of these speculations are weak. For instance, the microconvection hypothesis yields predictions conflicting with some experimental data. Moreover, not only do the theoretical studies not cohere with each other, but the nanofluid thermal conductivity data are also sparse and inconsistent [46]. All these inconsistencies are possibly due to:

- The broad range of experimental approaches;
- Incomplete characterization of the nanofluid samples;
- The differences in nanofluid synthesis, even for nominally similar nanofluids.

The enhancement in heat transfer by nanofluids depends on various parameters: particle material, size, shape, volume concentration, dispersity, as well as the base fluid material, temperature, additives, and acidity level [47]. Moreover, the geometry, clustering state, and surface resistance of nanoparticles are the main variables controlling the thermal conductivity enhancement in nanofluids [48]. As the additive amount is generally minimal, it is expected to not affect the nanofluid's thermal conductivity [49].

The inconsistencies between the nanofluids' thermal conductivity coming from measurements, theoretical and semi-empirical models are also observed over an extensive range of particle size, concentration, nanoparticle material, base fluid type, and surfactant amount. Experiments on CuO–water nanofluids (8 nm, 0.3 and 0.8 vol.%) reveal that the particle size, concentration, poly-dispersity, and cluster size play a crucial role in their thermal conductivity [16].

According to Jang and Choi, the key mechanism governing the effective thermal conductivity of nanofluids is not the type of the particle material but the Brownian motion of the nanoparticle [19]. Their model predicts the nanofluid's effective thermal conductivity and is validated with experimental results of both CuO–water (20 nm) and Al_2O_3 –water (30 nm) nanofluids. It demonstrates that the size of the nanoparticles affects the heat transfer the most [50]. On the contrary, Peyghambarzadeh et al. report that nanoparticle thermal conductivity is more critical than the particle size for the heat transfer improvement [51].

Buongiorno [52] theoretically explains that nanoparticle dispersion does not contribute to the energy transfer. Instead, the Brownian diffusion and thermophoresis influence the substantial variations in nanofluid thermal conductivity and viscosity within the laminar sub-layer of the turbulent boundary layer. Additionally, the nanofluid properties can noticeably vary within the thermal boundary layer due to the temperature gradient and thermophoresis. These effects may significantly decrease the viscosity within the laminar sub-layer, which eventually leads to the heat transfer enhancement.

The most common techniques implemented to measure the effective thermal conductivity of nanofluids are: the transient hot wire method [53], the steady-state method [54], temperature oscillation [55], and the hot strip method [56]. As reported earlier, the results strongly depend on the used measurement techniques, which impedes a clear understanding of the physical mechanisms taking place during nanofluid heat transfer processes. In all these techniques, the onset of natural convection of the base fluid is considered as one of the major causes of uncertainties and must be suppressed to obtain a higher accuracy of the results [57]. A single-sided horizontal method is proposed to measure the thermal conductivity of liquids, especially for nanofluids. In this method, the natural convection effects are suppressed by constraining the upper part of the system with a solid material, whose thermal properties are well known [58]. Das et al. measured the thermal diffusivity and thermal conductivity via the temperature oscillation method. They investigate the heat transfer enhancement by evaluating the temperature effect on thermal conductivity [55]. They note that there could be a threshold temperature for each particle size at which the nanofluid's effective thermal conductivity begins to deviate from that of typical slurries. Furthermore, the enhancement through the stochastic motion of the particles starts to dominate.

Hybrid nanofluids, consisting of two or more distinct materials of nanoparticles with higher thermal conductivity, can be synthesized to further enhance the heat transfer [59]. Mostly, three types of hybrid (composite) materials are prepared [60]: (i) ceramic ma-

trix nanocomposites $(Al_2O_3/TiO_2 \text{ and } CNT/Fe_3O_4)$, (ii) metal matrix nanocomposites $(Al_2O_3/Ni \text{ and } Al_2O_3/Cu)$, and (iii) polymer matrix nanocomposites (polymer/CNT and polyester/TiO₂). Because of its complexity, a complete understanding of the heat transfer mechanism of the hybrid nanofluids is still lacking and requires more research.

Due to its uncovered complexity and controversial results reported in the literature, no concrete conclusions have been drawn yet about the amount of enhancement and its occurring conditions [61,62]. All in all, the thermal conductivity enhancement of nanofluids is still a hotly debated topic.

5. Convective Heat Transfer Measurements

Similar to the ongoing debate on the nanofluid thermal conductivity, there has also been no consensus on the convective heat transfer coefficient of nanofluids. A part of the research community claims considerable potential for the heat transfer enhancement, while some researchers observe deterioration.

Many researchers have investigated nanofluids' convective heat transfer within the pipe flow. Selvakumar and Suresh experimentally show that the improvement in the convective heat transfer coefficient (\approx 29%) is possible for low particle concentrations (0.1 and 0.2 vol.%) [63]. Furthermore, the average increase in pumping power is only 15%, thus promising practical cooling applications of nanofluids, similar to the findings of Xuan and Li [64].

Hwang et al. illustrate that the flattening of the velocity profile could be the reason for the convective heat transfer enhancement, which cannot solely be attributed to an increase in the nanofluid thermal conductivity. The flattening of the velocity profile occurs owing to nanoparticles drifting towards the tube center-line due to Brownian diffusion and thermophoresis [65].

The simulations also show that the nanoparticles tend to migrate from the tube walls towards the tube center, causing a non-uniform particle concentration in the transverse direction. This incident can alter the spatial effective viscosity and the thermal conductivity due to the thinning of the boundary layer, which induces the change in the fluid flow and the heat transfer [66].

It is also experimentally observed that nanofluids promote heat transfer mostly in the developing region compared to the fully developed region [67]. The reason for this may be attributed to particle migration and thermal dispersion effects [68]. Likewise, better convective heat transfer coefficients are obtained in laminar flow for Al₂O₃-water nanofluids compared to CuO-water nanofluids, despite the higher thermal conductivity of CuO nanoparticles [12]. More specifically, larger thermal conductivity is not the only factor responsible for the heat transfer enhancement. Yet, other mechanisms, such as the dispersion and chaotic movement of the nanoparticles, Brownian motion, nanoparticle migration, and nanofluid viscosity, are proposed to interpret the experimental results. With the changes in the flow structure due to the presence of nanoparticles, as well as the increased thermal conductivity, Brownian motion, dispersions, and fluctuations of nanoparticles, especially in the vicinity of the pipe wall, are foreseen to promote the heat transfer rate.

On the other hand, Sommers and Yerkes [27] employ Al_2O_3 -propanol nanofluids (10 nm, 0.5 wt.%, 1 wt.%, and 3 wt.%) in an 18-inch-long copper pipe with ID = 3/4". Only with nanofluid at 1 wt.% did they observe a 15–20% enhancement in the heat transfer coefficient only between 2000 < Re < 3000. Since the physical mechanism behind this enhancement is believed to be an early transition from laminar to turbulent flow, the heat transfer enhancement occurs within a very narrow Re number range. In addition, the increase in the pressure drop and discoloration of the nanofluid, possibly due to abrasion, prevent them from recommending nanofluid usage.

Wong et al. study buoyancy induced flow in rectangular enclosures with Al_2O_3 -water nanofluids at 10 and 25 wt.% [69]. They observe a drastic change in the natural convective

behavior of the fluid due to the addition of nanoparticles. The use of nanofluids extends the multi-cellular flow to larger temperature differences with respect to the base fluid.

A reduction in the average Nusselt number in natural convection is also reported when the nanoparticle volume fraction is more than 5% [70]. However, such a decrease is not observed for lower particle concentrations. The results in the literature support that there is an optimal nanoparticle concentration, which enhances the heat transfer performance. Above this value, particle deposition may deteriorate the cooling mechanism [8].

6. Coupling of Microchannel Heat Sinks with Nanofluids

With the demand for removing excess heat reaching 500 W/cm², conventional forcedair heat exchangers have become insufficient. Hence, the control of overheating in processing units and the extension of their lifetime needs the development of specific cooling systems. Microchannel Heat Sinks (MCHSs) are one of the heat transfer augmenting tools to enhance microchip cooling, owing to the large surface-area-to-volume ratio [71]. In addition, the microscopic size of the channels lowers the thickness of the thermal boundary layer, i.e., reduced convective resistance to heat transfer yields higher cooling rates [72].

Tuckerman and Pease are the pioneers who introduced the concept of MCHS in 1981 [73]. They design and test a compact water-cooled integral heat sink by etching microscopic channels with 50-µm width and 300-µm depth on the silicon substrate. They demonstrate that the heat transfer coefficient is inversely related to the channel width in laminar and fully developed flows. In other words, the narrower the channels in the heat sink, the higher the heat transfer coefficient. However, the possible disadvantages are the increased pressure drop, manufacturing costs, and filtering requirements to avoid channel clogging.

A typical MCHS consists of numerous microchannels with the hydraulic diameter ranging between 10 and 1000 μ m. Although the rectangular geometry is mostly studied, various types of geometries are experimentally and numerically tested in the literature [74]. Some of them are illustrated in Figure 8 below.



Figure 8. Microchannel geometries reported in the literature. Adapted with permission from Ref. [75]: (a) parallel plate, (b) rectangular or square, (c) circular, (d) trapezoidal, (e) triangular, (f) straight, (g–j) converging and/or diverging, (k) sudden expansion, (l) zigzag, and (m) wavy.

According to Qu and Mudawar, the Navier–Stokes and energy equations apply to the predictions of the fluid flow and heat transfer within the MCHS [76]. Both the measured temperature distributions and the pressure drop agree well with the numerical predictions for Reynolds number Re < 1700 and characteristic length > 100 μ m. In addition to those, the experiments indicate that the flow and heat transfer characteristics are strongly linked to the hydraulic diameter and the channel aspect ratio [77]: The critical Re number diminishes with the reductions in the channel size.

Generally, air, water, fluorochemicals, and ethylene glycol are considered coolants suitable for MCHSs. However, these coolants are limited in terms of their thermal conductivity.

Jang and Choi [50] numerically investigate the potential of using MCHSs (small characteristic length) in combination with nanofluids (enhanced thermal conductivity) as the next generation of cooling devices for removing ultra-high heat fluxes. They show that reduced temperature and thermal resistance of the MCHS are obtained for diamond–water nanofluids compared to pure water. Nevertheless, they did not take into account the nanoparticles' stability and deposition inside the MCHS, which is expected to modify the heat transfer rate.

According to the theoretical calculations, the heat transfer enhancement by nanofluids appears to be more effective in laminar flows [67]. The laminar flow is typically anticipated thanks to the smaller hydraulic diameters and slower flow rates within the microchannels. Therefore, the use of nanofluids within the MCHSs seems a promising strategy to improve the cooling performance.

The combined benefits of microchannel flow and nanofluids are investigated for singlephase and two-phase systems. Among published experimental studies (<30% of the whole studies, including the numerical and the analytical ones as well) on the use of nanofluids as coolants within the MCHS, the results indicate the general trend of opted thermal behavior of nanofluids compared to the base fluids. Especially, nanofluids possessing a low fraction of nanoparticles improve thermal and hydrodynamic performance in microchannels [75].

The experiments of Ho et al. [78] show that the Al_2O_3 -water nanofluid-cooled MCHS outperforms the water-cooled one, thanks to the significantly higher average heat transfer coefficient (up to 70%) and substantially lower thermal resistance and wall temperatures. Despite the apparent rise in the dynamic viscosity values due to the dispersed nanoparticles, the friction factor only slightly increases. Similar results are also reported in [23]. Likewise, the improvement in the heat transfer coefficient of Al_2O_3 -water nanofluid becomes more distinguishable at a higher Re number in the laminar flow regime [79], particularly at the channel entrance [80].

Byrne et al. focus on the effect of surfactant use on the thermal and hydraulic performance of CuO–water nanofluids [81]. They conclude that proper suspension of nanoparticles cannot be maintained without surfactants, even at 0.1 vol.%. Furthermore, nanofluids containing surfactants outperform the pure water and surfactant-free nanofluids in terms of heat transfer. No significant penalty on pumping power is observed due to the presence of nanoparticles and the surfactant.

Chein and Chuang carry out both theoretical and experimental work to evaluate the MCHS performance using nanofluids [82]. They observe that the nanofluid-cooled MCHS could dissipate more heat compared to the water-cooled MCHS for low flow rates. On the other hand, if the flow rate becomes higher, it dominates the heat transfer process, and the contribution of the nanoparticles becomes less important. The measured MCHS wall temperature variations agree well with the theoretical predictions at low flow rates, whereas they do not entirely match the theoretical predictions at high flow rates. This is related to the triggering of particle clustering and deposition at high flow rates. Only a slight increase in the pressure drop within the MCHS is monitored during the experiments due to the presence of nanoparticles.

Peyghambarzadeh et al. also observe the deterioration of the heat transfer performance at high Re numbers due to the clustering and sedimentation of the nanoparticles within the channels [51]. They state that increased flow velocity for the nanofluids with low nanoparticle concentration helps to better disperse the nanoparticles. In contrast, it may trigger the instability of the nanofluids at high nanoparticle concentrations. Therefore, spotting the optimized concentration of nanoparticles is crucial, for which the heat transfer coefficient enhances with increased Re number.

7. Particle Clustering and Its Controversial Consequences

According to the hypothesis of heat percolation along the particle clusters [83,84], there might be an optimum level of clustering to attain thermal properties much greater than what is predicted in homogeneous suspensions [85], as schematized in Figure 9. This could be one of the paramount reasons for the large data scattering in the literature and the controversy regarding the nanofluids' potential as a coolant.





However, time-dependent measurements in water-based CuO nanofluid show that their thermal conductivity decreases with elapsed time due to the clustering of nanoparticles, as confirmed microscopically [16]. Indeed, these clusters are likely to settle within the base fluid due to the larger mass, thus causing particle concentration gradients in the fluid. As expected, nanofluids with higher particle concentrations tend to cluster due to the reduced mean interparticle distance, which leads to stronger van der Waals attraction forces. Sonication can break down the clusters into primary nanoparticles. For example, by extending the probe sonication time, the thermal conductivity of CuO–water nanofluid is increased up to 18% via the de-agglomeration process [87].

The experiments of Prasher et al. [88] are one of the first works taking into account the effect of particle clusters on the effective thermal conductivity of nanofluids. In other words, they combine the kinetics of nanoparticle clustering with the physics of thermal transport. They claim that the measured anomalies in nanofluid thermal conductivity reported in the literature can be described by considering the clustering kinetics.

Wu et al. experimentally and theoretically analyze the effects of altered cluster structure and particle size distribution on the effective thermal conductivity of SiO₂–water nanofluids (15 nm, 23 vol.%) [89]. They control the clustering kinetics by adjusting the nanofluid pH, starting from a well-dispersed system. No noticeable enhancement in the thermal conductivity is observed for clustered systems despite the high concentration. According to the generalized fractal model predictions, the conductive contribution due to the clustering may be canceled completely by the decreased convective contribution due to the particle growth (thermal conductivity of silica particles is very low, 1.4 W/(m·K)). Moreover, they recommend the use of nanoparticles with higher thermal conductivity and the optimized fractal dimension to maximize the enhancement of the thermal conductivity via particle clustering based on their predictions.

Although there is extensive research in the literature on the thermal performance of MCHSs with nanofluids, a few experiments are reported on the particulate fouling of nanofluids within the MCHSs [21,90]. For instance, Sarafraz et al. observe the rise in thermal resistance parameter with increased operating time for all nanofluids (Silverwater based, 0.01, 0.05, and 0.1 wt.%, for Re = 200). In addition, they distinguish three different stages over time: a linear increase in thermal resistance parameter in stage 1; a constant thermal resistance parameter in stage 2; and a sudden rise in the thermal resistance parameter in the last stage. These variations are then attributed to the change in the surface roughness and pressure drop, i.e., change in the fluid flow inside the MCHS, resulting from

the deposition of the nanoparticles. Other interesting results out of these studies are the following. With increased nanoparticle concentration, the thermal resistance is intensified. Moreover, the required time for the fouling thermal resistance parameter to reach a constant value is inversely proportional to the particle concentration. This is also attributed to the increase in the nanoparticle concentration and consequent augmented deposit on the MCHS walls. The larger pressure drop is due to the presence of nanoparticles in the bulk fluid, triggering the friction between the layers of the nanofluid (higher viscosity).

Particle deposition is a complex process since it involves particle–particle and particle– wall interactions. These interactions depend on the bulk fluid temperature, particle size and shape, electrochemical, and thermophysical properties of the bulk fluid, the particles, and the wall [91]. In particular, the particle size influences not only the strength of the physicochemical interactions between the particles and the channel wall but also the hydrodynamic forces, which reduce the tendency of the particles fouling on the surface [38]. The forces acting on a particle also affect particle deposition. More specifically, the temperature gradient present in the flow field causes thermophoretic forces exerted on the particle, pointing them to lower temperature regions, in addition to the lift and drag forces [92].

It is also speculated that the clustering of the particles cannot be avoided even if the surfactants are added, especially when the nanofluid is heated [82]. In fact, the irreversible degradation of the surfactants, generally exceeding 60 °C, is a well-known issue [84]. According to Arshad and Ali [93], both the hydrodynamic and thermal performances of graphene nanoplatelet–water nanofluid deteriorate with increased supplied heat flux due to the destabilization of the nanofluid at high temperatures. To sum up, the stability of nanofluids at high temperatures still remains a challenge.

8. Abrasion, Erosion, and Corrosion Characteristics of Nanofluids

Despite their attraction for more efficient heat transfer systems, nanofluids' encountered potential side effects are rarely assessed in the literature [94]. As an example, their tribological behavior is largely unknown [95]. It is vaguely mentioned that there could be erosion due to the clustered nanoparticles and that the removed material could impact the heat transfer performance of the nanofluids, as well as ending up with clogging, perforation, leakage, or sudden failure. Moreover, the study of long-term exposure effects of nanofluids, e.g., corrosion, on the metallic surfaces and components of the industrial systems is essential, but poorly reported [96].

The terms erosion and abrasion are often mixed. During the wear by erosion, the solid surface is damaged by the particles in liquids/gases impacting against a solid surface. In contrast, wear by abrasion causes material loss due to harder particles passing over the solid surface [97]. During corrosion, on the other hand, the surface of the material is chemically damaged so that it forms a chemically more stable state. The Mohs' scale of some materials are listed in Table 1 to give an idea about their hardness levels since the resistance to erosion is proportional to their bulk hardness for annealed pure metals [98].

Material	Mohs' Scale	Material	Mohs' Scale
Al	2.75	Silicon (Si)	6.5
Cu	2.5-3	Quartz (SiO ₂)	7
Plastic media	3–4	Borosilicate glass	7.5
CuO	3.5-4	ZrO ₂	8
Stainless steel (SS)	5-6.5	Al_2O_3	9
TiO ₂ -Anatase	5.5-6	SiC	9.5
TiO ₂ -Rutile	6-6.5	Diamond	10

 Table 1. Hardness levels of some common bulk materials [99–101].

It is interesting to note that these phenomena are interrelated [102,103]. Erosion can trigger corrosion by removing the passive protective layer on a material's surface, especially on metals. Likewise, corrosion can induce erosion via a preferential dissolution

of the material, or it can impede erosion by creating a passive protective layer on the surface. The test conditions also have a huge impact. For instance, under static conditions, the corrosion rate diminishes in time until the equilibrium is met, while a larger corrosion rate is expected under dynamic conditions with a continuous flow, especially under turbulent conditions. Additionally, erosion differently propagates on the surfaces of brittle and ductile materials.

As a theoretical a priori assumption, the mechanical (abrasion and erosion) and chemical (corrosion) secondary effects can be assumed negligible considering the nanoscale of the particles and their small weight percent in the nanofluid. That is, a negligible amount of momentum is expected to be transferred from the particles to the channel walls and the surfaces of circuit components in the fluidic loop. Nonetheless, the abrasive characteristics of the clustered particles can provoke erosion on the surfaces, and there has been no rigorous research investigating these phenomena in parallel. Similarly, it can be suggested that nanofluids have more corrosive characteristics to metals compared to their base liquids, while the corrosion inhibition characteristics of the nanoparticles to metals might be the case. Therefore, the center of the attention has been on the thermal behavior of the nanofluids rather than its adaption to the operating conditions similar to the industrial heat transfer applications.

Abrasion, erosion, and corrosion characterization is possible by several techniques [104,105]: Scanning electron microscopy (SEM), Transmission electron microscopy (TEM), Electron back-scatter diffraction (EBSD), X-ray diffraction (XRD), Energy-dispersive X-ray spectroscopy (EDS), Atomic force microscopy (AFM), Optical microscopy (OM), Electrochemical impedance spectroscopy, polarization resistance tests, surface roughness tests, weight loss tests, and visual inspection. By these methods, direct observation on the morphology of the sample's surface and the characterization of the modification (e.g., particle accumulation) can be carried out.

Sommers and Yerkes [27] are the only ones reporting the discoloration of nanofluids (see Figure 10), which becomes more severe at high flow rates and elevated temperatures. They obtain no heat transfer enhancement during those tests by using nanofluids. Furthermore, they observe a deterioration in the thermal performance of the nanofluid with increasing discoloration. Using X-ray powder diffraction (XRD) on the centrifuged samples before and after the heat transfer experiments, they try to understand the reason for the discoloration. They speculate that nano-abrasion occurs on the softer materials in the setup since the hardness level of Al_2O_3 is higher than the rest of the materials. Their speculation is indeed motivated by the fact that suspensions of submicron Al_2O_3 particles are commonly used in mechanical abrasive polishing [106]. We can also suppose that the severity level of the discoloration would be enhanced in microchannels due to the larger wetted area and the spatial constraints promoting the particle clustering.



After





In the past few years, the tribological characteristics of nanofluids on metals have gained some attention. An erosion rate of 0.1 vol.% TiO₂ nanofluid impingement is measured on ductile (aluminum) and brittle (cast iron) substrates [107]. The abrasive nature of the particles is the prominent mode of material loss in aluminum, and nanofluid impingement smoothens the aluminum surface according to AFM imaging. However, corrosion-assisted erosion is the responsible mechanism in cast iron due to the removal of the protective oxide layer by the nanofluid flow. The jet angle and impingement speed influence the amount of material loss from both of the surfaces, i.e., maximum erosion rate at an angle of impingement $\approx 90^{\circ}$ for cast iron (crack formation) and $\approx 20^{\circ}$ for aluminum.

Recent studies also examine the corrosion effects of nanofluids in the no-flow case. The corrosion resistance of brass owing to the presence of a sodium dodecyl benzene sulfonate (SDBS) surfactant is further increased by Al_2O_3 -water nanofluid, which improves the protective film occurring via adsorption on the brass [108,109]. Nevertheless, no enhanced brass resistance to TiO₂-water nanofluid is acquired with SDBS addition due to the lack of protective film [110]. In a similar way, Al_2O_3 -water nanofluid contributes to corrosion by weakening the SDBS protective layer on aluminum alloy 6061 [111].

The effect of CNTs–water nanofluid (with gum arabic (GA) as a surfactant) on the corrosion rate of 316L stainless steel, plunged for 10 h, is evaluated in terms of particle concentrations at 0.05, 0.1, 0.3, and 0.5 wt.% [112]. With the presence of CNTs, corrosion resistance is enhanced compared to GA–water solutions owing to the adsorption of CNTs on the substrate and forming a protective layer acting as a barrier towards the solution ions that attack the substrate. The lowest corrosion rate is attained at 0.1 wt.%, while the higher concentrations stimulate corrosion due to the uneven and non-uniform distribution of CNTs on the surface. In another study [113], the same research group investigates the effect of temperature at 22, 40, 60, and 80°C on the same phenomenon using 0.1 wt.% and 1 wt.% CNTs–GA–water nanofluids. Although CNTs behave as corrosion inhibitors for 316 L stainless steel, increasing temperatures decrease this advantage. The highest corrosion rate is observed both at the largest temperature (as CNTs can desorb at greater temperatures) and particle concentration due to the uneven accumulation of CNTs. By combining the information from these two articles, we can also interpret that the corrosion rate is largely affected by the temperature than the particle concentration.

Since erosion–corrosion effects originate from the interactions between the mechanical processes and the chemical reactions during a fluid flow [114], the material loss can become 25–100X larger than the uniform corrosion rate [115]. In addition, the coupled damage caused by these interactions is known as synergism or additive effects due to the larger amount of material loss compared to the summed loss from the pure mechanical erosion and pure chemical corrosion.

The work of Celata et al. [116] in 2014 is the first thorough study on the erosioncorrosion sensitivity of different nanofluid flows for a long term (weeks) at high particle concentrations (\geq 3 wt.%) on various metal surfaces, as detailed in Table 2. They compare the degree of consumption on the target metals by the nanofluids circulating in an industrial scale setup. It is remarked that stainless steel is insensitive to all tested types of nanofluids (no consumption). At the same time, aluminum substrates are the most sensitive and show a severe amount of consumption, and the results of copper are dependent on the type of nanofluid. They also demonstrate some other negative outcomes, i.e., wear on the pump gear and consequent impact on the gear rotational speed (rpm), resulting in an unstable rpm, as given in Figure 11.

			Water Tests		Nanofluid Tests		
Nanofluid	pН	Vol. [m ³]	Α1 [μm]	Cu [µm]	Α1 [μm]	Cu [µm]	Duration + Flow Rate
Al ₂ O ₃ (9 wt.%)	8.8	7.363	1.56	0.25	182.6	1.17	10 days @ 6 m/s + 5 days @ 3.3 m/s
Al ₂ O ₃ (3 wt.%)	8.6	9.6	0.45	0.74	263	0.88	16 days @ 5.3 m/s
TiO ₂ (9 wt.%)	7.3	7.7	0.87	-	1.13	-	13 days @ 5 m/s
TiO ₂ (9 wt.%)	7.3	12.2	-	0.36	-	-0.54	17 days @ 6 m/s + 20 days @ 0.7 m/s
ZrO ₂ (9 wt.%)	9	10.1	0.15	0.30	95.5	4.4	17 days @ 5.2 m/s
ZrO ₂ (base fluid)	7.4	9.323	0.73	0.88	0.55	0.53	13 days @ 6.3 m/s
SiC (3 wt.%)	5.9	6.1	0.8	1.09	0.67	0.68	9 days @ 6 m/s

Table 2. Resultant effects of abrasion, erosion, and corrosion on different metal targets in terms of a decrease in the material thickness, i.e., step height in μ m, due to the fluid flow [116,117].



Figure 11. (a) Abrasion on the gears in PTFE+carbon fiber. (b) Consequent change in the pump rpm (the ratio with regard to the base fluid conditions) due to gear wear with total circulated volume of different nanofluids. Reprinted from Ref. [116], Copyright 2014 Elsevier.

To further elaborate the abrasion, erosion, and corrosion effects, Bubbico et al. [117] evaluate the impact of a nanofluid's pH. In particular, the severe damages on the aluminum substrate happen when the pH of the fluid does not fall within the passivation zone on the Pourbaix diagram [118] of aluminum. In this zone, the aluminum substrate is covered with a protective oxide layer, i.e., Al₂O₃, which obstructs further oxidation, namely, corrosion. However, when the pH is out of the passivation zone, corrosion starts to take place, and this explains the high aluminum consumption due to Al₂O₃-water nanofluids (both at 3 wt.% and 9 wt.%) and ZrO_2 -water nanofluid. To prove that corrosion is the dominant effect on aluminum substrate, the pH of Al_2O_3 -water nanofluid (9 wt.%) is modified by keeping the other parameters constant. Small amounts of citric and malic acids are separately added so that the pH of two nanofluids lies in the passivation zone. The tests point out remarkable reductions in aluminum consumption for the two cases, but the damages remain severe. Additional pH measurements at the end of the experiments reveal that the reduced pHs are increased to the values in the corrosion zone, probably due to some side reactions between the additives promoted by the wall temperatures reaching 70°C. Two final experiments are conducted with Al₂O₃-water nanofluids (9 wt.% and 20 wt.%), containing some additives to stabilize the pH in the passivation zone of aluminum throughout the experimentation. These tests yield zero to a negligible amount of aluminum consumption, highlighting that the severe consumption is solely due to corrosion and not erosion, surprisingly at 20 wt.%. Consequently, it can be said that nanofluids flowing in the fluidic loop would cause zero to a negligible amount of damages on a range of traditional materials if the conditions causing corrosion are properly prevented.

To summarize, the following features play en essential role in the initiation and intensity of abrasion, erosion, and corrosion phenomena:

- Solid particle characteristics (clustered particles' shape and size, particle material, e.g., hardness);
- Particle flow characteristics (flow rate, pH, particle concentration and dispersity level, Stoke's number, impact speed, impact angle, temperature, turbulence);
- Wall characteristics (surface roughness, geometry, wall material, e.g., hardness);
- Particle–particle interactions, e.g., shielding effect.

9. Conclusions and Perspectives

This paper aims to review the published experimental works available in the literature that address single-phase nanofluids as coolants inside microchannels. In addition, the chemical nature of the nanofluids is discussed in terms of particle stability, clustering, and sedimentation, as well as their tribological effects, i.e., abrasion, erosion, and corrosion. The following factors could be the cause of the contradictory results reported in the literature even for the experiments performed under the supposedly same or similar experimental conditions:

- ♦ Heat transfer experiments running with nanofluids for different durations;
- ◊ Difficulty in establishing identical experimental conditions for the base fluid and the nanofluid;
- ◊ Erosion of clustered nanoparticles on metallic surfaces texturing the surfaces;
- Erosion of clustered nanoparticles on metallic surfaces detaching oxide grains;
- Corrosion effect of water (either as the base fluid or as the water-based nanofluid) on some metals, such as copper;
- ◊ Surface oxidation occurring in time.

The key findings and some future perspectives are listed below:

- Several factors influence the nanofluid thermal performance in MCHSs: the particle size distribution and particle concentration, particle–base fluid combination, the surfactant amount, the channel material, geometry and size, and the flow rate.
- Many studies present favored thermal behavior with the use of nanofluids in microchannels compared to the base fluids, especially at low particle concentrations. Plus, some marginal increase in the pumping power is observed due to the presence of the nanoparticles. Al₂O₃-water nanofluid is the most investigated type owing to its low cost, good stability, and thermal performance despite its strong hardness properties.
- The thermophoresis and the thermal boundary layer distortion are considered the primary underlying phenomena to explain the heat transfer improvement with nanofluids.
- There may be an optimal particle concentration for the enhanced cooling mechanism with nanofluids, above which particles tend to cluster and sediment, resulting in worsened thermal performance.
- Despite a vast number of published research articles, nanofluids have not been applied yet in practice for electronic cooling due to many factors, such as nanofluids' long-term stability issues, production, and maintenance costs. The scattering of the experimental results, observed even under similar experimental conditions, is the fundamental reason that triggers this the gap. The reported controversial and inconsistent experimental results may originate from nanoparticle clustering, which is almost inevitable due to van der Waals forces and other colloidal interactions affecting the nanofluid stability. To resolve these conflicts, there is a need for an in situ optical measurement technique that can be implemented to evaluate the effect of the presence of the nanoparticle clusters. A possibility is to use the Light Extinction Spectroscopy technique [119] to monitor the nanofluid stability.
- The immediate substitution of traditional coolants with nanofluids in cooling systems is certainly not straightforward. Their design would require elaborate mechanical adaptations to obtain the required heat transfer enhancement with nanofluids. Despite the high resistance of stainless steel to erosion and corrosion, current studies are unfortunately insufficient to draw conclusions, and there is no vision about nanofluids'

long-term effects. For example, the abrasion, erosion, and corrosion phenomena due to the clustered nanoparticles are not profoundly examined in the literature. They can not only alter the microchannel's walls and provoke clogging, but they might also influence the thermal performance. In short, to assess its negative interactions with various materials involved in the fluidic loop, preliminary testing of the selected nanofluid is a must before their commercialization in any kind of application.

Author Contributions: Conceptualization, P.E.; investigation, P.E. and Y.T.A.; writing—original draft preparation, P.E., Y.T.A. and M.R.V.; writing—review and editing, P.E., Y.T.A. and M.R.V., supervision, M.R.V.; funding acquisition, M.R.V. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by Interne Fondsen KU Leuven/Internal Funds KU Leuven (C24/18/057).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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